

## Response to the reviewer comments on the manuscript

### C-IFS-CB05-BASCOE: Stratospheric Chemistry in the Integrated Forecasting System of ECMWF

By V. Huijnen et al.

First, we would like to thank the reviewers for their critical, but useful comments. In view of their valuable suggestions in our revised manuscript we have:

- 1 included an additional model configuration containing full (tropospheric *and* stratospheric) chemistry within the whole atmosphere
- 2 revised our PSC-parameterization
- 3 extended our model evaluation with one additional year
- 4 revised some of our evaluations

The reviewer's comments are given in italic, and our responses in regular font. Textual modifications to the manuscript are highlighted in bold. Figure numbers refer to the revised manuscript.

#### **Response to anonymous Referee #1**

*The paper describes an update of the C-IFS model, including a stratospheric chemistry scheme in addition to the existing tropospheric scheme, and using the tropopause to switch between the schemes. This method is also used by other models, and the authors consider the possible inconsistency at the transition zone between the domains. The paper is fairly short and concise, although there are a few issues that should be improved or made clearer.*

*I therefore think minor revisions are necessary, although my questions on the relatively short simulation period may require more effort than the other comments.*

#### **Abstract**

*The abstract should contain some more on the motivation for including stratospheric chemistry in CAMS. Is forecasting skill part of the motivation?*

*It is stated that this is a first step, which makes me wonder how far away the next step really is.*

The essential motivation for including stratospheric chemistry within C-IFS is to enable the evaluation of ozone and methane oxidation chemistry processes in the stratosphere, to achieve an improved representation of ozone, water vapour and related trace gases within IFS. Combined with the availability of long-lived trace gases such as N<sub>2</sub>O, evaluation of such an atmospheric composition analysis and forecast system is expected on the long term to lead to improvements to IFS meteorological forecasts as well.

The reviewer is correct that the wording '*first step*' potentially implies to the reader that several major next steps are still needed before operationalization. However, in our conclusions we list the steps that are still foreseen, of which none of them prevents such operationalization. Also an extension of the simulation from 1.5 year to 2.5 year (see also below) showed that the stratospheric trace gas composition remains bounded, with no obvious drift. Even though we acknowledge the system is not yet perfect overall a clear improvement is obtained compared to the current system. Therefore, to

clarify the status of this work, we now judge this as the ‘key’ step towards operationalization, where essentially the next steps concern the acceptance of ECMWF of the model performance in operational conditions as well as the coding implementation. Depending on the CAMS schedule, final implementation and testing in the operational environment, including a new meteorological cycle, would be required. We now write in our abstract:

“This marks a **key** step towards a chemistry module within IFS that encompasses both tropospheric and stratospheric composition, **and could expand the CAMS analysis and forecast capabilities in the near future.**”

## 1. Introduction

*Page 1, Line 25: “Also analyses and forecasts of stratospheric ozone directly impact the forecast capabilities of surface solar irradiance” is a bit difficult to understand. Suggest changing to e.g. “Also, the amount of stratospheric ozone directly impact the forecast capabilities of surface solar irradiance, making good stratospheric ozone forecast important”.*

We thank the reviewer for this suggestion. We replaced this sentence into:

**“Also, the amount of stratospheric ozone directly impacts the forecast capabilities of surface solar irradiance (Qu et al., 2014), stressing the relevance of good stratospheric ozone forecasts.”**

*Page 3, Line 5: “We have developed a strategy”: Perhaps better to say “We present here a merging ...”. There is no need to state that there is a strategy, and the work done is the actual merging.*

We agree with the reviewer not to emphasize our strategy, but simply the approach we have taken in our the work. In this section we now write:

**“We have developed an approach for an optimized** merging the CB05 tropospheric chemistry scheme...”

While in the next section we write:

**“In this paper we describe two merging approaches...”**

## 2.1 Stratospheric chemistry

*Page 4, Line 15: Are the surface area densities (SAD) fixed? No size distribution?*

The surface area density (SAD) field for stratospheric aerosol is constructed from a zonal mean aerosol number density field assuming a constant lognormal size distribution with median radius of 0.07  $\mu\text{m}$  and geometric standard deviation of 1.76. The aerosol number density is taken from SAGE II extinction measurements (Hitchman et al 1994). This is different from the ‘Daerden et al, ACP 2007’ reference as stated in the manuscript. We apologize for this mis-representation. For ice and NAT PSCs fixed SADs are assumed as reported in the manuscript. We now write:

**“The surface area density of stratospheric aerosols uses an aerosol number density climatology based on SAGE-II observations (Hitchman et al., 1994).”**

*On the fact that you do not do sedimentation, but parameterise it using exponential decay of HNO<sub>3</sub> and H<sub>2</sub>O, is this a good approximation? If you have a situation of very cold temperatures over a long period, the PSCs may sediment out and the SAD will be reduced. Will the parameterisation cause too large denitrification/dehydration? Figure 9 could perhaps indicate this?*

Indeed Figures 6 and 9 in the original manuscript show lower HNO<sub>3</sub> and H<sub>2</sub>O in BASCOE-CTM and CIFS-TS than the MLS observations over the south pole, indicating a too efficient removal of HNO<sub>3</sub> and H<sub>2</sub>O through sedimentation as consequence of the simplistic parameterization based on a temperature criterion. We have worked on an improvement of the PSC scheme, see also the response to reviewer #2.

### **2.3 Merging procedure ...**

*Page 5, Line 24: 40 hPa as tropopause is very low. Does this occur often? To my knowledge, the tropopause pressure is seldom lower than 80hPa. Using 40 hPa may be of small consequence, and may even cause O<sub>3</sub> production to be better represented.*

In practice the O<sub>3</sub> and CO concentrations always define the interface between tropospheric and stratospheric chemistry, rather than the 40hPa level. This pressure criterion is only introduced to prevent any spurious detection of tropospheric conditions at the top of the atmosphere where CO (O<sub>3</sub>) increases (decreases), due to CO<sub>2</sub> and O<sub>3</sub> photolysis. We specify this now more clearly by adding the sentence:

**“With this definition the associated tropopause pressure ranges in practice between approx. 270 and 80 hPa for sub-tropics and tropics, respectively.”**

*Page 5, Line 26: “Specificaly” -> “Specifically”.*

We changed this, thank you.

*Page 5, Line 34: 10-day decay rate in the stratosphere. What happens to the lost species? Are they assumed to be converted to aerosols?*

The trace gases affected by this decay rate are currently lost, i.e. they do not contribute to the aerosol, CO or NO<sub>y</sub> components. Clearly, especially a coupling of tropospheric aerosol to the stratospheric ozone chemistry would be an interesting application, as we also mention in our conclusions section, but this is beyond the scope of the current system. We add a sentence for clarification:

**“These losses are currently not accounted for in the stratospheric chemical mechanisms and do not contribute either to the load of stratospheric aerosols.”**

*Page 6, Line 11-14: It would be very interesting to see how chemical composition changes when compared to this test. I think some more info should be given on this, as it could explain chemically why you use two domains.*

Also in response to reviewer #2 we now include an evaluation of this model setup where tropospheric and stratospheric chemistry schemes have been fully merged into one single reaction mechanism, which we refer to as C-IFS-Atmos. In addition we now show model profiles near the tropopause for a selection of components in a new Supplementary Material. Even though we see significant differences in the troposphere for short-lived chlorine and bromine-containing trace gases, the differences between C-IFS-Atmos and C-IFS-TS in the stratospheric composition remain small, as confirmed by the extended model evaluation including C-IFS-Atmos. This can be understood since  $\text{N}_2\text{O}$ , methyl chloride, methyl bromide and CFC's, which form the largest sources of  $\text{NO}_y$ , HCl and HBr reservoir trace gases in the stratosphere, are marginally affected by these tropospheric reactions. This aspect is also discussed at the start of Sec. 2.3.

Near the tropopause level, at the interface between tropospheric and stratospheric chemistry in C-IFS-TS, some vertical oscillations are visible in C-IFS-TS for  $\text{CH}_2\text{O}$  (Fig. S2), HBr and Br (Fig. S5), i.e. species with relatively short lifetimes which have a different chemistry in the tropospheric and stratospheric modules ( $\text{CH}_2\text{O}$ ) or no chemistry in the tropospheric module (HBr, Br). The limited difference between C-IFS-TS and C-IFS-Atmos for other short-lived components ( $\text{NO}_y$ , OH, chlorine-containing trace gases, as well as the long-lived trace gases such as  $\text{O}_3$ , CO,  $\text{N}_2\text{O}$ , CFC's and  $\text{CH}_4$ ) supports the assumption that the chemical split between troposphere and stratosphere is appropriate for key components.

Also the  $\text{CH}_3\text{Cl}$  (Fig. S4) and  $\text{CH}_3\text{Br}$  (Fig. S5) components do show a limited discrepancy between C-IFS-TS and C-IFS-Atmos in the troposphere, associated with the absence of chemical break-down in the tropospheric part in C-IFS-TS. We acknowledge that an extension of the tropospheric reaction mechanism with halogen species also impacts tropospheric ozone and, e.g., the methane lifetime. We found that differences in tropospheric OH,  $\text{O}_3$  and related components are generally small, while a closer inspection is beyond the scope of this work.

### **2.3.1 Merging photolysis rates**

*Page 6, Line 26-27: How is the interpolation/merging done? Some weighting for different layers?*

The photolysis rates are linearly interpolated with pressure between the BASCOE parameterization in the stratosphere and MBA parameterization in the troposphere, at four pressure levels around a fixed tropopause altitude. This altitude is somewhat different (a bit lower towards the troposphere) than the chemical tropopause level adopted for the selection of the solver.

*Page 7, Line 1: The merging only affects 4 layers, where some are in the troposphere, so I am not surprised the over-all impact is small. Perhaps it could be noted that  $\text{JO}_3$  does not seem to be used in troposphere? Did you check hourly composition at these grid boxes?*

Please note that the photolytic reaction  $\text{JO}_3 \rightarrow \text{O}^1\text{D}$  is clearly also active in the troposphere, see also Table 4, even though the  $\text{O}^1\text{D}$  reaction product in the CB05 tropospheric chemistry scheme is only implicitly accounted for (see also response to ref#2). We now present instantaneous model profiles near the tropopause in the Supplementary Material, where we also include results from the C-IFS-Atmos version. We now add in the manuscript:

“Even though such jumps are undesirable, no visible impact on local chemical composition was found, **for any of the trace gases involved in both tropospheric and stratospheric chemistry, see also Figures S1-S3 in the Supplementary Material. This can be explained by the sufficiently small difference in the photolysis rates at the merging altitude of the photolysis and chemistry schemes, combined with the sufficiently long lifetime of the affected trace gases.**”

### **2.3.2 Merging tracer transport**

*This is perhaps not really a merging.*

The reviewer is correct. We have changed the subsection title to **“Tracer transport settings”**.

*Regarding stratospheric H<sub>2</sub>O tracer: So you do not use tropospheric H<sub>2</sub>O (from q) as source of stratospheric H<sub>2</sub>O? I would expect some boundary condition is needed, at least in the tropics. How is this treated?*

In fact we do use tropospheric H<sub>2</sub>O, as constrained by q, to serve as boundary condition for the stratosphere. We clarify this better by changing the sentences as follows:

**“While a chemical H<sub>2</sub>O tracer is defined in the full atmosphere, in the troposphere H<sub>2</sub>O mass mixing ratios are constrained by the humidity (q) simulated in the meteorological model in the IFS and provide a boundary condition for water vapour in the stratosphere.”**

### **3.1 Observational data ...**

*Page 8, Line 21: “weighted part”: What kind of weight?*

The weighted contribution of tropospheric NO<sub>2</sub> to the stratospheric NO<sub>2</sub> columns are due to the applied air mass factor. We slightly revised this section and now write more explicitly:

**“Stratospheric NO<sub>2</sub> columns from SCIAMACHY presented here are in fact total columns derived by dividing retrieved slant columns of NO<sub>2</sub> by a stratospheric air mass factor and contains data over the clean Pacific ocean (180°E - 220°E) only (Richter et al., 2005). Although in this region the contribution of the troposphere to total column NO<sub>2</sub> is small, stratospheric column NO<sub>2</sub> from SCIAMACHY is still somewhat positively biased by a tropospheric contribution. However, stratospheric air mass factors for NO<sub>2</sub> are usually large compared to tropospheric ones, so that the uncertainty resulting from this should only have a minor impact on the data analysis presented in this study.”**

*Page 8, Line 23: What is the model output frequency? It would be helpful to specify this and whether you use instant model values.*

In all cases, including the NO<sub>2</sub> stratospheric column evaluation, three-hourly instantaneous model values are used. In the case of SCIAMACHY NO<sub>2</sub> observations these model values are interpolated in time to 10:00 LT. We now specify this at the end of Sec. 3.1:

**“Three-hourly C-IFS and BASCOE-CTM output has been interpolated in space and time to match with any of these observations.”**

#### **4 Model evaluation**

*Page 9, Line 14-20: The equatorial low bias and high bias at NH mid-lat could indicate too fast transport away from Equator?*

Indeed potential biases in transport would impact on stratospheric composition. This issue is now discussed in more detail in the revised manuscript (see reply to last comment of second reviewer). We believe the stratospheric ozone biases are also associated to biases in NO<sub>x</sub>, which tend to be positive over the NH mid-latitude, while neutral and/or slightly negative over the tropics from 2009 onwards (despite the corresponding positive bias in N<sub>2</sub>O). We add the following sentence:

**“The alternating biases in CIFS-TS and CIFS-Atmos are due to corresponding biases in chemically related species such as NO<sub>x</sub> and also to transport issues, as discussed in more detail below.”**

*Figure 2: The simulated time period is not long enough to make clear whether the bias will build up. This should be further explained.*

The reviewer is correct that a 1.5 year simulation is relatively short to assess the evolution of stratospheric composition, and the corresponding development of biases. For that, multi-year simulations are required. We have extended all model runs with one extra year (2010) and expanded the corresponding evaluations. This revealed that the biases in O<sub>3</sub> remained essentially the same in 2010 compared to 2009. We now include figures showing time series of O<sub>3</sub> partial columns and total columns, as well as NO<sub>2</sub> total columns over the full period. However, please note that the purpose of this system is to provide accurate analyses and short-term forecasts of atmospheric composition, rather than good multiyear simulations, as also acknowledged by the reviewer.

*Ozone hole: Do you overestimate PSCs (as commented earlier) and hence halogen activation?*

We revised our PSC-parameterization (see also Ref#2), which indeed led to an improvement in halogen activation. We now expand our assessment of O<sub>3</sub>, HNO<sub>3</sub> and ClO evolution during 2009 Austral spring. In the revised setup the modeled ClO follows very well the observations, although its decrease is about two weeks earlier than observed, as now documented in a new Figure (Fig. 6 in the revised manuscript). Still the O<sub>3</sub> appears under-estimated to a larger amount than BASCOE-CTM, suggesting that transport-related elements also contribute to biases in O<sub>3</sub>.

*Page 9, Line 20-32: Is the comparison done by extracting the same profiles (location and time) as in sondes? Should be specified.*

For our methodology for comparing the model ozone with the sondes we use three-hourly model output which has been interpolated in time and space to match with the observations. We now specify this in the manuscript, as discussed above.

*The sonde comparison should include some variability in sondes (e.g. as standard deviation), and possibly also from the model (Fig.3-5). (In Figure 3-4 this could be placed in either upper or lower row.)*

We now present the standard deviation of the variability in the O<sub>3</sub> profiles, both for the observations and the collocated model results. Also, in response to reviewer #2 Figures 3 and 4 now present the O<sub>3</sub> evaluation in terms of ppmv rather than mPa. We have adapted the figure legends accordingly.

*Figure 6: How is OH in IFS vs BASCOE? Do you have less HNO<sub>3</sub> in stratosphere because of hotter photochemistry? Some thoughts/explanation should be given for the higher low-SZA NO<sub>2</sub> in IFS than in BASCOE. NO<sub>2</sub>+NO<sub>3</sub> ↔ N<sub>2</sub>O<sub>5</sub> at night? Photochemistry?*

The OH between C-IFS-TS and BASCOE-CTM is essentially identical, as largely governed by O<sup>1</sup>D reaction with H<sub>2</sub>O, and in turn on O<sub>3</sub> abundance and photolysis. This indeed does not give rise to the differences in HNO<sub>3</sub> between C-IFS-TS and BASCOE-CTM, which occur mainly at an altitude of approx. 10-30 hPa. Considering the equal photochemistry in the stratosphere the reduced HNO<sub>3</sub> in C-IFS-TS compared to BASCOE-CTM the discrepancy points at transport-related features, as also discussed with Figure 9. Note that the (reasonable) daytime NO<sub>2</sub> maximum, which is found at an altitude of ~10 hPa, is not directly related to this aspect, nor to the anomalously higher NO<sub>2</sub> during night-time (i.e. at high SZA) in C-IFS-TS compared to BASCOE-CTM and observations. This positive offset is largely occurring in the 5-1 hPa altitude range, but remains limited to biases in daytime NO and nighttime NO<sub>2</sub> only, while other trace gases in this altitude range (N<sub>2</sub>O, NO<sub>3</sub>, N<sub>2</sub>O<sub>5</sub>, HNO<sub>3</sub>) do not explain this bias. The following sentence has been added at the end of the paragraph:

**Even though a clear improvement compared to run C-IFS-T is found, further investigation is necessary to diagnose the origins of the biases in night-time NO<sub>2</sub> above 10 hPa and in HNO<sub>3</sub> between 10 and 70 hPa.**

*Page 10, Line 3-4 (Figure 7): NH higher NO<sub>2</sub>: Could a possible reason be that SCIAMACHY assumes that a too high fraction of the column is located in the troposphere?*

The fact that model columns are generally larger than retrieved ones cannot be explained by the contribution of the troposphere to stratospheric columns from SCIAMACHY alone, as stratospheric columns from SCIAMACHY are positively biased by a tropospheric contribution to a minor extent only (see our earlier response above). Moreover, the overestimation compared to SCIAMACHY retrievals cannot be explained by error estimates for SCIAMACHY stratospheric NO<sub>2</sub> columns given in the manuscript (relative uncertainties of roughly 5-10% and additional absolute uncertainty of  $1 \times 10^{14}$  molec cm<sup>-2</sup>) alone, which only account for about  $0.3 \times 10^{15}$  molec cm<sup>-2</sup> of the positive bias compared to SCIAMACHY. Hence the slight positive bias suggests a C-IFS model issue, as is also confirmed with the NO<sub>2</sub> evaluation against MIPAS observations in Figure 6, where the model also shows larger values than the observations over the NH, around the 8hPa altitude.

*Page 10, Line 19: What about too fast horizontal transport? Is CH<sub>4</sub> fixed at surface?*

We constrain CH<sub>4</sub> at the surface using a monthly and latitudinally varying climatology, which is somewhat different than the null flux approach adopted in BASCOE-CTM. Nevertheless, the agreement with BASCOE-CTM (and observations) at ~ 100hPa does not suggest issues with the tropospheric CH<sub>4</sub> concentrations that could explain the discrepancies seen in the stratosphere. Indeed not only vertical transport, but also horizontal transport and mixing could be causes for differences between C-IFS and BASCOE-CTM in the 100-10 hPa altitude range. Thanks to the last comment by second reviewer, the discussion of the transport issue indicated by fig.9 has been completely re-written in order to avoid any over-interpretation:

**“Fig. 9 shows an evaluation of N<sub>2</sub>O and CH<sub>4</sub> profiles during September 2009 against observations by ACE-FTS. Owing to their long lifetimes these trace gases are good markers for the model ability to describe transport processes - i.e. not only the Brewer-Dobson circulation but also isentropic mixing, mixing barriers, descent in the polar vortex, and stratosphere-troposphere exchange (Shepherd, 2007). Moreover, N<sub>2</sub>O is the main source of reactive nitrogen in the stratosphere while CH<sub>4</sub> is one of the main precursors for stratospheric water vapour. The figure suggests reasonable profile shapes for both CH<sub>4</sub> and N<sub>2</sub>O in the upper stratosphere (10 hPa and higher) where their abundance is more strongly influenced by chemical loss but at lower altitudes (100-10 hPa) C-IFS-TS and C-IFS-Atmos show larger discrepancies to the observations than the BASCOE-CTM run, with weaker vertical gradients in the tropics and SH-mid latitudes and a sharper gradient in the extra-tropical Northern Hemisphere.**

**This discrepancy cannot be due to different wind fields because the BASCOE CTM experiment is driven by three-hourly output of the C-IFS-T experiment. We attribute it instead to the different numerical schemes for advection and/or to differences in the representation of sub-grid transport processes in the GCM and in the CTM. Convection and diffusion are indeed explicitly modelled in C-IFS but neglected in BASCOE CTM, which relies on the implicit diffusion properties of its flux-form advection scheme to represent sub-grid mixing (Lin and Rood, 1996; Jablonowski and Williamson, 2011).. Since lower stratospheric ozone is strongly determined by both chemistry and transport, the transport issue indicated by Fig. 9 could also contribute directly to the ozone biases seen below 10 hPa in Figures 3 and 4.”**

## **5 Conclusions**

*Page 11, Line 3: I generally do not think 1.5 years is enough for evaluating the chemistry and chemistry. A possible drift in the O<sub>3</sub> column (Fig. 2) should be investigated. If chemistry is adjusted in an assimilation system, a drift will probably not be very prominent or important, but used as a CTM 1.5 years is short.*

We sympathize with the comment of the reviewer that a 1.5 year simulation is too short to assess and quantify potential drifts. Therefore we have now extended the runs and corresponding evaluation with an additional year (2010). This indeed shows that biases in O<sub>3</sub> partial and total columns for 2010 are



essentially similar to 2009, and also the Antarctic O<sub>3</sub> hole period was modeled with similar skills. We extended the discussion in the full manuscript on this additional year.

*Page 11, Line 9-11: “a larger error” -> “larger errors”, “was” -> “were”, and fix rest of sentence.*

We changed this, thank you for the suggestion.

*Page 11, Line 12: Why is it necessary to do this first step? It seems another step is expected.*

See also our comments above. We now modify this as follows:

“This benchmark model evaluation of C-IFS-TS marks a **key** step towards merging tropospheric and stratospheric chemistry within IFS, aiming at a **possible configuration for** daily operational forecasts of lower and middle atmospheric composition **in the near future**.”

*Page 11, Line 18: What use would assimilation of long-lived species have in the IFS?*

Assimilation of long-lived species ensures the provision of observationally constrained trace gas fields which will intrinsically contribute to an improved quality of ozone chemistry specifically, and atmospheric composition forecasts in general, which is one of the essential purposes of the system.

*Page 12, Line 6: While monitoring capabilities may be important and interesting in the stratosphere, and also provide global products of species which are not globally observed, it would be interesting to hear the implications for forecasting.*

*Is it not only O<sub>3</sub> that is important for radiation calculations? Better stratospheric O<sub>3</sub> could improve stratospheric temperatures?*

Indeed better stratospheric O<sub>3</sub> is one of the key target products which are expected to lead to improved stratospheric temperature fields, as shown e.g. by de Grandpré et al. (2009). Also stratospheric water vapour which is also highly relevant to radiation, can potentially benefit from an improved representation of stratospheric chemistry. Note that improvement in description of these tracer fields can either be obtained through revised climatologies, as derived from stratospheric composition analyses, as well as through revised parameterization of prognostic variables.

We explicitly hint on these aspects in the introduction of our manuscript, as one of the motivations to develop this system. As for the conclusion, we believe the general statement ‘(...) stratospheric chemistry (...) may also contribute to advances in meteorological forecasting of the ECMWF IFS model in the future’ is appropriate, as at this stage it is too early to specify such applications.

## **Appendix**

*Table A1 should be sorted on names.*

The reviewer is correct that the ordering was not optimal. We have re-ordered the list of trace gases, first grouping trace gases active in the various regions (glb, trop, strat), and next sorting them more strictly on functional groups (e.g. grouping the hydrocarbons, the chlorine-containing trace gases, etc.)

### ***References***

*DOIs are missing for some references; please update.*

We have updated the list of references, including the missing DOIs. The additional references in the revised manuscript are listed after the reply to the second reviewer.