

Responses to the reviewer comments on the manuscript

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

5 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:

- 10 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

15 The two reviewer's comments are given in italic, and our responses in regular font; this text is identical to the reviewer responses given before. Textual modifications to the manuscript are highlighted in bold. Figure numbers refer to the revised manuscript. To the end of this document a marked-up version of the revised manuscript with the changes to the original GMDD manuscript is provided.

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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.

30 **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.

35 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 N₂O, 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
5 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
10 '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:

15 "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

20 *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:

25 "**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.**"

30 *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:

35 "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?*

5

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 μm 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:

10

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

15

On the fact that you do not do sedimentation, but parameterise it using exponential decay of HNO₃ and H₂O, 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?

20

Indeed Figures 6 and 9 in the original manuscript show lower HNO₃ and H₂O in BASCOE-CTM and CIFS-TS than the MLS observations over the south pole, indicating a too efficient removal of HNO₃ and H₂O 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.

25

30 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 O₃ production to be better represented.*

35

In practice the O₃ 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 (O₃) increases (decreases), due to CO₂ and O₃ 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.”

5

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

We changed this, thank you.

10

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_y 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.”

25

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 N₂O, methyl chloride, methyl bromide and CFC's, which form the largest sources of 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 CH₂O (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 (CH₂O) 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 (NO_y , OH, chlorine-containing trace gases, as well as the long-lived trace gases such as O_3 , CO, N_2O , CFC's and CH_4) supports the assumption that the chemical split between troposphere and stratosphere is appropriate for key components.

Also the CH_3Cl (Fig. S4) and CH_3Br (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, O_3 and related components are generally small, while a closer inspection is beyond the scope of this work.

10

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.

20 *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 JO3 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 O^1D 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:

30 **“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.”**

35

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**”.

5

Regarding stratospheric H₂O tracer: So you do not use tropospheric H₂O (from q) as source of stratospheric H₂O? I would expect some boundary condition is needed, at least in the tropics. How is this treated?

In fact we do use tropospheric H₂O, as constrained by q, to serve as boundary condition for the stratosphere.

10 We clarify this better by changing the sentences as follows:

“While a chemical H₂O tracer is defined in the full atmosphere, in the troposphere H₂O 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.**”

15

3.1 Observational data ...

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

20 The weighted contribution of tropospheric NO₂ to the stratospheric NO₂ columns are due to the applied air mass factor. We slightly revised this section and now write more explicitly:

25 “Stratospheric NO₂ columns from SCIAMACHY presented here are in fact total columns **derived by dividing retrieved slant columns of NO₂ 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 NO₂ is small, stratospheric column NO₂ from SCIAMACHY is still somewhat positively biased by a tropospheric contribution. However, stratospheric air mass factors for NO₂ 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.**”

30

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

35 In all cases, including the NO₂ stratospheric column evaluation, three-hourly instantaneous model values are used. In the case of SCIAMACHY NO₂ 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.”

5 **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_x, 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 N₂O). We add the following sentence:

15 **“The alternating biases in CIFS-TS and CIFS-Atmos are due to corresponding biases in chemically related species such as NO_x 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 O₃ remained essentially the same in 2010 compared to 2009. We now include figures showing time series of O₃ partial columns and total columns, as well as NO₂ 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.

30

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 O₃, HNO₃ 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 O₃ appears under-estimated to a larger amount than BASCOE-CTM, suggesting that transport-related elements also contribute to biases in O₃.

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

5

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.

10

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 O₃ profiles, both for the observations and the collocated model results. Also, in response to reviewer #2 Figures 3 and 4 now present the O₃ evaluation in terms of ppmv rather than mPa. We have adapted the figure legends accordingly.

15

Figure 6: How is OH in IFS vs BASCOE? Do you have less HNO₃ in stratosphere because of hotter photochemistry? Some thoughts/explanation should be given for the higher low-SZA NO₂ in IFS than in BASCOE. NO₂+NO₃ <-> N₂O₅ at night? Photochemistry?

20

The OH between C-IFS-TS and BASCOE-CTM is essentially identical, as largely governed by O¹D reaction with H₂O, and in turn on O₃ abundance and photolysis. This indeed does not give rise to the differences in HNO₃ 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 HNO₃ in C-IFS-TS compared to BASCOE-CTM the discrepancy points at transport-related features, as also discussed with Figure 9.

25

Note that the (reasonable) daytime NO₂ maximum, which is found at an altitude of ~10 hPa, is not directly related to this aspect, nor to the anomalously higher NO₂ 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 NO₂ only, while other trace gases in this altitude range (N₂O, NO₃, N₂O₅, HNO₃) do not explain this bias. The following sentence has been added at the end of the paragraph:

30

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 NO₂ above 10 hPa and in HNO₃ between 10 and 70 hPa.

35

Page 10, Line 3-4 (Figure 7): NH higher NO₂: Could a possible reason be that SCIAMACHY assumes that a too high fraction of the column is located in the troposphere?

5 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 NO₂ columns given in the manuscript (relative uncertainties of roughly 5-10% and additional absolute uncertainty of 1×10^{14} molec cm⁻²) alone, which only account for about 0.3×10^{15} molec cm⁻² of the positive bias compared to SCIAMACHY. Hence the slight positive bias suggests a C-IFS model issue, as is also confirmed with the NO₂ evaluation against MIPAS observations in Figure 6, where the model also shows larger values than the observations over the NH, around the 8hPa altitude.

15 Page 10, Line 19: What about too fast horizontal transport? Is CH₄ fixed at surface?

We constrain CH₄ 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 CH₄ 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:

25 **“Fig. 9 shows an evaluation of N₂O and CH₄ 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, N₂O is the main source of reactive nitrogen in the stratosphere while CH₄ is one of the main precursors for stratospheric water vapour. The figure suggests reasonable profile shapes for both CH₄ and N₂O 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.**

35 **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

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 O₃ 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 O₃ partial and total columns for 2010 are essentially similar to 2009, and also the Antarctic O₃ 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.

20

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.

25

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?

35 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.*

5 *Is it not only O₃ that is important for radiation calculations? Better stratospheric O₃ could improve stratospheric temperatures?*

Indeed better stratospheric O₃ 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
10 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.

15 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.

20

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
25 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

30 *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.

35

Anonymous Referee #2

5 *The purpose of the paper is to describe and benchmark a new version of the IFS model. This version has separate chemistry modules (mechanisms) for the troposphere and stratosphere, where the decision of which module to call is determined by the altitude of the grid box with respect to the tropopause. The stratospheric chemical mechanism comes from an assimilation system (BASCOE). The previous version of the IFS had tropospheric chemistry plus linearized stratospheric O3. The paper concludes that a new simulation that uses both chemical mechanisms (called CIFS-TS) has good stratospheric O3, NO2 and other reactive trace gases compared to satellite data sets.*

10

A goal of the paper is to demonstrate that their method of using the tropospheric mechanism/solver for tropospheric grid boxes and the stratospheric solver for stratospheric grid boxes is a computationally efficient way to calculate the full chemistry of the atmosphere. The biggest problem with this paper is that they did not actually test this. To demonstrate their method, they need to have run a simulation where tropospheric and stratospheric reactions were solved TOGETHER and NOT split into 2 mechanisms.

15

Those results could then be compared with their 'split' method. Ideally, this would show that their method was faster (how much faster?) yet produced essentially the same results. I recommend they do this and then rewrite this manuscript. This experiment would not only satisfy the stated goal of the paper but it would also eliminate the confusion in the comparisons (see below) regarding transport/advection differences between BASCOE-CTM and the CIFS-TS.

20

We thank the reviewer for his/her valuable comments on our manuscript. Indeed a system with tropospheric and stratospheric chemistry resolved throughout the atmosphere (in the remainder referred to as 'C-IFS-Atmos'), as opposed to the reported more efficient approach in C-IFS-TS, had been already developed and briefly mentioned in the manuscript but its evaluation was missing. The main reason for not presenting this was that the stratospheric chemistry was treated very similar and hence small differences in model results between C-IFS-Atmos and C-IFS-TS could be expected in the stratosphere. Discrepancies to the observations mainly raise from common stratospheric chemistry model assumptions (e.g. PSC treatment, photolysis) and differences in transport treatment between C-IFS and BASCOE-CTM. Larger differences can be only be expected when approaching the tropopause. We would like to point out that the original manuscript does mention that C-IFS-Atmos is 50% more expensive than C-IFS-TS (at the end of Sec. 2.3), essentially due to the larger chemical mechanism throughout the atmosphere that needs to be solved.

25

30

In response to the reviewer we acknowledge that an explicit evaluation of the differences between C-IFS-TS and C-IFS-Atmos does clarify our goal, which indeed also aims at presenting our methodology with separate tropospheric and stratospheric chemistry in C-IFS-TS. We therefore now include explicitly C-IFS-Atmos in our model evaluation and show that the differences with the more efficient approach in C-IFS-TS are as small as expected.

35

This expansion of the model evaluation also increases the usefulness of the comparison with BASCOE-CTM since it is now possible to compare two models with the same stratospheric chemistry but different transport schemes (BASCOE-CTM versus C-IFS-TS) and two models with the same transport but (slightly) different chemical schemes in the stratosphere (C-IFS-TS versus C-IFS-Atmos). A recent study by de Grandpré *et al.* (2016) is now cited to illustrate the type of issues raised in the stratosphere by the semi-lagrangian advection scheme.

I don't agree with the statement in the abstract that the new model configuration shows good performances of stratospheric O₃, NO₂, and other tracers. The figures chosen to demonstrate good representation of various stratospheric constituents in the CIFS-TS model generally show fair to poor agreement with observations. Stratospheric O₃, for example, often looks worse (or at least no better) than it did in the CB05 (trop only) or BASCOE (strat only) versions. This new model does not appear to be an improvement over previous model versions.

The comparisons between CIFS-TS and BASCOE-CTM are confusing. When stratospheric species such as NO₂, HNO₃, and O₃ are compared, the results are different. I thought the primary goal of the paper was to compare the chemical mechanisms, but since the results are rather different, there must be transport (or meteorological field) differences too. This is alluded to on page 7, lines 26-28. The transport/advection needs to be the same in the two simulations in order to compare the chemical mechanisms.

It should be made clearer in the text what the differences are between CIFS-TS and BASCOE-CTM. My overall recommendation is to test a combined (strat+trop) solver and compare the results to trop only, strat only, and the 'split' method presented here. The results will provide a good benchmark and will be easier to interpret if all experiments are performed with the same transport code and meteorological fields.

The reviewer appears confused by the selection of model setups chosen in our manuscript. In essence, the C-IFS is a Global Circulation Model (GCM) designed for meteorological analyses and forecasts where a module for chemistry has been included to extend its abilities in terms of atmospheric composition. On the other hand, the BASCOE system is a dedicated data-assimilation system for stratospheric composition, based on a Chemistry Transport Model (CTM) environment, i.e. a completely independent system to C-IFS.

The impact of various chemical mechanisms was evaluated through comparison of C-IFS-T (with linear ozone treatment in the stratosphere) and C-IFS-TS (which uses the identical chemical parameterization in the stratosphere as BASCOE-CTM).

The BASCOE-CTM is driven by meteorological fields from the C-IFS run, but still uses a different numerical scheme for the advection, and is running on a different grid as compared to the C-IFS runs. Note that in this setup of the BASCOE system the chemical data-assimilation is switched off, hence purely reflecting the forward model capabilities. Hence comparison between BASCOE-CTM and C-IFS-TS is a clean method to evaluate differences due to the representation of transport with identical meteorological fields, and not suited for the evaluation of differences in the chemical treatment since there are no such differences.

The additional model run, C-IFS-Atmos, where tropospheric chemistry is extended throughout the stratosphere, and vice versa, is now included to assess the impact of assumptions of the reduced chemistry in C-IFS-TS.

In response to the reviewer's concerns we now extend the table describing the model versions (see also the comment below), and extend the description between differences of the various setups. Notably in the
5 introduction we now write:

**“The CB05 tropospheric scheme has been combined with the stratospheric scheme from BASCOE-CTM to form a single chemistry mechanism that encompasses tropospheric and stratospheric chemistry throughout the atmosphere, here referred to as C-IFS-Atmos. However, this approach appears computationally
10 expensive, due to the extended chemical mechanism. Therefore”**

And also:

**“In this optimized approach we developed a flexible setup where -within a single framework- either the
15 tropospheric or stratospheric chemistry modules are addressed, referred to as C-IFS-TS. In this approach the parameterizations for the chemistry, including the respective chemistry mechanisms as optimized for troposphere and stratosphere separately, are retained.**

In this paper we describe our **two merging approaches** and provide benchmark evaluations of the **C-IFS-Atmos and C-IFS-TS** systems with focus on the stratospheric composition. **The ancestor BASCOE-CTM is also included
20 in the comparison through a forward model run (without chemical data assimilation) in order to provide insight in the differences caused by the treatment of transport between C-IFS and BASCOE.”**

The model evaluation has been extended to include results obtained with C-IFS-Atmos, as well as an evaluation of the stratospheric composition (including O₃, HNO₃ and NO₂) in C-IFS-T, to explicitly identify the impact of the
25 newly implemented stratospheric chemistry within the C-IFS framework.

Other points

*30 It would be helpful to add a table that lists the specifications of each of the models used and notes how dynamical fields are obtained (e.g., forecast, assimilation, . . .?), chemical mechanism, resolution, etc. For example, BASCOE is an assimilation system, but it's only the BASCOE stratospheric chemical mechanism that used here, right? And BASCOE-CTM means the assimilated (reanalysis) fields have been saved and then
35 are being used in an offline chemistry transport model? Presumably it is the same offline model that the C-IFS forecast fields are used in? If what I am asking does not make sense, please take this as an indication that I am confused by the descriptions of the models.*

In response to the reviewer's concerns in Section 2.3 we expanded Table 2 which lists the specifics of the various model systems, as also given below. Further, we want to make clear that the C-IFS experiments have been run in 'nudged meteo' mode, by relaxation of the meteorology towards ERA-Interim, as we also write in Section 3. The BASCOE-CTM run is driven by the identical meteorology from the C-IFS experiment (and in turn from ERA-Interim), but applies its own advection algorithm which is clearly different from the one used in IFS. In Section 3 we also make more clear what are the differences between C-IFS and BASCOE-CTM. As discussed above, the BASCOE-CTM results are included as a reference of what can optimally be achieved with C-IFS-TS and C-IFS-Atmos in the stratosphere, using only simulations nudged with specified dynamics and unconstrained composition. Specifically we now write:

10

"Meteorology in the C-IFS runs is relaxed towards ERA-Interim (...) The performance of the C-IFS runs has further been compared against the BASCOE-CTM (without **chemical** data assimilation), using the same chemical mechanism and parameterizations for **photolysis and heterogeneous chemistry** as implemented in the C-IFS-TS. **This serves as a model reference for the C-IFS implementation of stratospheric chemistry. While C-IFS evaluates tracer transport on a reduced Gaussian grid, the BASCOE-CTM uses a regular latitude-longitude grid. It is run here with a resolution of 1.125° lon / lat similar to the resolution chosen for C-IFS, and on the same vertical grid of 60 levels. The BASCOE-CTM is driven by temperature, pressure and wind fields simulated by the C-IFS runs. However, while BASCOE adopts a flux-form advection scheme (Lin and Rood, 1996) the IFS uses the Semi-Lagrangian scheme for advection, accounts for vertical diffusion and includes a parameterization for convection (ECMWF, 2015).** Using essentially the same dynamical fields together with an identical implementation of the chemistry code should allow to identify differences **due to the different** transport schemes between C-IFS and the BASCOE-CTM. Common chemical biases between both systems also point at issues in the chemical parameterizations **such as reaction mechanism, photolysis, heterogeneous chemistry and sedimentation.**"

25

Table 2. Number of trace gases, the chemistry scheme in troposphere and stratosphere, and corresponding number of reactions (gas-phase / heterogeneous and photolytic), as well as specification of the circulation model and computational expenses of a one-month run on T255L60 in terms of system billing units (SBU) for various C-IFS model versions. For completeness also the BASCOE-CTM system is indicated.

30

	C-IFS-T	C-IFS-S	C-IFS-Atmos	C-IFS-TS	BASCOE-CTM
No. trace gases	55	59	99	99	59
Chemistry scheme in troposphere	CB05	BASCOE (P<400hPa)	CB05+BASCOE	CB05	BASCOE (P<400hPa)
Chemistry scheme in stratosphere	CB05/ Cariolle	BASCOE	CB05+BASCOE	BASCOE	BASCOE
No. reactions (gas / het / photo)	93/3/18	142/9/52	211/11/60	93/3/18 or 142/9/52	142/9/52
Circulation model	GCM	GCM	GCM	GCM	CTM
SBU	2075	2500	4563	3076	- ^a

^aBASCOE does not run on the ECMWF supercomputing facility and hence cannot be compared directly to C-IFS in terms of computational resources.

- 5 *Regarding ‘tracer species’ or similar expression found in many places, ‘tracer’ means a species that is unreactive and can be used to trace something, like transport. I think you mean ‘trace gas’ rather than tracer because that can be used in a general way to talk about any type of constituent in the model. Please search on ‘tracer’ in the document to identify where you mean trace gas or constituent.*
- 10 We thank the reviewer for this comment, and changed the wording accordingly throughout the document.

- p. 3, l.24. Are you saying the chemistry in the modules is parameterized? Or are you referring to the chemical mechanisms when you say ‘chemical parameterization’? A parameterization for chemistry is not the same thing as a chemical mechanism. Sometimes ‘chemical schemes’ is used, which is fine for referring to the mechanism.*
- 15

This confusion occurs throughout the paper. Please check each occurrence of ‘parameterization’ to verify the right words were chosen.

In this occasion the phrase ‘chemistry parameterization’ referred to all chemical conversion processes that require a parameterization, including aqueous phase and heterogeneous reactions as well as photolysis and parameterizations for sedimentation. Indeed this refers to more than just the definition of the chemical mechanism. To accommodate the concerns of the reviewer we had a critical look at our terminology for ‘parameterization’ in the complete manuscript, and changed it where appropriate (see also below). In this instance mentioned by the reviewer in Sec. 2.0 (p.3, l.24) we only wish to guide the reader forward to the specific sections on stratospheric/tropospheric chemistry, but to prevent potential confusion we now write “tropospheric (CB05-based) chemistry parameterizations”.

p. 4, l. 15. The threshold temperature for NAT formation is pressure dependent. The manuscript indicates that 194 K was chosen as the threshold regardless of pressure. That would not be the correct way to calculate it.

While the BASCOE CTM was used some time ago for detailed studies of the processes leading to polar ozone depletion (Daerden et al., ACP, 2007), the corresponding microphysical module was removed (due to huge computational costs) and replaced by this very crude parameterization. Indeed the BASCOE CTM is now designed as a generic model which (until now) needs only to be good enough to allow the successful assimilation of satellite observations of stratospheric composition. Yet both reviewers indicated a simple improvement which could be implemented quickly enough for this revision of the manuscript.

Hence we have revised the PSC-parameterization, which is no longer purely temperature-dependent. We now remove H₂O and HNO₃ where their respective partial pressures exceed the equilibrium values, according to Murphy and Koop (2005), and Hanson and Mauersberger (1988). The time scale for irreversible removal of HNO₃ has been revised from 100 days in the original setup to 20 days, in accordance with the smaller regional and temporal extent where NAT particles are assumed to exist. This led to significant improvements in the H₂O and HNO₃ bias in the region where PSC formation is possible, and accordingly to a slight improvement in O₃ profile shapes in terms of a reduced positive bias at 100 hPa and reduced negative bias at 20hPa during August-September over the Neumayer and Syowa stations (see also below). Nevertheless, the HNO₃ timeseries for the BASCOE-CTM, CIFS-TS and C-IFS-Atmos models suggest that denitrification proceeds more slowly and ends one month later than observed by Aura MLS observations, which may be attributed to our crude modelling approach for the formation and sedimentation impact of NAT PSC. We have modified the respective section as follows:

“Ice PSCs are presumed to exist at any grid point in the winter/spring polar regions where **water vapour partial pressure exceeds the vapour pressure of water ice (Murphy and Koop, 2005)**. Nitric Acid Tri-hydrate (NAT)

PSCs are assumed **when the nitric acid (HNO₃) partial pressure exceeds the vapour pressure of condensed HNO₃ at the surface of NAT PSC particles (Hanson and Mauersberger, 1988).**”

5

p. 5, l. 31. I don't understand what is meant by O1D and O3P being described implicitly, as opposed to being treated explicitly.

10 Within the troposphere the O¹D is produced from O₃ photolysis and assumed to react instantaneously, with only reaction products H₂O and again O₃. As the O¹D (and O) lifetime is much shorter than the integration time, while only reactions with N₂ and O₂ are assumed in the troposphere, the O¹D concentration can be considered in equilibrium over the integration time and hence does not need to be treated explicitly. The same argumentation holds for O³P, produced from O₂ photolysis in upper troposphere, and assumed to only react
15 with O₃ to form O₂, and with O₂ to form O₃. This is different for the stratosphere, where O1D and O3P are involved in many more reactions. To clarify in the manuscript we reformulate this as follows:

“It is worth mentioning that the constituents O¹D and O³P, produced from O₃ and O₂ photolysis, **are not explicitly computed in the troposphere, as O¹D and O³P are assumed to react with O₂, O₃ and N₂ only. This is**
20 **different for the stratosphere, where O¹D and O³P are involved in many reactions.**”

p. 6, l. 24. ‘solar radiation reaches the stratosphere earlier than the surface. . .’ as written this sounds like it is referring to delay caused by the speed of light! I doubt this was intended; it needs better wording.

25 The reviewer is clearly technically correct. We changed the formulation to a more compact formulation, leaving out the suggestion of a different timing:

“Also the presence of sunlight at solar zenith angles (SZA) larger than 90° at high altitudes needs to be accounted for in the stratosphere **due to the Earth’s curvature, but may be neglected** in the troposphere. **This plays a role in the timing of springtime ozone depletion in the polar lower stratosphere.**”

30

*p. 7, Section 2.3.1. For JO3, the lack of a ‘jump’ in O3 may be because photolysis is unimportant (slow) near 100 hPa, so O3 is probably long-lived relative to the photochemical lifetime. JNO2 is much larger so I’m not sure why there isn’t a jump – can you explain this? It would be useful if you showed the simulated O3 and NO2 profiles in
35 Fig. 1 to demonstrate the lack of a jump. What is the meaning of ‘JO3_TB’ in the title of one plot? No similar title for the other plot.*

The reviewer is correct in that the presence or absence of jumps associated to the change in the reaction mechanism depends on the lifetime of the species, in combination with the magnitude of the change in the dominating reaction (or photolysis) rate with the different chemical mechanism. For O₃ the photolysis is a dominating loss term in this altitude range, but still the reaction rate is sufficiently low (i.e. the O₃ lifetime sufficiently long) such that jumps in the photolysis rate do not lead to jumps in O₃ concentrations. For NO₂ the photolysis rate is much larger, and resulting in a short (less than 1 hour) NO₂ lifetime. Jumps in photolysis rate potentially result in jumps in NO and NO₂ concentrations. Nevertheless, the jump is sufficiently small (for J-NO₂ we verified that the difference in photolysis rates around the tropopause is generally below 5%), such that the NO₂ concentrations do not show a significant jump. We now provide figures in the supplementary material where we present instantaneous profiles of a range of trace gases at the tropopause interface. We extended the discussion on this aspect with the following sentences:

“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.**”

20 *Section 2.3.2, l. 8. It's unclear whether you're saying NO, NO₂, and have the mass fixed applied or whether they are the few species where the mass fixed isn't applied. How badly is H₂O not conserved in the stratosphere? This will conceivably cause problems for stratospheric chemistry. It would be useful to see a 1-year time series of the H₂O mass above 100 hPa.*

25 As explained in the manuscript the reason for switching off the mass fixer for the stratospheric H₂O tracer is because otherwise mass conservation errors originating from the troposphere lead to spurious redistribution of H₂O mass towards the stratosphere. Therefore, in fact due to switching off the mass fixer, the H₂O mass in the stratosphere remains very stable. We illustrate this by Figure R1 (below), which shows indeed absence of any trend in stratospheric H₂O columns over the years, indicating that H₂O mass conservation is sufficiently well
30 ensured in the stratosphere. This figure also shows that H₂O total columns are essentially identical in C-IFS-Atmos and C-IFS-TS.

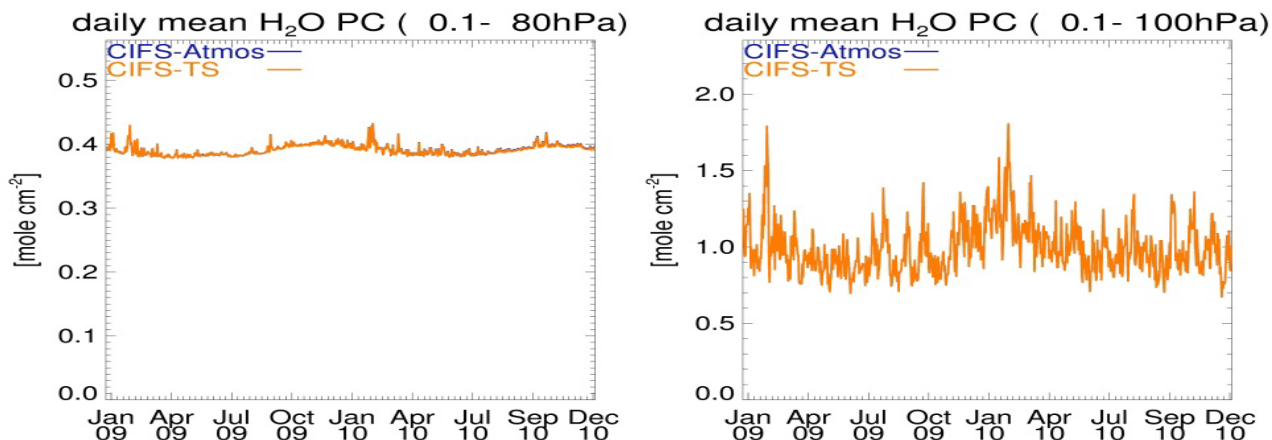


Figure R1. Evolution of global, daily mean H₂O partial columns (left: 0.1-80 hPa, right: 0.1 – 100 hPa) for the runs C-IFS-Atmos (blue) and C-IFS-TS (orange) for January 2009 to December 2010. C-IFS-TS is on top of C-IFS-Atmos.

5 In the manuscript we now write:

“The global advection errors in H₂O that essentially originate from the tropospheric part because by far most H₂O mass is located in the troposphere and the spatial gradients are much more pronounced. This should not affect the stratospheric H₂O mass budget, therefore the global mass fixer for the stratospheric H₂O tracer has been switched off. This prevents spurious trends in stratospheric H₂O columns over the years (not shown),
 10 **indicating that H₂O mass conservation is well ensured in the stratosphere.”**

p. 7, last 3 lines. This sentence says you are looking to identify differences in transport schemes. This confuses the issue of evaluating the chemical mechanisms (and their implementation). This evaluation should be
 15 *performed using the same dynamical fields with the same model. If the advection schemes are also different, then we cannot actually test the impact of chemical mechanisms alone. And does ‘parameterization’ in line 28 refer to the different chemical mechanisms?*

For a discussion on the selection of the model setups evaluated in our manuscript we refer to our response to
 20 the reviewer’s first general comment. We now extend the evaluation with results from run C-IFS-T, to explicitly identify the impact of the newly implemented stratospheric chemistry within the C-IFS framework. Indeed, the BASCOE-CTM run uses identical chemistry to C-IFS-TS and is not introduced to assess the chemical mechanisms, but rather differences due to the transport scheme while using the same dynamical fields. Here, the ‘parameterizations’ refer to the reaction mechanism, photolysis, heterogeneous chemistry and sedimentation,
 25 as we now explicitly write.

p. 8, l. 27, ‘first Science Satellite’?

Indeed this is the meaning of the abbreviation ‘SCISAT-1’.

5 *p. 8, l. 30-31. Suggest to change to ‘. . .between 6-30 km agree to within 15% of independent . . .’ For all the figures that are line plots (starting with Figure 2), the blue and black lines are hard to distinguish. Please do something with the line thickness and colors to improve readability.*

10 We changed this according to the reviewer’s suggestions, thank you. We have improved color-coding and general figure quality, which unfortunately also had seen some degradation in the stage of pdf-generation from the word-document.

20 *Section 4, Model Evaluation p. 9, lines 14-19. This paragraph would benefit by a general statement of the purpose of this comparison. It appears the purpose is to show that the TS mechanism looks more like the observed total column O3 than does the trop-only code (with linearized strat O3). One would expect the TS O3 to be better than the linearized O3 of CB05, but there should also be a comparison with the stratonly code. Comparing with the O3 results in Fig. 6, I think the strat chem O3 columns would be lower than the TS mechanism. I guess they aren’t the same because the BASCOE-CTM has different transport. Again, not having the same transport in all the simulations really interferes with a useful comparison.*

25 We now replace this figure with an evaluation of the partial columns (10-100hPa) against Aura MLS observations, to emphasize the performance in the stratosphere. We now also include results from C-IFS-Atmos and C-IFS-T, as well as from BASCOE-CTM to assess the impact of different chemistry approaches, and different transport scheme. The new evaluation shows more clearly the benefits and limitations of the new approach in C-IFS-TS, as compared to C-IFS-T (with linearized O3), as well as differences with BASCOE-CTM (which contains stratospheric chemistry only and uses the same dynamical fields as C-IFS but with a different transport algorithm). We have moved the assessment of O₃ total columns against the Multi-Sensor Reanalysis to the Supplementary Material . This does not include results from BASCOE-CTM, considering it’s missing tropospheric contribution. The manuscript text has been revised accordingly.

30 Furthermore, in Sec. 3 we now include a few general statements to clarify the purpose of the various model evaluations:

35 **“Intercomparison of the runs C-IFS-TS and C-IFS-Atmos aims to provide a justification of our approach to split the chemistry into two regions, while intercomparison of C-IFS-TS with C-IFS-T can be used to identify the changes to stratospheric composition modelling between full stratospheric chemistry and the baseline approach with the linear ozone scheme.”**

p. 9, discussion of Figs. 3-4. I do not agree that there are meaningful, reduced biases in the TS version. The linearized O₃ chemistry of the trop mechanism gives different results from the TS version, but not really worse. These figures show that TS is not an improvement over trop-only. I think the use of mPa for the O₃ bias (lower panels) is misleading and probably minimizes the appearance of the disagreement in the middle stratosphere.

By evaluating O₃ profiles in terms of partial pressure biases in the original manuscript we intended to focus on the contribution of each pressure region to the O₃ TC, with larger weights in the lower stratosphere. This is now assessed in detail in the revised Figure 2 that presents the evolution of the O₃ partial columns (10-100hPa). Hence in accordance with the reviewer request we now present results of O₃ profiles in units ppmv, indeed giving more focus to the altitude with maximum O₃ concentration, at around 10 hPa. Also we now average over all profiles in 2009 and 2010, to improve the statistics, and include results from C-IFS-Atmos. We agree with the reviewer that we have been too positive when describing the C-IFS-TS results as compared to C-IFS-T (with linear chemistry). We have rewritten this section to provide a more balanced discussion.

15

p. 9, discussion of Fig. 5. I cannot tell the difference between obs and CIFS-T lines in the figure. There is no line color/style for the observations in each panel's legend. The TS O₃ agrees with one of the black lines (obs or CIFS-T??) near and below 100 hPa – sometimes – but the TS O₃ consistently has poor agreement above 50 hPa. Why? Since the TS (red) line often does not agree with either black line – I see no basis for claiming good agreement. Additionally, Syowa is often near the vortex and has large daily variability. Were the simulated profiles used in this figure calculated from the same days of the month as the Syowa data?

Figure 5 in the manuscript has been regenerated based on the revised model simulations. Color-coding has been updated, and error-bars denoting the model and observation variability are now included. Note that all comparisons with observations, including fig. 5, use three-hourly model output which has been collocated in time and space to the observations. This is now explained in Section 3. Please also note that in our section describing Figure 5 we do not claim general good agreement, as the reviewer suggests, but rather point at regions and months where C-IFS-TS performs well, and others where it shows biases compared to observations. The revised simulations have seen some improvement in terms of vertical profile shape during ozone hole conditions, see also Figure R2, below, for an assessment of the differences to the C-IFS simulations presented in the GMDD manuscript. This is due to the improved PSC parameterization, especially above 50hPa in August and September where PSCs were allowed to exist in the C-IFS-TS run for the original GMDD version. The remaining discrepancies could still be caused by the limitations of the revised PSC parameterization. We now write:

35

“For the 2009 Antarctic ozone hole season (Fig. 5) the C-IFS-TS and C-IFS-Atmos shows a positive bias at ~100 hPa for August and September, and negative bias at higher altitudes (50-10 hPa), where C-IFS-T shows a positive bias.”

Additionally we now provide a closer analysis of the performance during polar ozone depletion, by presenting time series of HNO₃, ClO and O₃ during the 2009 ozone depletion over Antarctica (the new Figure 6). This new figure clearly shows the abilities and limitations of the different versions of C-IFS to describe this event. Specifically we now show that denitrification, which is clearly not modelled in C-IFS-T, starts at the correct time in the models with stratospheric chemistry, although it appears to last about one month too long as compared to the observations. We note that in the original manuscript, where this parameterization depended only on T, the denitrification started one month too late.

10

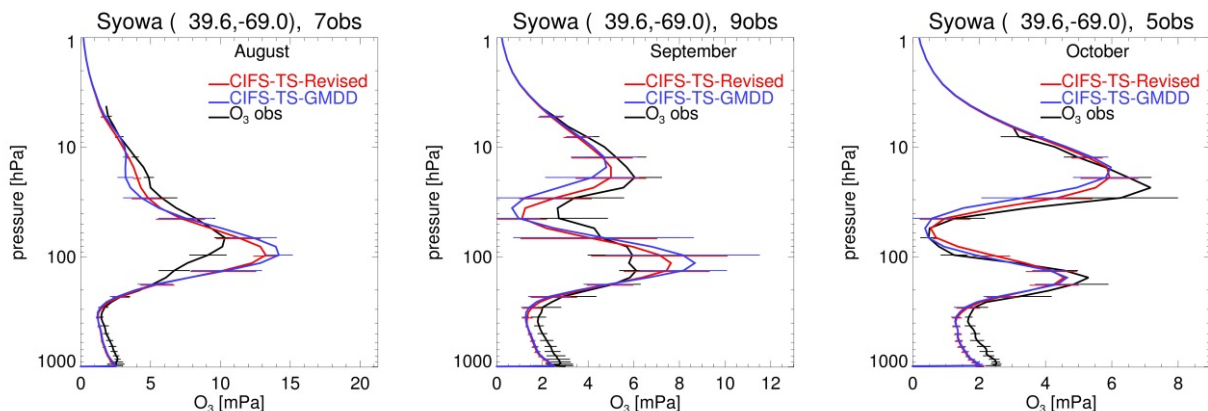


Figure R2. Evaluation of ozone in units mPa against WOUDC ozone sondes at Syowa station during August-October 2009. Black: ozone sonde, red: C-IFS-TS in the Revised model version, blue: C-IFS-TS in original GMDD version. Error bars denote the 1-sigma spread in the models and observations.

15

p. 9, lines 31-32. If you made a difference plot between MIPAS and the simulations, then you might be able to say whether there is good agreement. As presented, the conclusion can't be drawn that there are 'small biases'. Near the tropical maximum the TS looks slightly better than the BASCOE-CTM. Again, assuming that some of the differences are due to dynamical fields or advection scheme, this comparison isn't very useful.

20

As argued before, the inclusion of BASCOE-CTM is especially useful to diagnose if model biases arise due to different advection schemes or due to different chemistry schemes. To accommodate the reviewer's comments to better quantify the C-IFS versions, as compared to BASCOE-CTM we now include results from C-IFS-Atmos and C-IFS-T, and provide a more balanced discussion. Finally we have strengthened the evaluation of ozone with two new figures in the Supplementary Material: the quantitative comparison is strengthened by a new comparison of vertical profiles with Aura MLS (Fig. S7) and the discussion of Fig. 7 (top row) is confirmed with a corresponding evaluation also using Aura MLS (Fig. S8). The discussion of ozone on fig. 7 now reads:

25

5 “The evaluation of the zonal mean ozone **mixing ratios** against MIPAS observations shows good general agreement, Fig. 7, with **all four modelling experiments providing similar features. The tropical maximum of O₃ mixing ratio at 10 hPa is under-estimated in all experiments but to a larger extent in those which model stratospheric photochemistry explicitly (BASCOE CTM, C-IFS-TS, C-IFS-Atmos) than in C-IFS-T, in line with the evaluation against O₃ sondes for June-July-August (figure 4). The same evaluation against MLS observations provides exactly the same conclusions (figure S8, supplementary material).”**

10 *p.10, lines 5-9. What is the message here? The CIFS has a terrible high bias in nighttime NO₂ and a large low bias in HNO₃. Why is the CIFS simulation worse than BASCOE? There is no clear explanation here.*

15 We acknowledge that these results are not satisfying; unfortunately at current stage we do not have a clear explanation for this. Nevertheless, we want to highlight that the model performance has still improved compared to C-IFS-T, whose results we now include. Also we explicitly provide these figures to indicate current limitations of our model. We now write:

20 **“Even though a clear improvement compared to run C-IFS-T is found, further investigation is necessary to diagnose origins of the biases in night-time NO₂ above 10 hPa and in HNO₃ between 10 and 70 hPa.”**

Also in the conclusions section we include such a sentence.

25 *p. 10, lines 10-20 (Fig. 8). N₂O and CH₄ profiles do NOT assess vertical transport. Their profiles below 10 hPa represent a balance between the vertical and horizontal components of the residual mean circulation. That balance depends on latitude, that is, whether the profile is from the tropical upwelling region or somewhere in the midlatitudes (horizontal and vertical motions matter and so does mixing), or isolated inside the polar vortex (descent). Above 10 hPa, profiles are more strongly influenced by chemical loss so the 2 simulations should look very similar there. The CIFS-TS simulation tends to look worse than the BASCOE CTM or the observations between 10-50 hPa. This suggests circulation and/or mixing problems in the tropics and SH.*

30 *O₃ at 20 hPa is strongly influence by chemistry, not just transport. These paragraphs indicate a lack of understanding of transport circulation and its diagnosis, as well as any understanding of what controls stratospheric ozone distributions.*

35 These two paragraphs are indeed rather vague and mistakenly use Figure 8 as a diagnostic for “vertical” transport. We thank the reviewer for pointing this out and setting us on the right track.

The reviewer’s suggestion about circulation and/or mixing problems confirms that figure 8 is a preliminary yet valid diagnostic for transport processes in general, and that it indicates an unidentified issue for the representation of these processes in C-IFS. We do not think that circulation is the culprit because the BASCOE

CTM is driven by meteorological fields which are the output of C-IFS. As noted above, the revised manuscript gives (at the beginning of section 3) a few more details about the modelling of transport in both models:

5 **“The BASCOE-CTM is driven by temperature, pressure and wind fields simulated by the C-IFS runs. However, while BASCOE adopts a flux-form advection scheme (Lin and Rood, 1996) the IFS uses the Semi-Lagrangian scheme for advection, explicitly accounts for horizontal diffusion and includes a parameterization for convection (Ritchie et al., 1995; ECMWF, 2014).”**

10 The revised manuscript also lists the relevant transport processes in the stratosphere along with a general reference on this topic (Shepherd, 2007) and states what specific pieces of C-IFS may be responsible for the problem(s). Since O₃ at 20 hPa is strongly influenced by both chemistry and transport, we stand with the statement that this transport issue “could also contribute” (directly) to the ozone biases noted below 10 hPa. But their attribution to an “excess of vertical transport” was clearly a mistake. No further statement can be made on this topic because further evaluation of stratospheric transport processes in C-IFS is beyond the scope
15 of this paper. The two problematic paragraphs in section 4 have thus been re-written as follows:

**“Fig. 9 shows an evaluation of N₂O and CH₄ 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
20 the polar vortex, and stratosphere-troposphere exchange (Shepherd, 2007). Moreover, N₂O is the main source of reactive nitrogen in the stratosphere while CH₄ is one of the main precursors for stratospheric water vapour. The figure suggests reasonable profile shapes for both CH₄ and N₂O 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
25 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
30 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.”**

35 The revised conclusions mention that this area deserves further investigation:

“For the long-lived tracers CH₄ and N₂O, larger errors with respect to limb-sounding retrievals were found between 10 hPa and 100 hPa than with the BASCOE-CTM, suggesting difficulties in representing slow

transport processes. The BASCOE CTM experiment shown here was driven by three-hourly wind fields output of the C-IFS-T experiment. Hence this discrepancy is due to a difference in the representation of the transport processes between the GCM and the CTM, i.e. the numerical scheme used for advection (Semi-Lagrangian versus Flux-Form), the convection (parameterized in C-IFS but neglected in BASCOE CTM) or the diffusion (parameterized in C-IFS but not explicitly considered in the CTM). Hence, stratospheric transport in C-IFS will be an area for further evaluation and developments.”

Additional references

de Grandpré, J., Tanguay, M., Qaddouri, A., Zerroukat, M., and McLinden, C.A. (2016), Semi-Lagrangian Advection of Stratospheric Ozone on a Yin–Yang Grid System. *Monthly Weather Review* 144:3, 1035-1050. DOI: <http://dx.doi.org/10.1175/MWR-D-15-0142.1>

ECMWF: IFS documentation – Cy41r1. Operational implementation 12 May 2015. Available from <http://www.ecmwf.int/en/forecasts/documentation-and-support/changes-ecmwf-model/ifs-documentation> (last access on 5 July 2016), 2015.

Froidevaux, L., Jiang, Y. B., Lambert, A., Livesey, N. J., Read, W. G., Waters, J. W., Browell, E. V., Hair, J. W., Avery, M. A., McGee, T. J., Twigg, L. W., Sumnicht, G. K., Jucks, K. W., Margitan, J. J., Sen, B., Stachnik, R. A., Toon, G. C., Bernath, P. F., Boone, C. D., Walker, K. A., Filipiak, M. J., Harwood, R. S., Fuller, R. A., Manney, G. L., Schwartz, M. J., Daffer, W. H., Drouin, B. J., Cofield, R. E., Cuddy, D. T., Jarnot, R. F., Knosp, B. W., Perun, V. S., Snyder, W. V., Stek, P. C., Thurstans, R. P., and Wagner, P. A.: Validation of Aura Microwave Limb Sounder stratospheric ozone measurements, *J. Geophys. Res.*, 113, doi:10.1029/2007JD008771, 2008a.

Hanson, D., and Mauersberger, K.: Laboratory studies of the nitric acid trihydrate: Implications for the south polar stratosphere, *GRL* 15 (8), pp 855–858, DOI: 10.1029/GL015i008p00855, 1988.

Hitchman, M. H., M. McKay, and C. R. Trepte (1994), A climatology of stratospheric aerosol, *J. Geophys. Res.*, 99(D10), 20689–20700, doi:10.1029/94JD01525.

Jablonowski, C., and Williamson, D. L.: The pros and cons of diffusion, filters and fixers in atmospheric general circulation models, in: *Numerical Techniques for Global Atmospheric Models* (pp. 381-493). Springer Berlin Heidelberg, doi:10.1007/978-3-642-11640-7, 2011.

Lin, S.-J. and Rood, R. B.: Multidimensional flux-form semi-Lagrangian transport schemes, *Mon. Weather Rev.*, 124, 2046–2070, 1996.

Murphy, D. M. and Koop, T. (2005), Review of the vapour pressures of ice and supercooled water for atmospheric applications. *Q.J.R. Meteorol. Soc.*, 131: 1539–1565. doi: 10.1256/qj.04.94

Ritchie, H., Temperton, C., Simmons, A., Hortal, M., Davies, T., Dent, D. and Hamrud, M.: Implementation of the Semi-Lagrangian Method in a High-Resolution Version of the ECMWF Forecast Model, *Mon. Wea. Rev.*, 123, 489–514, 1995.

5 Santee, M. L., A. Lambert, W. G. Read, N. J. Livesey, G. L. Manney, R. E. Cofeld, D. T. Cuddy, W. H. Daffer, B. J. Drouin, L. Froidevaux, R. A. Fuller, R. F. Jarnot, B. W. Knosp, V. S. Perun, W. V. Snyder, P. C. Stek, R. P. Thurstans, P. A. Wagner, J. W. Waters, B. Connor, J. Urban, D. Murtagh, P. Ricaud, B. Barret, A. Kleinboehl, J. Kuttippurath, H. Kuellmann, M. von Hobe, G. C. Toon, and R. A. Stachnik. Validation of the Aura Microwave Limb Sounder CIO measurements. *J. Geophys. Res.*, 113:D15S22, doi:10.1029/2007JD008762, 2008.

10 Shepherd, T. G.: Transport in the Middle Atmosphere, *J. Met. Soc. Jap.*, 85B, 165–191, doi: 10.2151/jmsj.85B.165, 2007.

Astrid Kerkeweg, Executive Editor

Please add a version number or unique identifier either for each of the models you combined or for your newly created model in the title upon your revised submission to

5 GMD.

We thank Dr Kerkeweg for referring to the requirements of papers published in GMD. In fact also the editor of this manuscript, Dr Grewe, had pointed us to these requirements, where we responded that version labeling was not yet introduced, in line with arguments given by Flemming et al. (2015): The cycle number of the IFS
10 reflect the development of the NWP code, but are not yet linked to the development of the chemistry modules. The current chemistry scheme is used for several IFS cycles. As part of the Copernicus Atmosphere Monitoring Service a proper version naming convention will be introduced. In case our response is not acceptable for acceptance in GMD we will now introduce such a version number to this new model implementation.

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

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Abstract. We present a model description and benchmark evaluation of an extension of the tropospheric chemistry module in the Integrated Forecasting System (IFS) of the European Centre for Medium-Range Weather Forecasts (ECMWF) with stratospheric chemistry, referred to as C-IFS-CB05-BASCOE (for brevity here referred to as C-IFS-TS). The stratospheric chemistry originates from the one used in the Belgian Assimilation System for Chemical Observations (BASCOE), and is here combined with the modified CB05 chemistry module for the troposphere as currently used operationally in the Copernicus Atmosphere Monitoring Service (CAMS). In our approach either the tropospheric or stratospheric chemistry module is applied depending on the altitude of each individual grid box with respect to the tropopause. An evaluation of a 42.5 year long C-IFS-TS simulation with respect to various satellite retrieval products and in-situ observations indicates good performance of the system in terms of stratospheric ozone, ~~nitrogen dioxide as well as other reactive tracers in comparison to various satellite retrieval products and a general improvement in terms of stratospheric composition compared to the C-IFS predecessor model version.~~ Possible issues with transport processes in the stratosphere are identified. This marks a first 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

25 Existing earth observation systems in combination with global circulation models (GCMs) help to provide a better understanding of the Earth's atmospheric composition and changes therein (Hollingsworth et al., 2008). For the troposphere, hemispheric transport and chemical conversion of atmospheric composition influences regional air quality (Pausata et al., 2012; Im et al., 2015, Marécal et al., 2015). Also ~~analyses and forecasts the amount~~ of stratospheric ozone directly ~~impact~~ impacts the forecast capabilities of surface solar irradiance (Qu et al., 2014), stressing the relevance of good stratospheric ozone forecasts. Stratospheric ozone further affects the chemical composition in the troposphere because of stratosphere-troposphere transport of ozone (Stevenson et al., 2006, Gaudel et al., 2015), and its radiative properties influencing the tropospheric photolysis rates. Beyond such direct implications on the troposphere a comprehensive

description of stratospheric composition allows a more complete understanding of processes taking place in the stratosphere, ranging from tracking the ozone hole (Lefever et al., 2015) and understanding the concentrations of ozone depleting substances (Chipperfield et al., 2015), to the assessment of dynamical effects such as the Quasi-Biennial Oscillation (QBO, Baldwin et al., 2001), and from implications of sudden stratospheric warmings on circulation patterns (Manney et al., 2015) to general radiative feedbacks of ozone, water ~~vapor~~vapour and CO₂ on weather and climate (Solomon et al., 2010). These aspects have long been studied in the framework of Chemistry Transport Models (CTMs) and, more recently, in GCMs, see, e.g., the SPARC Chemistry-Climate Model Validation Activity (CCMVal, 2010). In GCMs the role of stratospheric ozone chemistry on the tropospheric climate can explicitly be studied (e.g. Scaife et al., 2011). But also meteorological models can benefit from having a good representation of the stratospheric composition and its variability, considering the radiative effects and the resulting impact on stratospheric as well as tropospheric temperatures (Monge-Sanz et al., 2013), which becomes relevant for tropospheric forecast skills on long-range to seasonal time scales (Maycock et al., 2011).

Within a series of MACC (Monitoring Atmospheric Composition and Climate) European research projects a global forecast and assimilation system has been built, which is the core for the global system of the Copernicus Atmosphere Monitoring Service, (CAMS, <http://atmosphere.copernicus.eu>). In CAMS, forecasts of atmospheric composition are carried out (Flemming et al., 2015, Morcrette et al., 2009, Engelen et al. 2009), which benefit from assimilation of satellite retrievals (Inness et al., 2015, Benedetti et al., 2009), to improve the initial conditions for composition fields in terms of reactive gases, aerosols and greenhouse gases. Here a tropospheric chemistry scheme has been embedded in ECMWF's Integrated Forecast System, referred to as Composition-IFS (C-IFS, Flemming et al., 2015). Even though the current operational version of C-IFS based on the Carbon Bond chemistry scheme (CB05) provides good model capability on tropospheric composition (Eskes et al., 2015), the stratosphere is only realistically constrained in terms of ozone. This is because so far the model ozone is based on a linear scheme (Cariolle and Tyssède, 2007) which is suitable owing to the data-assimilation capabilities of C-IFS of both total column and profile satellite retrievals (Flemming et al., 2011; Inness et al., 2015; Lefever et al., 2015).

Also it is recognized that the applicability of radiation feedbacks of ~~tracer fields~~trace gases, such as ozone and water ~~vapor~~vapour, as produced through CH₄ oxidation, are hampered by schemes that are based on linearizations (Cariolle and Morcrette, 2006; de Grandpré et al., 2009), This is due to ~~their~~the intrinsic dependencies to climatologies which are used to construct such schemes and hence they may behave poorly in anomalous situations. Having full stratospheric chemistry available in the IFS therefore would not only allow to study a wider range of atmospheric composition processes, but also a more independent assessment of radiation feedbacks on temperature, hence providing the potential for improvements in stratospheric and tropospheric meteorology. These considerations drive the need for extension of C-IFS with a module for stratospheric chemistry. For this we use the chemistry scheme from the Belgian Assimilation System for Chemical Observations (BASCOE), Errera et al. (2008), which was developed to assimilate satellite observations of stratospheric composition.

BASCOE is based on a Chemistry Transport Model (CTM) of the stratosphere which is used to investigate stratospheric photochemistry (Theys et al., 2010; Muncaster et al., 2012). The assimilation system uses the 4D-VAR algorithm (Talagrand and Courtier, 1987) to produce reanalyses of stratospheric composition (Viscardy et al., 2010) which compare favourably well with similar systems (Geer et al., 2006; Thornton et al., 2009) and facilitate detailed studies of transport processes in the stratosphere (Lahoz et al., 2011). The photochemistry module from the BASCOE-CTM was implemented into the Canadian assimilation system GEM, demonstrating the potential of a coupled chemical-dynamical assimilation system for stratospheric studies (de Grandpré et al., 2009; Robichaud et al., 2010). BASCOE has been used and evaluated within the framework of MACC as an independent system for the provision of Near Real-Time analyses of stratospheric ozone and for the validation of the corresponding product by the main assimilation system (Lefever et al., 2015; Eskes et al., 2015).

~~We have developed a strategy for merging~~The CB05 tropospheric scheme has been combined with the stratospheric scheme from BASCOE-CTM to form a single chemistry mechanism that encompasses tropospheric and stratospheric chemistry throughout the atmosphere, here referred to as C-IFS-Atmos. However, this approach appears computationally expensive, due to the extended chemical mechanism. Therefore we have developed an approach for an optimized merging of the CB05 tropospheric chemistry scheme and the stratospheric chemistry scheme used in BASCOE within C-IFS. An assessment of the two chemistry schemes showed that there is only partial overlap in ~~tracers~~trace gases and reactions that are essential in both regimes. For instance, 15 out of the full list of 99 ~~tracers~~trace gases need to be treated in the chemical mechanisms for both troposphere and stratosphere. Also the modelling of the photolysis rates and heterogeneous reactions have been optimized for application in troposphere and stratosphere separately. ~~Therefore we did not aim at a full integration of the chemistry schemes, but rather choose~~In this optimized approach we developed a flexible setup where -within a single framework- either the tropospheric or stratospheric chemistry modules are addressed, referred to as C-IFS-TS. In this approach the parameterizations for the chemistry, including the respective chemistry mechanisms as optimized for troposphere and stratosphere separately, are retained.

In this paper we describe ~~our modeling strategy~~two merging approaches and provide a benchmark ~~evaluation~~evaluations of the ~~merged~~C-IFS-Atmos and C-IFS-TS ~~systems~~systems with focus on the stratospheric composition. The ancestor BASCOE-CTM is also included in the comparison through a forward model run (without chemical data assimilation), in order to provide insight in the differences caused by the treatment of transport between C-IFS and BASCOE. The paper is organized as follows: In Sect 2 the chemistry modules for the stratosphere are described and the merging with the tropospheric scheme is explained.. Section 3 provides details on the setup of the model runs, and the observational data used for the model evaluation. Section 4 provides a basic model evaluation of the system. We finalize this manuscript with conclusions and an outlook for further work.

2. Atmospheric chemistry in C-IFS

For general aspects related to chemistry modeling in ~~the~~ C-IFS the reader is referred to Flemming et al. (2015). The meteorological model in the current version of C-IFS is based on IFS cycle 41r1 (<http://www.ecmwf.int/en/forecasts/documentation-and-support/changes-ecmwf-model>), ECMWF, 2015). The advection is simulated with a three-dimensional semi-Lagrangian advection scheme, which applies a quasi-monotonic cubic interpolation of the departure values.

In the following two subsections we describe the C-IFS modules for the stratospheric (~~referred to as~~ BASCOE-based) and tropospheric (CB05-based) chemistry parameterizations, continued by a section describing the merging procedure of these two modules to form the C-IFS-TS system. The full list of trace gases is given in Table A1 in the Appendix, including the domains where they are actively treated within the chemistry.

2.1 Stratospheric chemistry

From the BASCOE system (Errera et al., 2008) the chemical scheme and the parameterization for Polar Stratospheric Clouds (PSC) has been implemented in ~~the~~ C-IFS. The BASCOE chemical scheme used here is labelled “sb14a”. It includes 58 species interacting through 142 gas-phase, 9 heterogeneous and 52 photolytic reactions. This chemical scheme merges the reaction lists developed by Errera and Fonteyn (2001) to produce short-term analyses, with the list included in the SOCRATES 2-D model for long-term studies of the middle atmosphere (Brosseur et al., 2000; Chabrilat and Fonteyn, 2003). The resulting list of species (see Table A1) includes all the ozone-depleting substances and greenhouse gases necessary for multi-decadal simulations of the couplings between dynamics and chemistry in the stratosphere, as well as the reservoir and short-lived species necessary for a complete description of stratospheric ozone photochemistry.

Gas-phase and heterogeneous reaction rates are taken from JPL evaluation 17 (Sander et al., 2011) and JPL evaluation 13 (Sander et al., 2000), respectively. Lookup tables of photolysis rates were computed offline by the TUV package (Madronich and Flocke, 1999) as a function of log-pressure altitude, ozone overhead column and solar zenith angle. The photolysis tables used in chemical scheme sb14a are based on absorption cross-sections from JPL evaluation 15 (Sander et al., 2006). The kinetic rates for heterogeneous chemistry are determined by the parameterization of Fonteyn and Larsen (1996), using classical expressions for the uptake coefficients on sulfate aerosols (Hanson and Ravishankara, 1994) and on Polar Stratospheric Clouds (PSCs) (Sander et al., 2000).

The surface area density of stratospheric aerosols uses ~~the same an aerosol number density climatology as Daerden based on SAGE-II observations (Hitchman et al. (2007), while the surface area densities of PSCs is computed from a simple cold-point parameterization., 1994).~~ Ice PSCs are presumed to exist at any grid point in the winter/spring polar regions where ~~the temperature is colder than 186 K, water vapour partial pressure exceeds the vapour pressure of water ice (Murphy and Koop, 2005).~~

Nitric Acid Tri-hydrate (NAT) PSCs ~~where the temperature is colder than 194 K~~ are assumed when the nitric acid (HNO_3) partial pressure exceeds the vapour pressure of condensed HNO_3 at the surface of NAT PSC particles (Hanson and Mauersberger, 1988). The surface area density is set to $2 \times 10^{-6} \text{ cm}^2/\text{cm}^3$ for ice PSCs and $2 \times 10^{-7} \text{ cm}^2/\text{cm}^3$ for NAT PSCs. The sedimentation of PSC particles causes denitrification and dehydration. This process is approximated by an exponential decay of HNO_3 with a characteristic time-scale of ~~10020~~ days for gridpoints where NAT particles are supposed to exist, and an exponential decay of HNO_3 and H_2O with a characteristic time-scale of 9 days for gridpoints where ice particles are supposed to exist.

Mass mixing ratios for N_2O , CO_2 and a selection of anthropogenic and organic halogenic trace gases are constrained at the surface by a global mean constant value, Table 1. Assuming that trace gases are well mixed in the troposphere, this essentially serves as lower boundary conditions for the stratospheric chemistry.

2.2 Tropospheric chemistry

The tropospheric chemistry in the C-IFS is based on the CB05 mechanism (Yarwood et al., 2005). It adopts a lumping approach for organic species by defining a separate tracer species for specific types of functional groups. The scheme has been modified and extended to include an explicit treatment of C1 to C3 species as described in Williams et al., (2013), and SO_2 , di-methyl sulphide (DMS), methyl sulphonic acid (MSA) and ammonia (NH_3) (Huijnen et al., 2010). A coupling to the MACC aerosol model is available (Huijnen et al., 2014), but not switched on for this study. The reaction rates follow the recommendations given in either Sander et al. (2011) or Atkinson et al. (2006). The modified band approach (MBA), which is adopted for the computation of photolysis rates (Williams et al., 2012), uses 7 absorption bands across the spectral range 202 – 695 nm. At instances of large solar zenith angles ($71\text{--}85^\circ$) a different set of band intervals is used. In the MBA the radiative transfer calculation using the absorption and scattering components introduced by gases, aerosols and clouds is computed on-line for each of the predefined band intervals. The complete chemical mechanism as applied for the troposphere is extensively documented in Flemming et al. (2015). ~~A~~ There a specification of the emissions and deposition of tropospheric reactive trace gases is provided as well.

2.3 Merging ~~procedure~~procedures for the tropospheric and stratospheric chemistry

~~In this section we describe the strategy for merging~~ Here we investigate two options to merge tropospheric and stratospheric chemistry, as also summarized in Table 2. The chemistry mechanism for C-IFS-Atmos is composed by simply combining the reaction mechanisms for troposphere and stratosphere into one large mechanism, removing reactions that are duplicated. In contrast to this model version here we propose an approach for a more efficient merging of the chemistry modules for the troposphere and stratosphere to form the C-IFS-TS system. Key chemical cycles differ between troposphere and stratosphere, hence ~~requiring~~allowing different ~~parameterizations~~chemical mechanisms. For example, the oxidation of non-methane hydrocarbons (NMHC's) is essentially taking place in the troposphere and represents an important driver for tropospheric O_3 production. The chemical evolution of PAN and organic nitrate can be neglected in the stratosphere. On the

other hand, N₂O and CFC's are essentially chemically inactive in the troposphere and will only ~~photolyse~~be photolysed by UV radiation in the stratosphere. Therefore, ~~only the transport of those trace chemical reactions involving these~~ gases ~~needs do not need~~ to be accounted for in the troposphere. ~~Associated chemistry involving single atom radicals, such as N, O, Br, Cl, can only be produced in the stratosphere.~~ Also the parameterization of the photolysis rates leads to different

5 requirements for the troposphere and stratosphere, as will be discussed in the next subsection. Finally the numerical solver of the chemical mechanism contributes substantially to the total costs of the model run in terms of run-time, depending on the size of the reaction mechanism. These elements have motivated us to divide the chemistry in the C-IFS-TS system into a tropospheric and stratospheric part. Note that there is only one set of transported atmospheric trace gases and only the position of the grid box above or below the tropopause determines if the tropospheric or stratospheric chemistry is applied.

10 The tropopause can be defined based on a different criteria. A common approach is to use dynamical criterion such as the isentropic potential vorticity (e.g., Thuburn and Craig, 1997) but this fails in regions of small absolute vorticity, notably in the tropics. A definition based on the lapse rate (WMO, 1957) is an alternative, but may not be well defined in the presence of multiple stable layers. We therefore choose to base our criterion on the chemical composition of the atmosphere considering that the tropopause is associated with sharp gradients in trace gases (e.g., Gaudel et al., 2015). This has the

15 advantage that parcels with tropospheric/stratospheric composition can be traced dynamically, and the most appropriate chemistry scheme can be adopted to it. In our simulation we use a chemical definition of the tropopause level, where tropospheric grid cells are defined at O₃<200 ppb and CO>40 ppb, for P > 40 hPa. With this definition the associated tropopause pressure ranges in practice between approx. 270 and 80 hPa for sub-tropics and tropics, respectively.

For both troposphere (CB05) and stratosphere (BASCOE) the numerical solver is generated using the Kinetic Pre-Processor (KPP, Sandu and Sander, 2006) software. Specifically~~Specifically~~ we adopt the standard four-stages, third order Rosenbrock solver (Rodas3). This is different from the ~~hard-coded~~ Eulerian backward implicit solver as used in Flemming et al. (2015), and is motivated by the improved coding flexibility and accuracy.

Most of the gas phase reactions that take place both in the troposphere and stratosphere, such as NO_x and HO_x reactions, are simulated in identical ways in both chemistry schemes. It is worth mentioning that the ~~tracers~~constituents O¹D and O³P, produced from O₃ and O₂ photolysis, are ~~described implicitly not explicitly computed~~ in the troposphere, ~~while they as O¹D and O³P are treated explicitly in~~ assumed to react with O₂, O₃ and N₂ only. This is different for the stratosphere, where O¹D and O³P are involved in many reactions. For trace gases whose chemistry is currently neglected in the stratosphere (the NMHC's, PAN, Organic nitrate, SO₂) we adopt a 10-day decay rate to prevent ~~their~~ spurious accumulation ~~of these tracers~~ in the stratosphere. Hence these losses are currently not accounted for in the stratospheric chemical mechanisms and do not

25 contribute either to the load of stratospheric aerosols. Note that tropospheric halogen chemistry, which contributes to near-surface ozone depletion in spring-time polar region and to changes in oxidative capacity in the tropical marine boundary layer (von Glasow and Crutzen, 2007) is currently neglected, even though related trace gases are available. By inspection of individual ~~tracer~~constituents fields we have ensured that the merging strategy does not result in spurious jumps at the interface between troposphere and stratosphere. In case of running, see also Supplementary Figures S2-S5. When the system

30

is run with stratospheric chemistry only (C-IFS-S), all chemistry and emissions are switched off at altitudes below 400 hPa and ~~replaced~~constrained by surface boundary conditions.

The ~~three~~four options to run this type of C-IFS experiments and the computational ~~cost~~costs are given in Table 2. As compared to the C-IFS-T experiments, the costs of running an experiment including full stratospheric chemistry with the C-IFS-TS system have increased by ~50%. ~~The additional burden for~~Most of this increase is caused by the computation of the chemistry and not the tracer transport due to the ~~increase in the number of tracers only marginally increases the computational time, because of the~~ efficiency of the semi-Lagrangian advection scheme for multiple tracers. ~~A test experiment~~The C-IFS-Atmos setup where tropospheric and stratospheric chemistry were merged into a single reaction mechanism, ~~where all reactions are activated in the whole atmosphere~~, led to an increase in costs by ~50% compared to C-IFS-TS, indicating the benefit of having separate solver codes for tropospheric and stratospheric chemistry. ~~Finally this also allows for an easy switch between system setups.~~The C-IFS-TS implementation allows for an easy switch between system setups compared to the C-IFS-Atmos implementation. For completeness also specifications of the BASCOE-CTM are provided in Table 2, which is identical in terms of stratospheric chemistry parameterization compared to C-IFS-TS and C-IFS-S. Clearly the essential difference compared to the C-IFS setup refers to the fact that BASCOE is used here as a CTM, while C-IFS is a GCM. Most notably this implies a different advection treatment and a different horizontal grid (see section 3).

2.3.1 Merging photolysis rates

For parameterization of the photolysis rates the Modified Band Approach (MBA, Williams et al., 2012) and the lookup table approach (Errera and Fonteyn, 2001) ~~as are retained, see Table 3, as these~~ have been optimized in the past for applications in the troposphere and stratosphere ~~are retained, see Table 3, respectively~~. While for tropospheric conditions scattering and absorption properties of clouds and aerosol strongly impact the magnitude of photolysis rates and hence the local chemical composition, this is of less relevance in the stratosphere. Wavelengths shorter than 202 nm, on the other hand, are largely blocked by stratospheric ozone and oxygen and do not contribute to radiation in the troposphere (Williams et al., 2012). At higher altitudes these short wavelengths contribute to the Chapman cycle and to the break down of CH₄, CFC's and N₂O either directly or through oxidation by O¹D. Also the presence of sunlight at solar zenith angles (SZA) larger than 90° at high altitudes needs to be accounted for in the stratosphere ~~but not necessarily in the troposphere. Solar radiation reaches the stratosphere earlier than the Earth's surface, due to the Earth's curvature which, amongst others, triggers the polar spring stratospheric. This plays a role in the timing of springtime~~ ozone depletion. ~~in the polar lower stratosphere, but may be neglected in the troposphere.~~

Table 4 lists the photolysis rates that are active both in the troposphere and stratosphere. Photolysis rates for reactions occurring both in the troposphere and stratosphere are merged at the interface, in order to ensure a smooth transition between the two schemes. This is done by an interpolation at four model levels around the interface level between both

parameterizations, for $SZA < 85^\circ$. For larger SZA the original value for the photolysis rate is retained in case of stratospheric chemistry, while it is switched off for the troposphere.

Note that even though the reaction rates have been merged, the products from the same photolytic reactions are sometimes different as a consequence of the different reaction mechanisms between the troposphere and stratosphere.

- 5 An example of the merging strategy is given in Fig. 1. It shows that at the interface for $J O_3$ and $J NO_2$ on average a small increase of the merged photolysis rate is seen towards lower altitudes, with the switch to MBA in the troposphere, which is a consequence of the combination of differences in the parameterizations. 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~~ **Tracer transport settings**

15 Tracer transport is treated identically for all individual chemical ~~tracers~~ trace gases. Since the semi-Lagrangian advection does not formally conserve mass (Flemming and Huijnen, 2011; de Grandpré et al., 2016) a global mass fixer is applied (Diamantakis and Flemming, 2014) to all but a few ~~tracers~~ constituents, including NO, NO₂ and H₂O. Rather than conserving mass during the advection step of the individual components NO and NO₂, this is enforced to a stratospheric NO_x tracer defined as the sum of NO and NO₂. While a chemical H₂O ~~tracer~~ trace gas is defined in the full atmosphere, in the troposphere H₂O mass mixing ratios are constrained by the humidity (q) simulated in the meteorological model in IFS and provide a boundary condition for water vapour in the IFS stratosphere. Stratospheric H₂O (i.e. above the tropopause level) is governed by chemical production and loss. ~~Stratospheric H₂O mass is not strictly conserved considering that the~~ The global advection errors in H₂O that essentially originate from the tropospheric part (where because by far most H₂O mass is located with large in the troposphere and the spatial gradients), and are much more pronounced. This should not affect the stratospheric H₂O mass budget (where total, herefore the global mass fixer for the stratospheric H₂O tracer has been switched off. This prevents spurious trends in stratospheric H₂O columns over the years (not shown), indicating that H₂O mass is much lower and H₂O mixing ratio gradients are much smoother), conservation is well ensured in the stratosphere.

3. Model setup and observations used

25 We have executed ~~a run of~~ runs with C-IFS-TS and C-IFS-Atmos for the period April 2008 until ~~December 2009~~ December 2010. Stratospheric ozone in C-IFS-TS is further compared to that of the C-IFS-T system (Flemming et al., 2015) ~~which~~. This uses the ECMWF standard linear ozone scheme (version 2a, Cariolle and Teyssède, 2007) in the stratosphere, while stratospheric HNO₃ is constrained through a climatological ratio of HNO₃/O₃ at 10 hPa (Flemming et al., 2015).

We have initialized ~~all C-IFS-TS and C-IFS-T~~ runs on 1 April 2008 using assimilated concentration fields from the BASCOE system in the stratosphere for this date. The horizontal resolution of these runs is T255 (i.e. approx. 0.7° lon / lat) with 60 levels in the vertical. Meteorology in the C-IFS runs is relaxed towards ERA-Interim.

Intercomparison of the runs C-IFS-TS and C-IFS-Atmos aims to provide a justification of our approach to split the chemistry into two regions, while intercomparison of C-IFS-TS with C-IFS-T can be used to identify the changes to stratospheric composition modelling between full stratospheric chemistry and the baseline approach with the linear ozone scheme.

The performance of the C-IFS-TS runs has further been compared against the BASCOE-CTM (without chemical data assimilation), using the same chemical mechanism and parameterizations for photolysis and heterogeneous chemistry as implemented in the C-IFS. ~~The BASCOE-CTM is -TS. It uses~~ This serves as a model reference for the C-IFS implementation of stratospheric chemistry. While C-IFS evaluates tracer transport on a reduced Gaussian grid, the BASCOE-CTM uses a regular latitude-longitude grid. It is run here with a resolution of 1.0125° lon / lat similar to the resolution ~~of~~ chosen for C-IFS used ~~here~~, and on the same vertical grid of 60 levels. ~~It uses~~ The BASCOE-CTM is driven by temperature, pressure and wind fields simulated by the C-IFS runs. Using ~~However, while BASCOE adopts a flux-form advection scheme (Lin and Rood, 1996) the IFS uses the Semi-Lagrangian scheme for advection, accounts for vertical diffusion and includes a parameterization for convection (ECMWF, 2015). Using essentially~~ the same dynamical fields together with an identical implementation of the chemistry code should allow to identify differences ~~in~~ due to the different transport schemes between C-IFS and the BASCOE-CTM. Common chemical biases between both systems also point at issues in the chemical ~~parameterization~~ parameterizations such as reaction mechanism, photolysis, heterogeneous chemistry and sedimentation.

3.1 Observational data used for validation

We evaluate the C-IFS-TS runs in terms of stratospheric O₃, NO₂, N₂O, CH₄, H₂O and HCl, and for this purpose use a range of observation-based products.

Model results are compared with retrievals (version 3) of O₃ (Froidevaux et al., 2008a), ClO (Santee et al., 2008), H₂O (Read et al., 2007) and HCl (Froidevaux et al., 2008b) from the Microwave Limb Sounder (MLS) onboard the satellite Aura and with retrievals (version 6) of O₃ (Ceccherini et al., 2008), HNO₃ (Wang et al., 2007) and NO₂ (Wetzel et al., 2007) from limb emission spectra recorded by the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) onboard the European satellite Envisat.

The MLS error budget is reported in Livesey et al. (2011). For HCl observations between 1-20 hPa the precision and accuracy are below 10 and 15% respectively. Between 46 and 100 hPa, these are below 0.3 and 0.2 ppbv, respectively. For H₂O between 0.46 and 100 hPa, precision and accuracy are below 15 and 8%. MIPAS random and systematic errors for various trace gases are reported by Raspollini et al. (2013). For NO₂ between 25 and 50 km altitude these are below 10 and 20% respectively. For HNO₃ between 15 and 30 km, these are below 8 and 15% while for O₃ between 20 and 55 these are below 5 and 10%. At 15 km, these errors increase to 10 and 20%, respectively.

Total column O₃ is validated against KNMI's multi sensor reanalysis version 2 (MSR, van der A et al., 2015) which, for the 2008-20092010 time period is based on Solar Backscattering Ultraviolet radiometer (SBUV/2), Global Ozone Monitoring Experiment (GOME), SCanning Imaging Absorption spectroMeter for Atmospheric CartographY (SCIAMACHY) and Ozone Monitoring Instrument (OMI) observations. The satellite retrieval products used in the MSR are bias-corrected with respect to Brewer and Dobson Spectrophotometers to remove discrepancies between the different satellite data sets. The uncertainty in the product, as quantified by the bias of the observation-minus-analysis statistics, is in general less than 1 DU. O₃ profiles are compared to ozonesonde data that are acquired from the World Ozone and Ultraviolet radiation Data Centre (WOUDC). The precision of the ozonesondes is on the order of 5% in the stratosphere (Hassler et al., 2015), when based on electrochemical concentration cell (ECC) devices (~85% of all soundings). Larger random errors (5-10%) are found for other sonde types, and in the presence of steep gradients and where the ozone amount is low. Sondes at 19, 12, 2 and 1 individual stations are used for the evaluation over northern hemisphere midlatitudes, tropics, southern hemisphere midlatitudes and Antarctic, respectively.

Stratospheric NO₂ columns are compared to observational data from the SCIAMACHY (Bovensmann et al., 1999) UV-VIS (ultraviolet-visible) and NIR (near-infrared) sensor onboard the Envisat satellite. The satellite retrievals are based on applying the Differential Optical Absorption Spectroscopy (DOAS) (Platt and Stutz, 2008) method to a 425-450 nm wavelength window. Stratospheric NO₂ columns from SCIAMACHY presented here are in fact total columns derived using by dividing retrieved slant columns of NO₂ by a stratospheric air mass factor (Richter et al., 2005). To minimize the impact of the troposphere, only and contains data over the clean Pacific region are used ocean (180°E - 220°E). Still, the amount considered here as being only (Richter et al., 2005). Although in this region the contribution of the troposphere to total column NO₂ is small, stratospheric includes column NO₂ from SCIAMACHY is still somewhat positively biased by a weighted part of the tropospheric background NO₂-contribution. However, stratospheric air mass factors for NO₂ 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.

Monthly mean stratospheric NO₂ columns are associated with relative uncertainties of roughly 5-10% and an additional absolute uncertainty of 1×10^{14} molec cm⁻². To account for differences in observation and model output time, simulations are interpolated linearly to the equator crossing time of SCIAMACHY (10:00 LT). In addition, only model data for which satellite observations exist are included in the corresponding comparisons.

Furthermore, satellite-based observations are used from the Atmospheric Chemistry Experiment - Fourier Transform Spectrometer (ACE-FTS), onboard of the Canadian satellite mission SCISAT-1 (first Science Satellite, Bernath et al., 2005). This is a high spectral resolution Fourier transform spectrometer operating with a Michelson interferometer. Vertical profiles of atmospheric volume mixing ratios of trace constituents are retrieved from the occultation spectra, as described in Boone et al. (2005), with a vertical resolution of 3-4 km at maximum. Here we use level 2 retrievals (version 3.0) of N₂O and CH₄.

ACE-FTS N₂O observations between 6-~~and~~-30 km are agree to within ±15% compared against of independent observations, while above they agree to within ±4 ppbv (Strong et al., 2008). The uncertainty in ACE-FTS CH₄ observations is within 10%

in the upper troposphere – lower stratosphere, and within 25% in the middle and higher stratosphere up to the lower mesosphere (<60 km) (De Mazière et al. 2008).

~~Model results are also compared with observations. Three-hourly C-IFS and BASCOE-CTM output has been interpolated in space and time to match with any of these observations.~~

~~of O_3 (Ceccherini et al., 2008), HNO_2 (Wang et al., 2007) and NO_2 (Wetzel et al., 2007) retrieved from limb emission spectra recorded by the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) onboard the European satellite Envisat, and with observations of H_2O (Read et al., 2007) and HCl (Froidevaux et al., 2008) retrieved from the Microwave Limb Sounder (MLS) onboard the satellite Aura.~~

~~MIPAS random and systematic errors for various trace gases are reported by Raspollini et al. (2013). For NO_2 between 25 and 50 km altitude these are below 10 and 20% respectively. For HNO_2 between 15 and 30 km, these are below 8 and 15% while for O_3 between 20 and 55 these are below 5 and 10%. At 15 km, these errors increase to 10 and 20%, respectively. The MLS error budget is reported in Livesey et al. (2011). For HCl observations between 1–20 hPa the precision and accuracy are below 10 and 15% respectively. Between 46 and 100 hPa, these are below 0.3 and 0.2 ppbv, respectively. For H_2O between 0.46 and 100 hPa, precision and accuracy are below 15 and 8%.~~

4. Model evaluation

~~Fig. 2 shows the zonal mean O_3 total columns against the MSR at various latitude bands. It shows that for the extra-tropical mid-latitudes the positive and negative biases remain below 20 DU (6%), while for the tropics the bias increases towards 18 DU (8%) at the end of the model simulation. Over Antarctica (70S–90S) the zonal, monthly mean average bias is generally less than 20 DU, except for the ozone hole period when the minimum ozone is underestimated by up to 35 DU (25%). In contrast, the Cariolle scheme shows an over-estimation of O_3 column outside the ozone hole period, and a relatively appropriate magnitude of the ozone minimum. While over the northern hemisphere C-IFS-TS shows a clear improvement compared to C-IFS-T with Cariolle, for the tropical and southern hemisphere both versions show a similar performance. Fig. 2 shows the mean O_3 partial columns (PC) against observations from Aura MLS v3.0 over the poles and the tropics. In C-IFS-T, applying the Cariolle parameterization, the annual cycle over the Arctic is very well simulated but a constant overestimation of 50 DU (20%) is found. In the Tropics the bias is much smaller, with a slight underestimation (10 DU, 5%). In the Antarctic, the results are remarkably good during the ozone hole episodes but there is a serious overestimation developing from February until the beginning of August, when it reaches 50 DU (30%) i.e. as much as in the Arctic. CIFS-Atmos and CIFS-TS provide very similar results over the full time period, suggesting that our approach to keep two different solvers in each region is valid for stratospheric ozone. Also after an initialization period of a few months the model runs do not present any obvious drift, and the differences with BASCOE-CTM are very small. This implies that differences due to the model configuration regarding transport are not crucial for lower stratospheric ozone at these timescales. In the Tropics the C-IFS-TS and C-IFS-Atmos results are slightly better than those with BASCOE-CTM, potentially due to the missing~~

parameterization for convection. In the Antarctic, the parameterization of PSC leads to an overestimation of springtime ozone depletion while the Cariolle parameterization simulates very well the lowest columnar values observed in September, as discussed in more detail below. The recovery of ozone is overestimated by 20DU (10%) in December-January. While the amplitude of the annual cycle is overestimated above the Antarctic, its structure matches well the observations.

5 An evaluation of O₃ total columns (TC) against the MSR at various latitude bands is given in Figure S6 in the Supplementary material. Considering the missing tropospheric chemistry in the BASCOE-CTM this system is not well constrained in terms of the O₃ TC which implies that it is not useful to include its results here. The TC comparison confirms the evaluation with PC from Aura MLS observations, showing a strong positive bias over the NH mid latitudes and Arctic for C-IFS-T, which is reduced for C-IFS-Atmos and C-IFS-TS. These model versions do not show a significant trend during
10 the 2009 – 2010 period. For the tropical and southern hemisphere mid-latitudes all C-IFS versions show a similar performance with C-IFS-Atmos showing a small positive offset with respect to C-IFS-TS of approx. 2-8 DU depending on the latitude band and season.

15 Closer inspection of O₃ profiles against sondes averaged over the NH-mid latitudes, tropics and SH-mid latitudes for the DJF and JJA seasons (Figures 3 and 4) also shows reduced biases most prominently visible at the 10-30 hPa altitude range in the sub-tropics for C-IFS-TS. Nevertheless, this experiment still shows in 2009 and 2010 (Figures 3 and 4) shows biases in generally similar order of magnitude, although frequently with opposite sign, for C-IFS-TS and C-IFS-Atmos compared to C-IFS-T. Especially over the extra-tropics the C-IFS-TS and C-IFS-Atmos model versions show lower mixing ratios than C-IFS-T at the middle stratosphere (~10 hPa), generally leading to an improvement compared to the observations. For the NH mid-latitudes this also explains the improved O₃ TC and O₃ PC in these runs compared to C-IFS-T as discussed above.
20 Nevertheless, these experiments still show a positive bias near the ozone maximum in terms of partial pressure (~50 hPa) and also at lower altitudes during the northern hemispheric spring season. Furthermore, in the tropics the use of the full stratospheric chemistry implies a slight degradation against the linear scheme around the ozone maximum, where the Cariolle parameterization is very well tuned while the The negative bias in the lower stratosphere, as also found in C-IFS-TS is not improved.

25 compared to C-IFS-T. These alternating biases in CIFS-TS and C-IFS-Atmos are due to corresponding biases in chemically related species such as NO_x and also to transport issues, as discussed in more detail below. The very similar performance of C-IFS-TS with respect to C-IFS-Atmos, especially in this altitude range, once again gives confidence in our approach to split chemistry scheme for tropospheric or stratospheric conditions. A similar evaluation against MLS observations, but for the period September-October-November 2009, provides very similar conclusions (Figure S7, supplementary material). For the
30 2009 Antarctic ozone hole season (Fig. 5) the C-IFS-TS shows and C-IFS-Atmos show a positive bias at ~100 hPa for August and September, but and negative bias at higher altitudes (50-10 hPa), where C-IFS-T shows a positive bias. Still the depth of the ozone hole is well modelled in October. During the closure phase in November and December the O₃ variability with altitude is better captured in C-IFS-TS than in C-IFS-T. The evaluation of the zonal mean ozone concentrations against

MIPAS observations shows good general agreement, Fig. 6, with small biases of similar magnitude as the ones for the BASCOE-CTM simulation.

A closer analysis of the processes responsible for springtime polar ozone depletion is given in Fig. 6. In both the C-IFS-TS and C-IFS-Atmos runs as well as BASCOE-CTM there is an HNO_3 deficit at the beginning of the winter. Denitrification, which is not modelled in C-IFS-T, starts at the correct time in the models with stratospheric chemistry indicating that NAT PSC appear at about the right time. However, denitrification proceeds more slowly and ends one month later than observed by Aura-MLS. We attribute this shortcoming to the crude modelling of NAT PSC which does not calculate the amount of condensed nitric acid and water, keeps the surface area densities of PSC particles fixed at an arbitrary value and parameterizes sedimentation through irreversible removal. Chlorine activation starts at exactly the right time and is as strong in the C-IFS runs as in the Aura-MLS observations until the beginning of September, but starts decreasing afterwards while it lasts two more weeks in the observations. Hence the overestimation of ozone depletion during August and September in the models with explicit stratospheric chemistry is probably not due to an overestimation of chemical removal. This feature is more pronounced in CIFS-TS and CIFS-Atmos than in the BASCOE-CTM, suggesting that it may be associated to differences in the modelling of transport.

The evaluation of the zonal mean ozone mixing ratios against MIPAS observations shows good general agreement, Fig. 7, with all four modelling experiments providing similar features. The tropical maximum of O_3 mixing ratio at 10 hPa is underestimated in all experiments but to a larger extent in those which model stratospheric photochemistry explicitly (BASCOE CTM, C-IFS-TS, C-IFS-Atmos) than in C-IFS-T, in line with the evaluation against O_3 sondes for June-July-August (figure 4). The same evaluation against MLS observations provides exactly the same conclusions (Figure S8, supplementary material).

The assessment of NO_2 against MIPAS daytime NO_2 observations, acquired by sampling the ascending orbits from Envisat, shows good agreement with both models. Also the C-IFS-TS describes the models, although C-IFS-TS and C-IFS-Atmos tend to show a positive bias. The C-IFS-TS and C-IFS-Atmos runs describe well the seasonal variation in zonal mean stratospheric NO_2 columns at different latitude bands, Fig. 78, with monthly mean biases with respect to the SCIAMACHY observations of less than $\pm 0.5 \times 10^{15} \text{ molec cm}^{-2}$ in the tropics and at mid-latitudes. The positive bias is larger in C-IFS-Atmos than C-IFS-TS. In contrast, poor performance can be seen for C-IFS-T, due to the lack of stratospheric NO_x chemistry in that version.

However, a positive NO_2 bias with respect to night-time MIPAS NO_2 observations appears larger for C-IFS-TS and C-IFS-Atmos than for the BASCOE-CTM (Fig. 67). In contrast, this figure also shows a negative bias in HNO_3 with respect to MIPAS observations in both the BASCOE-CTM, and C-IFS-TS, again Atmos, and even more marked in the C-IFS-TS experiment. Considering that daytime NO_2 bias in Even though a clear improvement compared to run C-IFS-TST is small and similar to that for BASCOE-CTM, found, further investigation is necessary to diagnose the larger negative bias in C-IFS HNO_3 is likely not caused by origins of the biases in its chemical precursors, night-time NO_2 above 10 hPa and in HNO_3 between 10 and 70 hPa.

Fig. 89 shows an evaluation of N₂O and CH₄ 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 ~~(vertical) 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, N₂O is the main source of reactive nitrogen in the stratosphere while CH₄ is one of the main precursors for stratospheric water vapour. The figure suggests reasonable profile shapes for both CH₄ and N₂O in the upper stratosphere (10 hPa and higher), ~~which is also rather similar as found in the BASCOE-CTM control run. Even though the absolute difference between C-IFS N₂O and observations from MIPAS and MLS is somewhat different in absolute terms than found for the evaluation against ACE-FTS, the general features are very similar~~ 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. ~~At lower altitudes (100-10 hPa) C-IFS-TS N₂O and CH₄ shows larger discrepancies to the observations, and to the BASCOE-CTM run with an over-estimation most prominently around 30 hPa in the tropics and SH-mid latitudes, suggesting too much vertical transport within the middle and lower stratosphere. This feature could also contribute to the positive biases seen in O₂ at ~20 hPa in Figures 3 and 4.~~

~~This discrepancy cannot be due to different wind fields because the BASCOE-CTM experiment is driven by three-hourly output of the C-IFS 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.~~

Fig. 910 shows a good consistency between H₂O modelled by C-IFS-TS and the BASCOE-CTM results, albeit with a slight negative bias with respect to MLS observations above 5 hPa, and a positive bias around 30 hPa in the tropics, associated with corresponding biases in CH₄. This figure also shows globally a good agreement between HCl modelled by C-IFS-TS and MLS observations, although with a positive bias of 0.8 ppbv confined in the region of ozone depletion above Antarctica.

5. Conclusions

We have presented a model description and benchmark evaluation of an extension of the C-IFS system with stratospheric ozone chemistry of the BASCOE model added to the already existing tropospheric scheme CB05, ~~referred. We refer to this~~ system as C-IFS-CB05-BASCOE, or C-IFS-TS in short. In our approach we have retained a separate treatment for tropospheric and stratospheric chemistry, and select the most appropriate scheme depending on the altitude with respect to the tropopause level. This has the advantage that ~~parameterizations~~ mechanisms which are optimized for tropospheric and

stratospheric chemistry, respectively, can be retained, which also substantially reduces the computational costs of the chemical solver compared to an approach where all reactions are activated in the whole atmosphere, referred to as C-IFS-Atmos. Also, it allows for an easy switch between system setups. To avoid jumps in ~~tracer~~trace gas concentrations at the interface the consistency in gas-phase reaction rates has been verified while the photolysis rates from the two parameterizations are interpolated across the interface. We showed that differences between C-IFS-TS and C-IFS-Atmos are overall small, hence our basic assumption to have different chemistry solvers for troposphere and stratosphere is valid for our applications.

An evaluation of a 12.5 year simulation of C-IFS-TS indicates good performance of the system in terms of stratospheric ozone, of similar quality as its ancestor BASCOE-CTM model results, and a considerable general improvement in terms of stratospheric composition compared to the C-IFS-T predecessor model version which applied a linear ozone scheme in the stratosphere.

The O₃ ~~total~~partial columns (10-100 hPa) show biases mostly smaller than ±20 DU when compared to the MSR-v2Aura MLS observations. Also the profiles were generally well captured, and show an improvement with respect to the C-IFS-T linear ozone scheme in the stratosphere over mid-latitudes. The depth and variability of the ozone hole over Antarctica is modelled well. While also the C-IFS-T shows a remarkably good agreement to the observations during the ozone hole episodes it develops a significant overestimation of the partial columns during other months. The tropical maximum of the mixing ratio, around 10 hPa, is the only stratospheric region where C-IFS-T agrees better all-year-long with observations.

Also evaluation of other trace gases (NO₂, HNO₃, CH₄, N₂O, HCl) against observations derived from various satellite retrievals (SCIAMACHY, ACE-FTS, MIPAS, MLS) indicates a good performance. But for CH₄ and N₂O a larger error with respect to limb-sounding retrievals was found at around 30 hPa than the BASCOE-CTM. This could point at too fast vertical transport within the middle and lower stratosphere in the C-IFS framework. illustrate the clear improvements obtained with C-IFS-TS compared to C-IFS-T, even though C-IFS-TS still suffers from positive biases in stratospheric NO₂, whereas HNO₃ is biased low. For the long-lived tracers CH₄ and N₂O, larger errors with respect to limb-sounding retrievals were found between 10 hPa and 100 hPa than with the BASCOE-CTM. The BASCOE-CTM experiment shown here was driven by three-hourly wind fields output of the C-IFS experiments. Hence this discrepancy is due to a difference in the representation of the transport processes between the GCM and the CTM, i.e. the numerical scheme used for advection (Semi-Lagrangian versus Flux-Form), the convection (parameterized in C-IFS but neglected in BASCOE CTM) or the diffusion (parameterized in C-IFS but not explicitly considered in the CTM). Hence, stratospheric transport in C-IFS will be an area for further evaluation and developments.

This benchmark model evaluation of C-IFS-TS marks a first 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 for in the entire atmosphere near future. Future work will/could focus on the following aspects:

- Chemical data-assimilation: initial tests with data-assimilation of O₃ total column and profile retrievals suggest that stratospheric ozone is successfully constrained in C-IFS-TS. However, observational constraints on other components

driving ozone chemistry are currently lacking in the assimilation system. Our extension opens the possibility for assimilation of additional ~~tracers~~trace gases such as N₂O and HCl. However, for the 4D-VAR assimilation of short-lived species such as NO₂ and ClO an adjoint chemistry module would likely be required as implemented in the BASCOE DA system.

5 - Alignment of the reaction mechanism and photolysis rates: while at current stage the gas-phase and photolytic reaction rates of the parent schemes are retained, we foresee a further integration to ensure better alignment of the chemical mechanisms. Especially the existing jumps in photolysis rates as a consequence of the different parameterizations are not desirable, even though they are not harmful for model stability nor visibly lead to any degradation in model performance. The alignment in terms of gas-phase reaction rate expressions can be achieved by the introduction of the KPP solver in C-IFS, for both tropospheric and stratospheric chemistry, which allows for a better traceable model development than the hard-
10 coded Euler Backward Integration solver as adopted in Flemming et al. (2015).

- Improvement of the representation of stratospheric sulphate aerosols and Polar Stratospheric Clouds: the current climatology for these aerosols, and parameterization for PSCs could easily be improved. While the current results are satisfactory for a general-purpose monitoring system, these improvements would especially allow better simulations of the composition in in the polar lower stratosphere during springtime.

15 - Extension of tropospheric and stratospheric chemistry schemes: the availability of a comprehensive set of ~~tracers~~trace gas fields allows for a relatively easy extension of the tropospheric reaction mechanism by including selective reactions originating from the stratospheric chemistry, and vice versa. Examples are the introduction of halogen chemistry in the troposphere (von Glasow and Crutzen, 2007), or SO₂ conversion to sulphate aerosol in the stratosphere, relevant in case of strong volcanic events (Bândă, et al., 2015).

20 - Optimization of solver efficiency: even though the use of KPP has simplified the code maintenance and may result in a higher numerical accuracy of the solution, it also caused a considerable slow-down of the numerical efficiency as compared to the Euler Backward Integration solver, as that solver had been optimized for tropospheric ozone chemistry in C-IFS-CB05. Solutions could be an optimization of the initial chemical time step for the KPP solver, depending on prevailing chemical and physical conditions, and an optimization of the automated solver code, which allows for a more efficient code
25 structure (KP4, Jöckel et al., 2010).

In summary, the extension towards stratospheric chemistry in C-IFS broadens its ability for forecast and assimilation of stratospheric composition, which is beneficial to the monitoring capabilities in CAMS, and may also contribute to advances in meteorological forecasting of the ECMWF IFS model in the future.

Code availability

30 The C-IFS source code is integrated into ECMWF's IFS code, which is available subject to a licence agreement with ECMWF, see also Flemming et al. (2015) for details. The stratospheric chemistry module of C-IFS was originally developed

in the framework of BASCOE. Readers interested in the BASCOE code can contact the developers through <http://bascoe.oma.be>.

Appendix A

Table A1. Trace gases in C-IFS-TS, along with their chemically active domain: troposphere (Trop), stratosphere (Strat) or ~~both (Global~~atmosphere (WA).

Short name	Long name	Active domain
O3	ozone	GB WA
<u>OH</u>	<u>hydroxyl radical</u>	WA
H2O2	Hydrogen <u>hydrogen</u> peroxide	GB WA
HO2	Hydroperoxy <u>hydroperoxy</u> radical	GB WA
OH	Hydroxyl radical	GB
CH4	methane	GB
CO	Carbon <u>carbon</u> monoxide	GB WA
CH2O	formaldehyde	GB WA
CH3O2	Methylperoxy <u>methylperoxy</u> radical	GB WA
CH3OOH	methylperoxide	GB WA
<u>CH4</u>	<u>methane</u>	WA
NO	Nitrogen <u>nitrogen</u> monoxide	GB WA
NO2	Nitrogen <u>nitrogen</u> dioxide	GB WA
NO3	Nitrate <u>nitrate</u> radical	GB WA
HNO3	Nitric <u>nitric</u> acid	GB WA
HO2NO2	Pernitric <u>pernitric</u> acid	GB WA
N2O5	Dinitrogen <u>dinitrogen</u> pentoxide	GB WA
<u>Rn</u>	<u>radon</u>	WA
<u>PARPb</u>	<u>paraffins</u> lead	Trop

C2H4	ethene	Trop
<u>C2H6</u>	<u>ethane</u>	<u>Trop</u>
<u>C2H5OH</u>	<u>ethanol</u>	<u>Trop</u>
<u>C3H8</u>	<u>propane</u>	<u>Trop</u>
<u>C3H6</u>	<u>propene</u>	<u>Trop</u>
<u>C5H8</u>	<u>isoprene</u>	<u>Trop</u>
<u>C10H16</u>	<u>terpenes</u>	<u>Trop</u>
<u>CH3COCHO</u>	<u>methylglyoxal</u>	<u>Trop</u>
<u>CH3COCH3</u>	<u>acetone</u>	<u>Trop</u>
<u>CH3OH</u>	<u>methanol</u>	<u>Trop</u>
<u>HCOOH</u>	<u>formic acid</u>	<u>Trop</u>
<u>MCOOH</u>	<u>methacrylic acid</u>	<u>Trop</u>
<u>PAR</u>	<u>paraffins</u>	<u>Trop</u>
OLE	olefins	Trop
ALD2	aldehydes	Trop
PAN	Peroxyacetyl nitrate	Trop
ROOH	peroxides	Trop
<u>PAN</u>	<u>peroxyacetyl nitrate</u>	<u>Trop</u>
ONIT	Organic <u>organic</u> nitrates	Trop
SO2	Sulfur <u>sulfur</u> dioxide	Trop

SO4	sulfate	Trop
DMS	Dimethyl dimethyl sulfide	Trop
<u>MSA</u>	<u>methanesulfonic acid</u>	<u>Trop</u>
NO3_A	nitrate	Trop
<u>NH2</u>	<u>amine</u>	<u>Trop</u>
NH3	ammonia	Trop
NH4	ammonium	Trop
MSA	Methanesulfonic acid	Trop
CH3COCHO	methylglyoxal	Trop
C2O3	Peroxyacetyl peroxyacetyl radical	Trop
<u>ISPD</u>	<u>methacrolein_MVK</u>	<u>Trop</u>
<u>ACO2</u>	<u>acetone product</u>	<u>Trop</u>
<u>IC3H7O2</u>	<u>IC3H7O2</u>	<u>Trop</u>
<u>HYPROPO2</u>	<u>HYPROPO2</u>	<u>Trop</u>
ROR	Organic ethers	Trop
RXPAR	PAR budget corrector	Trop
XO2	NO to NO2 operator	Trop
XO2N	NO to alkyl nitrate operator	Trop
<u>QCH3OH</u>	<u>methanol oxygen atom (ground state)</u>	Trop Strat
<u>HCOOH01D</u>	<u>Formic acid oxygen atom (first</u>	Strat <u>Trop</u>

<u>MCOOH</u>	Methacrylic acid hydrogen atom	<u>Strat</u> Trop
<u>C2H6</u>	ethane hydrogen	Trop Strat
<u>C2H5OH</u> <u>H2O</u>	ethanol/Water	<u>Strat</u> Trop
<u>C3H8</u>	propane	Trop
<u>C3H6</u>	propene	Trop
<u>C5H8</u>	isoprene	Trop
<u>C10H16</u>	terpenes	Trop
<u>CH3COCH3</u>	acetone	Trop
<u>ISPD</u>	Methacrolein-MVK	Trop
<u>ACO2</u>	Acetone product	Trop
<u>IC3H7O2</u>	<u>IC3H7O2</u>	Trop
<u>HYPROPO2</u>	<u>HYPROPO2</u>	Trop
<u>NH2</u>	amine	Trop
<u>Rn</u>	radon	Glb
<u>Pb</u>	lead	Trop
<u>CH3</u>	<u>Methyl</u> <u>methyl</u> radical	Strat
<u>CH3O</u>	<u>Methoxy</u> <u>methoxy</u> radical	Strat
<u>HCO</u>	<u>Formyl</u> <u>formyl</u> radical	Strat
<u>CO2</u>	<u>carbondioxide</u>	<u>Strat</u>
<u>N</u>	<u>nitrogen atom</u>	<u>Strat</u>

N2O	Nitrous <u>nitrous</u> oxide	Strat
H2O <u>CL</u>	water <u>chlorine atom</u>	Strat
CL2 <u>O</u> CL<u>O</u>	Chlorine dioxide <u>chlorine</u>	Strat
HCL	Hydrogen <u>hydrogen</u> chloride	Strat
<u>HOCL</u>	<u>hypochlorous acid</u>	<u>Strat</u>
<u>CH3CL</u>	<u>methyl chloride</u>	<u>Strat</u>
<u>CH3CCL3</u>	<u>methyl chloroform</u>	<u>Strat</u>
<u>CCL4</u>	<u>tetrachloromethane</u>	<u>Strat</u>
CLONO2	chlorine_nitrate	Strat
HOCL <u>CL</u> <u>NO2</u>	Hypochlorous acid <u>chloro(oxo)azane</u>	Strat
CL2 <u>CLO</u>	chlorine <u>monoxide</u>	Strat
O <u>CL</u> O <u>H</u> BR	Hydrogen bromide <u>chlorine dioxide</u>	Strat
BR <u>NO2</u> <u>CLO</u>	Bromine nitrate <u>asy mmetric chlorine</u>	Strat
CL2O2	dichlorine_dioxide	Strat
<u>BR</u>	<u>bromine atom</u>	<u>Strat</u>
<u>BR2</u>	<u>bromine atomic ground state</u>	<u>Strat</u>
<u>CH3BR</u>	<u>methyl bromide</u>	<u>Strat</u>
<u>CH2BR2</u>	<u>dibromomethane</u>	<u>Strat</u>
<u>CHBR3</u>	<u>bromoform</u>	<u>Strat</u>
<u>BRONO2</u>	<u>bromine nitrate</u>	<u>Strat</u>

<u>BRO</u>	<u>bromine monoxide</u>	<u>Strat</u>
<u>HBR</u>	<u>hydrogen bromide</u>	<u>Strat</u>
HOBR	Hypobromous <u>hypobromous</u> acid	Strat
BRCL	Bromine <u>bromine</u> monochloride	Strat
<u>HF</u>	<u>hydrofluoric acid</u>	<u>Strat</u>
CFC11	trichlorofluoromethane	Strat
CFC12	dichlorodifluoromethane	Strat
CFC113	trichlorotrifluoroethane	Strat
CFC114	1,2-	Strat
CFC115	Chloropentafluoroethane <u>chloropentafluoroethane</u>	Strat
<u>CCL4</u>	<u>tetrachloromethane</u>	<u>Strat</u>
<u>CLNO2</u>	Chloro(oxo)azane oxide	<u>Strat</u>
<u>CH3CCL3</u>	<u>Methyl chloroform</u>	<u>Strat</u>
<u>CH3CL</u>	<u>Methyl chloride</u>	<u>Strat</u>
HCFC22	chlorodifluoromethane	Strat
<u>CH3BR</u>	<u>Methyl bromide</u>	<u>Strat</u>
<u>HF</u>	<u>Hydrofluoric acid</u>	<u>Strat</u>
HA1301	Bromotrifluoromethane <u>bromotrifluoromethane</u>	Strat
HA1211	Bromochlorodifluoromethane <u>bromochlorodifluoromethane</u>	Strat
<u>CHBR3</u>	<u>Bromoform</u>	<u>Strat</u>

CLOO	Asymmetric chlorine dioxide radical	Strat
O	Oxygen atom	Strat
O1D	Excited oxygen atom	Strat
N	Nitrogen atom	Strat
CLO	Chlorine monoxide	Strat
CL	Chlorine atom	Strat
BR	Bromine atom	Strat
BRO	Bromine monoxide	Strat
H	Hydrogen atom	Strat
H2	hydrogen	Strat
CO2	carbondioxide	Strat
BR2	Bromine atomic ground state	Strat
CH2BR2	dibromomethane	Strat

Acknowledgments

MACC III was funded by the European Union's Seventh Framework Programme (FP7) under Grant Agreement no. 283576. We are grateful to the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) for providing ozone sonde observations and to the GOME-2, MIPAS, ACE-FTS and MLS teams for providing satellite observations.

5 References

- Atkinson, R., Baulch, D. L., Cox, R. A., Crowley, J. N., Hampson, R. F., Hynes, R. G., Jenkin, M. E., Rossi, M. J. and Troe, J.: evaluated kinetic and photochemical data for atmospheric chemistry: Volume I – gas phase reactions of Ox, HOx, NOx and SOx species, *Atmos. Chem. Phys.*, 4, 1461–1738, doi:10.5194/acp-4-1461-2004, 2004.
- Atkinson, R., Baulch, D. L., Cox, R. A., Crowley, J. N., Hampson, R. F., Hynes, R. G., Jenkin, M. E., Rossi, M. J., Troe, J.,
10 and IUPAC Subcommittee: Evaluated kinetic and photochemical data for atmospheric chemistry: Volume II – gas phase reactions of organic species, *Atmos. Chem. Phys.*, 6, 3625–4055, doi:10.5194/acp-6-3625-2006, 2006.
- Baldwin, M. P., Gray, L. J., Dunkerton, T. J., Hamilton, K., Haynes, P. H., Randel, W. J., Holton, J. R., Alexander, M. J., Hirota, I., Horinouchi, T., Jones, D. B. A., Kinnersley, J. S., Marquardt, C., Sato, K. and Takahashi, M.: The quasi-biennial oscillation, *Rev. Geophys.*, 39(2), 179–229, doi:10.1029/1999RG000073, 2001.
- 15 Bândă, N., Krol, M., van Weele, M., van Noije, T., Le Sager, P., and Röckmann, T.: Can we explain the observed methane variability after the Mount Pinatubo eruption?, *Atmos. Chem. Phys. Discuss.*, 15, 19111–19160, [doi:10.5194/acpd-15-19111-2015](https://doi.org/10.5194/acpd-15-19111-2015), 2015.
[acp-16-195-2016](https://doi.org/10.5194/acp-16-195-2016), 2016.
- Benedetti, A., Morcrette, J.-J., Boucher, O., Dethof, A., Engelen, R. J., Fisher, M., Flentje, H., Huneus, N., Jones, L., Kaiser, J. W., Kinne, S., Mangold, A., Razinger, M., Simmons, A. J., Suttie, M., and the GEMS-
20 AER team: Aerosol analysis and forecast in the European Centre for Medium-Range Weather Forecasts Integrated Forecast System: 2. Data assimilation, *J. Geophys. Res.*, 114, D13205, doi:10.1029/2008JD011115, 2009.
- Bovensmann, H., Burrows, J. P., Buchwitz, M., Freerick, J., Noël, S., Rozanov, V. V., Chance, K. V., and Goede, A. P. H.: SCIAMACHY: Mission Objectives and Measurement Modes, *J. Atmos. Sci.*, 56, 127–150, [doi: 10.1175/1520-0469\(1999\)056<0127:SMOAMM>2.0.CO;2](https://doi.org/10.1175/1520-0469(1999)056<0127:SMOAMM>2.0.CO;2), 1999.
- 25 Brasseur, G., A. Smith, R. Khosravi, T. Huang, S. Walters, S. Chabrilat and Kockarts, G.: Natural and human-induced perturbations in the middle atmosphere: A short tutorial, In *Atmospheric Science Across the Stratopause*, pages 7-20, AGU Geophys. Monograph, vol. [123123](https://doi.org/10.1029/GM123p0007), DOI: [10.1029/GM123p0007](https://doi.org/10.1029/GM123p0007), 2000.
- Cariolle, D. and Morcrette, J.-J.: A linearized approach to the radiative budget of the stratosphere: Influence of the ozone distribution, *Geophys. Res. Lett.*, 33, L05806, doi:10.1029/2005GL025597, 2006.
- 30 Cariolle, D. and Teyssède, H.: A revised linear ozone photochemistry parameterization for use in transport and general circulation models: multi-annual simulations, *Atmos. Chem. Phys.*, 7, 2183-2196, doi:10.5194/acp-7-2183-2007, 2007.

- Ceccherini, S., Cortesi, U., Verronen, P. T., and Kyrölä, E.: Technical Note: Continuity of MIPAS-ENVISAT operational ozone data quality from full- to reduced-spectral-resolution operation mode, *Atmospheric Chemistry and Physics*, 8, 2201–2212, doi:10.5194/acp-8-2201-2008, 2008.
- Chabrillat, S. and Fonteyn, D.: Modelling long-term changes of mesospheric temperature and chemistry, *Adv. Space Res.*, 32, 9, pages 1689-1700, [doi:10.1016/S0273-1177\(03\)90464-9](https://doi.org/10.1016/S0273-1177(03)90464-9), 2003.
- Chipperfield, M. P., Dhomse, S. S., Feng, W., McKenzie, R. L., Velders, G.J.M., Pyle, J. A.: Quantifying the ozone and ultraviolet benefits already achieved by the Montreal Protocol. *Nat Commun.*, 6, <http://dx.doi.org/10.1038/ncomms8233>, 2015.
- de Grandpré J., Ménard R., Rochon Y., Charette C., Chabrillat S. and Robichaud A. : Radiative impact of ozone on temperature predictability in a coupled chemistry-dynamics data assimilation system, *Monthly Weather Rev.*, 137, 679-692, doi:10.1175/2008MWR2572.1, 2009.
- [de Grandpré, J., Tanguay, M., Qaddouri, A., Zerroukat, M., and McLinden, C.A.: Semi-Lagrangian Advection of Stratospheric Ozone on a Yin–Yang Grid System. Monthly Weather Review 144:3, 1035-1050. doi: 10.1175/MWR-D-15-0142.1 .2016.](https://doi.org/10.1175/MWR-D-15-0142.1)
- De Mazière, M., Vigouroux, C., Bernath, P. F., Baron, P., Blumenstock, T., Boone, C., Brogniez, C., Catoire, V., Coffey, M., Duchatelet, P., Griffith, D., Hannigan, J., Kasai, Y., Kramer, I., Jones, N., Mahieu, E., Manney, G. L., Piccolo, C., Randall, C., Robert, C., Senten, C., Strong, K., Taylor, J., Tétard, C., Walker, K. A., and Wood, S.: Validation of ACE-FTS v2.2 methane profiles from the upper troposphere to the lower mesosphere, *Atmos. Chem. Phys.*, 8, 2421–2435, doi:10.5194/acp-8-2421-2008, <http://www.atmos-chem-phys.net/8/2421/2008/>, 2008.2008.
- [ECMWF: IFS documentation – Cy4r1. Operational implementation 12 May 2015. Available from http://www.ecmwf.int/en/forecasts/documentation-and-support/changes-ecmwf-model/ifs-documentation \(last access on 5 July 2016\). 2015.](https://www.ecmwf.int/en/forecasts/documentation-and-support/changes-ecmwf-model/ifs-documentation)
- Engelen, R. J., Serrar, S., and Chevallier, F.: Four-dimensional data assimilation of atmospheric CO₂ using AIRS observations, *J. Geophys. Res.*, 114, D03303, doi:10.1029/2008JD010739, 2009.
- Errera, Q. and Fonteyn, D.: Four-dimensional variational chemical assimilation of CRISTA stratospheric measurements, *J. Geophys. Res.*, 106, 12,253-12, 265, doi: 10.1029/2001JD900010 2001.
- Errera, Q., Daerden, F., Chabrillat, S., Lambert, J. C., Lahoz, W. A., Viscardy, S., Bonjean, S., and Fonteyn, D.: 4D-Var assimilation of MIPAS chemical observations: ozone and nitrogen dioxide analyses, *Atmos. Chem. Phys.*, 8, 6169-6187, doi:10.5194/acp-8-6169-2008, 2008.
- Eskes, H., Huijnen, V., Arola, A., Benedictow, A., Blechschmidt, A.-M., Botek, E., Boucher, O., Bouarar, I., Chabrillat, S., Cuevas, E., Engelen, R., Flentje, H., Gaudel, A., Griesfeller, J., Jones, L., Kapsomenakis, J., Katragkou, E., Kinne, S., Langerock, B., Razinger, M., Richter, A., Schultz, M., Schulz, M., Sudarchikova, N., Thouret, V., Vrekoussis, M., Wagner, A., and Zerefos, C.: Validation of reactive gases and aerosols in the MACC global analysis and forecast system, *Geosci. Model Dev.*, 8, 3523-3543, doi:10.5194/gmd-8-3523-2015, 2015a2015.

- Flemming, J., Inness, A., Flentje, H., Huijnen, V., Moinat, P., Schultz, M. G., and Stein, O.: Coupling global chemistry transport models to ECMWF's integrated forecast system, *Geosci. Model Dev.*, 2, 253-265, doi:10.5194/gmd-2-253-2009, 2009.
- 5 Flemming, J., Inness, A., Jones, L., Eskes, H. J., Huijnen, V., Schultz, M. G., Stein, O., Cariolle, D., Kinnison, D., and Brasseur, G.: Forecasts and assimilation experiments of the Antarctic ozone hole 2008, *Atmos. Chem. Phys.*, 11, 1961–1977, doi:10.5194/acp-11-1961-2011, 2011 .
- Flemming, J., Huijnen, V., Arteta, J., Bechtold, P., Beljaars, A., Blechschmidt, A.-M., Diamantakis, M., Engelen, R. J., Gaudel, A., Inness, A., Jones, L., Josse, B., Katragkou, E., Marecal, V., Peuch, V.-H., Richter, A., Schultz, M. G., Stein, O., and Tsikerdekis, A.: Tropospheric chemistry in the Integrated Forecasting System of ECMWF, *Geosci. Model Dev.*, 10 8, 975-1003, doi:10.5194/gmd-8-975-2015, 2015.
- Flemming, J. and Huijnen, V.: IFS Tracer Transport Study, MACC Deliverable G-RG 4.2, Tech. rep., ECMWF, available at: http://www.gmes-atmosphere.eu/documents/deliverables/g-rg/ifs_transport_study.pdf (last access: 15 December 2015), 2011.
- Fonteyn, D. and Larsen, N.: Detailed PSC formation in a two-dimensional chemical transport model of ~~the stratosphere~~
15 [the stratosphere](https://doi.org/10.1007/s00585-996-0315-0), *Ann. Geophys.*, 14, 315–328, doi:10.1007/s00585-996-0315-0 1996.
- [Froidevaux, L., Jiang, Y. B., Lambert, A., Livesey, N. J., Read, W. G., Waters, J. W., Browell, E. V., Hair, J. W., Avery, M. A., McGee, T. J., Twigg, L. W., Summicht, G. K., Jucks, K. W., Margitan, J. J., Sen, B., Stachnik, R. A., Toon, G. C., Bernath, P. F., Boone, C. D., Walker, K. A., Filipiak, M. J., Harwood, R. S., Fuller, R. A., Manney, G. L., Schwartz, M. J., Daffer, W. H., Drouin, B. J., Cofield, R. E., Cuddy, D. T., Jarnot, R. F., Knosp, B. W., Perun, V. S., Snyder, W. V., Stek, P. C., Thurstans, R. P., and Wagner, P. A.: Validation of Aura Microwave Limb Sounder stratospheric ozone measurements, *J. Geophys. Res.*, 113, doi:10.1029/2007JD008771, 2008a.](https://doi.org/10.1029/2007JD008771)
- 20 Froidevaux, L., Jiang, Y., Lambert, A., Livesey, N., Read, W., Waters, J., Fuller, R., Marcy, T., Popp, P., Gao, R., et al.: Validation of Aura microwave limb sounder HCl measurements, *Journal of Geophysical Research: Atmospheres* (1984–2012), 113, [2008](https://doi.org/10.1029/2007JD009025), doi:10.1029/2007JD009025 2008b.
- 25 Gaudel, A., Clark, H., Thouret, V., Jones, L., Inness, A., Flemming, J., Stein, O., Huijnen, V., Eskes, H., Nédélec, P., Boulanger, D.: On the use of MOZAIC-IAGOS data to assess the ability of the MACC reanalysis to reproduce the distribution of ozone and CO in the UTLS over Europe. *Tellus B*, 67, 27955. doi:<http://dx.doi.org/10.3402/tellusb.v67.27955>, 2015.
- Geer, A. J., Lahoz, W. A., Bekki, S., Bormann, N., Errera, Q., Eskes, H. J., Fonteyn, D., Jackson, D. R., Juckes, M. N., Massart, S., Peuch, V.-H., Rharmili, S., and Segers, A.: The ASSET intercomparison of ozone analyses: method and first
30 results, *Atmos. Chem. Phys.*, 6, 5445-5474, doi:10.5194/acp-6-5445-2006, 2006.
- Hanson, D. R. and Ravishankara, A. R.: Reactive Uptake of ClONO₂ onto Sulfuric Acid Due to Reaction with HCl and H₂O, *J. Phys. Chem.*, 98, 5728–5735, doi:10.1021/j100073a026, 1994.

- Hassler, B., Petropavlovskikh, I., Staehelin, J., August, T., Bhartia, P. K., Clerbaux, C., Degenstein, D., Mazière, M. De, Dinelli, B. M., Dudhia, A., Dufour, G., Frith, S. M., Froidevaux, L., Godin-Beekmann, S., Granville, J., Harris, N. R. P., Hoppel, K., Hubert, D., Kasai, Y., Kurylo, M. J., Kyrölä, E., Lambert, J.-C., Levelt, P. F., McElroy, C. T., McPeters, R. D., Munro, R., Nakajima, H., Parrish, A., Raspollini, P., Remsberg, E. E., Rosenlof, K. H., Rozanov, A., Sano, T., Sasano, Y., Shiotani, M., Smit, H. G. J., Stiller, G., Tamminen, J., Tarasick, D. W., Urban, J., van der A, R. J., Veefkind, J. P., Vigouroux, C., von Clarmann, T., von Savigny, C., Walker, K. A., Weber, M., Wild, J., and Zawodny, J. M.: Past changes in the vertical distribution of ozone – Part 1: Measurement techniques, uncertainties and availability, *Atmos. Meas. Tech.*, 7, 1395-1427, doi:10.5194/amt-7-1395-2014, 2014.
- Im, U., R. Bianconi, E. Solazzo, I. Kioutsioukis, A. Badia, A. Balzarini, R. Baro, R. Bellasio, D. Brunner, C. Chemel, G. Curci, J. Flemming, R. Forkel, L. Giordano, P. Jimenez-Guerrero, M. Hirtl, A. Hodzic, L. Honzak, O. Jorba, C. Knote, J.J.P. Kuenen, P.A. Makar, A. Manders-Groot, L. Neal, J.L. Perez, G. Pirovano, G. Pouliot, R. San Jose, N. Savage, W. Schröder, R.S. Sokhi, D. Syrakov, A. Torian, P. Tuccella, K. Werhahn, R. Wolke, K. Yahya, R. Žabkar, Y. Zhang, J. Zhang, C. Hogrefe, S. Galmarini. Evaluation of operational online-coupled regional air quality models over Europe and North America in the context of AQMEII phase2. Part I: ozone. *Atmos. Environ.*, 115–(2015), pp. 404–420
<http://dx.doi.org/10.1016/j.atmosenv.2014.09.042>, 2015.
- Hanson, D., and Mauersberger, K.: Laboratory studies of the nitric acid trihydrate: Implications for the south polar stratosphere. *GRL* 15 (8), pp 855–858. DOI: 10.1029/GL015i008p00855, 1988.
- Hitchman, M. H., M. McKay, and C. R. Trepte, A climatology of stratospheric aerosol. *J. Geophys. Res.*, 99(D10), 20689–20700. doi:10.1029/94JD01525, 1994.
- Hollingsworth, A., Engelen, R.J., Textor, C., Benedetti, A., Boucher, O., Chevallier, F., Dethof, A., Elbern, H., Eskes, H., Flemming, J., Granier, C., Kaiser, J.W., Morcrette, J.-J., Rayner, P., Peuch, V.H., Rouil, L., Schultz, M.G., Simmons, A.J and The GEMS Consortium: Toward a Monitoring and Forecasting System For Atmospheric Composition: The GEMS Project. *Bull. Amer. Meteor. Soc.*, 89, 1147-1164, doi:10.1175/2008BAMS2355.1, 2008.
- Huijnen, V., Williams, J., van Weele, M., van Noije, T., Krol, M., Dentener, F., Segers, A., Houweling, S., Peters, W., de Laat, J., Boersma, F., Bergamaschi, P., van Velthoven, P., Le Sager, P., Eskes, H., Alkemade, F., Scheele, R., Nédélec, P., and Pätz, H.-W.: The global chemistry transport model TM5: description and evaluation of the tropospheric chemistry version 3.0, *Geosci. Model Dev.*, 3, 445-473, doi:10.5194/gmd-3-445-2010.
- Huijnen, V., Williams, J. E., and Flemming, J.: Modeling global impacts of heterogeneous loss of HO₂ on cloud droplets, ice particles and aerosols, *Atmos. Chem. Phys. Discuss.*, 14, 8575-8632, doi:10.5194/acpd-14-8575-2014, 2014.
- Inness, A., Blechschmidt, A.-M., Bouarar, I., Chabrilat, S., Crepulja, M., Engelen, R. J., Eskes, H., Flemming, J., Gaudel, A., Hendrick, F., Huijnen, V., Jones, L., Kapsomenakis, J., Katragkou, E., Keppens, A., Langerock, B., de Mazière, M., Melas, D., Parrington, M., Peuch, V. H., Razinger, M., Richter, A., Schultz, M. G., Suttie, M., Thouret, V., Vrekoussis, M., Wagner, A., and Zerefos, C.: Data assimilation of satellite-retrieved ozone, carbon monoxide and nitrogen dioxide with ECMWF's Composition-IFS, *Atmos. Chem. Phys.*, 15, 5275-5303, doi:10.5194/acp-15-5275-2015, 2015.

[Jablonowski, C., and Williamson, D. L.: The pros and cons of diffusion, filters and fixers in atmospheric general circulation models, in: Numerical Techniques for Global Atmospheric Models \(pp. 381-493\). Springer Berlin Heidelberg, doi:10.1007/978-3-642-11640-7, 2011.](#)

Jöckel, P., Kerkweg, A., Pozzer, A., Sander, R., Tost, H., Riede, H., Baumgaertner, A., Gromov, S., and Kern, B.:
5 Development cycle 2 of the Modular Earth Submodel System (MESSy2), *Geosci. Model Dev.*, 3, 717-752,
doi:10.5194/gmd-3-717-2010, 2010.

Kaiser, J. W., Heil, A., Andreae, M. O., Benedetti, A., Chubarova, N., Jones, L., Morcrette, J.-J., Razinger, M., Schultz, M.
G., Suttie, M., and van der Werf, G. R.: Biomass burning emissions estimated with a global fire assimilation system
based on observed fire radiative power, *Biogeosciences*, 9, 527-554, doi:10.5194/bg-9-527-2012, 2012.

10 Lahoz, W. A., Errera, Q., Viscardy, S., and Manney, G. L.: The 2009 stratospheric major warming described from
synergistic use of BASCOE water vapour analyses and MLS observations, *Atmos. Chem. Phys.*, 11, 4689-4703,
[doi:10.5194/acp-11-4689-2011](#), 2011.

Lefever, K., van der A, R., Baier, F., Christophe, Y., Errera, Q., Eskes, H., Flemming, J., Inness, A., Jones, L., Lambert, J.-
C., Langerock, B., Schultz, M. G., Stein, O., Wagner, A., and Chabrillat, S.: Copernicus stratospheric ozone service,
15 2009–2012: validation, system intercomparison and roles of input data sets, *Atmos. Chem. Phys.*, 15, 2269-2293,
doi:10.5194/acp-15-2269-2015, 2015.

[Lin, S.-J. and Rood, R. B.: Multidimensional flux-form semi-Lagrangian transport schemes, *Mon. Weather Rev.*, 124, 2046–2070, 1996.](#)

Livesey, N. J., Read, W. G., Froidevaux, L., Lambert, A., Manney, G. L., Pumphrey, H. C., Santee, M. L., Schwartz, M. J.,
20 Wang, S., Cofield, R. E., Cuddy, D. T., Fuller, R. A., Jarnot, R. F., Jiang, J. H., Knosp, B. W., Stek, P. C., Wagner, P. A.,
and Wu, D. L.: Earth Observing System (EOS) Aura Microwave Limb Sounder (MLS) Version 3.3 Level 2 data quality
and description document, Tech. Rep. D-33509, JPL, 2011.

Madronich, S. and Flocke, S.: The Role of Solar Radiation in Atmospheric Chemistry, in: *Environmental Photochemistry*,
edited by Boule, P., vol. 2 / 2L of *The Handbook of Environmental Chemistry*, pp. 1–26, Springer Berlin Heidelberg,
25 doi:10.1007/978-3-540-69044-3_1, 1999.

Manney, G. L., Lawrence, Z. D., Santee, M. L., Livesey, N. J., Lambert, A., and Pitts, M. C.: Polar processing in a split
vortex: Arctic ozone loss in early winter 2012/2013, *Atmos. Chem. Phys.*, 15, 5381-5403, doi:10.5194/acp-15-5381-
2015, 2015.

30 Marécal, V., Peuch, V.-H., Andersson, C., Andersson, S., Arteta, J., Beekmann, M., Benedictow, A., Bergström, R.,
Bessagnet, B., Cansado, A., Chéroux, F., Colette, A., Coman, A., Curier, R. L., Denier van der Gon, H. A. C., Drouin,
A., Elbern, H., Emili, E., Engelen, R. J., Eskes, H. J., Foret, G., Friese, E., Gauss, M., Giannaros, C., Guth, J., Joly, M.,
Jaumouillé, E., Josse, B., Kadyrov, N., Kaiser, J. W., Krajsek, K., Kuenen, J., Kumar, U., Liora, N., Lopez, E.,
Malherbe, L., Martinez, I., Melas, D., Meleux, F., Menuet, L., Moinat, P., Morales, T., Parmentier, J., Piacentini, A., Plu,
M., Poupkou, A., Queguiner, S., Robertson, L., Rouïl, L., Schaap, M., Segers, A., Sofiev, M., Thomas, M., Timmermans,

- R., Valdebenito, Á., van Velthoven, P., van Versendaal, R., Vira, J., and Ung, A.: A regional air quality forecasting system over Europe: the MACC-II daily ensemble production, *Geosci. Model Dev. Discuss.*, 8, 2739-2806, doi:10.5194/gmdd-8-2739-2015, 2015.
- Maycock, A. C., Keeley, S. P. E., Charlton-Perez, A. J., and Doblas-Reyes, F. J.: Stratospheric circulation in seasonal forecasting models: implications for seasonal prediction, *Clim. Dynam.*, 36, 309–321, doi:10.1007/s00382-009-0665-x, 2011.
- McGregor, J. L.: C-CAM Geometric Aspects and Dynamical Formulation, Tech. Rep. 70, CSIRO Atmospheric Research, Aspendale, Victoria, 2005.
- Monge-Sanz, B. M., Chipperfield, M. P., Untch, A., Morcrette, J.-J., Rap, A., and Simmons, A. J.: On the uses of a new linear scheme for stratospheric methane in global models: water source, transport tracer and radiative forcing, *Atmos. Chem. Phys.*, 13, 9641-9660, doi:10.5194/acp-13-9641-2013, 2013.
- Morcrette, J.-J., Boucher, O., Jones, L., Salmond, D., Bechtold, P., Beljaars, A., Benedetti, A., Bonet, A., Kaiser, J. W., Razingerg, M., Schulz, M., Serrar, S., Simmons, A. J., Sofiev, M., Suttie, M., Tompkins, A. M., and Untch, A.: Aerosol analysis and forecast in the [ECMWF European Centre for Medium-Range Weather Forecasts](http://www.ecmwf.int) Integrated Forecast System ~~Part I: Forward modelling~~ [modeling](http://www.ecmwf.int), *J. Geophys. Res.*, 114, D06206, doi:10.1029/2008JD011235, 2009.
- Michou M., P. Laville, D. Serça, A. Fotiadi, P. Bouchou and V.-H. Peuch, Measured and modeled dry deposition velocities over the ESCOMPTE area, *Atmos. Res.*, 74 (1-4), 89-116, 2004, doi:10.1016/j.atmosres.2004.04.011, 2004.
- Muncaster, R., Bourqui, M. S., Chabrilat, S., Viscardy, S., Melo, S., and Charbonneau, P.: A simple framework for modelling the photochemical response to solar spectral irradiance variability in the stratosphere, *Atmos. Chem. Phys.*, 12, 7707-7724, doi:10.5194/acp-12-7707-2012, 2012.
- [Murphy, D. M. and Koop, T.: Review of the vapour pressures of ice and supercooled water for atmospheric applications. Q.J.R. Meteorol. Soc., 131: 1539–1565. doi: 10.1256/qj.04.94, 2005.](http://www.jrnl.org)
- Pausata, F. S. R., Pozzoli, L., Vignati, E., and Dentener, F. J.: North Atlantic Oscillation and tropospheric ozone variability in Europe: model analysis and measurements intercomparison, *Atmos. Chem. Phys.*, 12, 6357-6376, doi:10.5194/acp-12-6357-2012, 2012.
- Platt, U. and Stutz, J.: Differential Optical Absorption Spectroscopy, *Physics of Earth and Space Environments*, Berlin: Springer, available at: <http://www.springerlink.com/content/978-3-540-21193-8> (last access: February 2016), 2008.
- Read, W., Lambert, A., Bacmeister, J., Cofield, R., Christensen, L., Cuddy, D., Daffer, W., Drouin, B., Fetzer, E., Froidevaux, L., et al.: Aura Microwave Limb Sounder upper tropospheric and lower stratospheric H₂O and relative humidity with respect to ice validation, *Journal of Geophysical Research: Atmospheres*, 112, doi:10.1029/2007JD008752, 2007.
- Robichaud, A., Ménard, R., Chabrilat, S., de Grandpré, J., Rochon, Y. J., Yang, Y. and Charette, C : Impact of energetic particle precipitation on stratospheric polar constituents: an assessment using monitoring and assimilation of operational MIPAS data, *Atmos. Chem. Phys.*, 10, 1739-1757, doi:10.5194/acp-10-1739-2010, 2010.

- Qu, Z., Gschwind, B., Lefevre, M., and Wald, L.: Improving HelioClim-3 estimates of surface solar irradiance using the McClear clear-sky model and recent advances in atmosphere composition, *Atmos. Meas. Tech.*, 7, 3927-3933, doi:10.5194/amt-7-3927-2014, 2014.
- Raspollini, P., Carli, B., Carlotti, M., Ceccherini, S., Dehn, A., Dinelli, B. M., Dudhia, A., Flaud, J.-M., López-Puertas, M., Niro, F., Remedios, J. J., Ridolfi, M., Sembhi, H., Sgheri, L., and von Clarmann, T.: Ten years of MIPAS measurements with ESA Level 2 processor V6 – Part 1: Retrieval algorithm and diagnostics of the products, *Atmos. Meas. Tech.*, 6, 2419–2439, doi:10.5194/amt-6-2419-2013, <http://www.atmos-meas-tech.net/6/2419/2013/2013.2013>.
- Richter, A., Burrows, J. P., Nüß, H., Granier, C., Niemeier, U.: Increase in tropospheric nitrogen dioxide over China observed from space, *Nature*, 437, 129-132, doi: 10.1038/nature04092, 2005.
- Sander, S.P., et al., Chemical Kinetics and Photochemical Data for Use in Stratospheric Modeling. Supplement to Evaluation 12: Update of Key Reactions, Evaluation Number 13, JPL Publication 00-03, Jet Propulsion Laboratory, Pasadena, 2000.
- Sander, S.P., et al., Chemical Kinetics and Photochemical Data for Use in Atmospheric Studies. Evaluation Number 15. JPL Publication 06-2, Pasadena, 2006.
- Sander, S. P., Abbatt, J. R., Burkholder, J. B., Friedl, R. R., Golden, D. M., Huie, R. E., Kolb, C. E., Kurylo, G., Moortgat, K., Orkin, V. L. and Wine, P. H.: Chemical kinetics and Photochemical Data for Use in Atmospheric studies, Evaluation No.17, JPL Publication 10-6, Jet Propulsion Laboratory, Pasadena, 2011.
- Sandu, A. and Sander, R.: Technical note: Simulating chemical systems in Fortran90 and Matlab with the Kinetic PreProcessor KPP-2.1, *Atmos. Chem. Phys.*, 6, 187-195, doi:10.5194/acp-6-187-2006, 2006.
- [Santee, M. L., A. Lambert, W. G. Read, N. J. Livesey, G. L. Manney, R. E. Cofeld, D. T. Cuddy, W. H. Daffer, B. J. Drouin, L. Froidevaux, R. A. Fuller, R. F. Jamot, B. W. Knosp, V. S. Perun, W. V. Snyder, P. C. Stek, R. P. Thurstans, P. A. Wagner, J. W. Waters, B. Connor, J. Urban, D. Murtagh, P. Ricaud, B. Barret, A. Kleinboehl, J. Kuttippurath, H. Kuellmann, M. von Hobe, G. C. Toon, and R. A. Stachnik: Validation of the Aura Microwave Limb Sounder ClO measurements. *J. Geophys. Res.*, 113:D15S22. doi:10.1029/2007JD008762. 2008.](#)
- Scaife, A., Spanghel, T., Fereday, D., Cubasch, U., Langematz, U., Akiyoshi, H., Bekki, S., Braesicke, P., Butchart, N., Chipperfield, M., Gettelman, A., Hardiman, S., Rozanov, M. M. E., and Shepherd, T.: Climate change projections and stratosphere-troposphere interaction, *Climate Dyn.*, 38, 2089–2097, doi:10.1007/s00382-011-1080-7, 2012.
- [Shepherd, T. G.: Transport in the Middle Atmosphere, *J. Met. Soc. Jap.*, 85B, 165–191, doi: 10.2151/jmsj.85B.165. 2007.](#)
- Solomon, S., Rosenlof, K. H., Portmann, R.W., Daniel, J. S., Davis, S. M., Sanford, T. J., and Plattner, G.-K.: Contributions of stratospheric water vapor to decadal changes in the rate of global warming, *Science*, 327, 1219–1223, [2010doi:10.1126/science.1182488](https://doi.org/10.1126/science.1182488) 2010
- Stevenson, D. S. and Dentener, F. J. and Schultz, M. G. and Ellingsen, K. and van Noije, T. P. C. and Wild, O. and Zeng, G. and Amann, M. and Atherton, C. S. and Bell, N. and Bergmann, D. J. and Bey, I. and Butler, T. and Cofala, J. and

- Collins, W. J. and Derwent, R. G. and Doherty, R. M. and Drevet, J. and Eskes, H. J. and Fiore, A. M. and Gauss, M. and Hauglustaine, D. A. and Horowitz, L. W. and Isaksen, I. S. A. and Krol, M. C. and Lamarque, J.-F. and Lawrence, M. G. and Montanaro, V. and Müller, J.-F. and Pitari, G. and Prather, M. J. and Pyle, J. A. and Rast, S. and Rodriguez, J. M. and Sanderson, M. G. and Savage, N. H. and Shindell, D. T. and Strahan, S. E. and Sudo, K. and Szopa, S. : Multimodel ensemble simulations of present-day and near-future tropospheric ozone, *J. Geophys. Res.*, 111, D08301, doi:10.1029/2005JD006338, 2006.
- Strong, K., Wolff, M. A., Kerzenmacher, T. E., Walker, K. A., Bernath, P. F., Blumenstock, T., Boone, C., Catoire, V., Coffey, M., De Mazière, M., Demoulin, P., Duchatelet, P., Dupuy, E., Hannigan, J., Höpfner, M., Glatthor, N., Griffith, D. W. T., Jin, J. J., Jones, N., Jucks, K., Kuellmann, H., Kuttippurath, J., Lambert, A., Mahieu, E., McConnell, J. C., Mellqvist, J., Mikuteit, S., Murtagh, D. P., Notholt, J., Piccolo, C., Raspollini, P., Ridolfi, M., Robert, C., Schneider, M., Schrems, O., Semeniuk, K., Senten, C., Stiller, G. P., Strandberg, A., Taylor, J., Tétard, C., Toohey, M., Urban, J., Warneke, T., and Wood, S.: Validation of ACE-FTS N₂O measurements, *Atmos. Chem. Phys.*, 8, 4759–4786, doi:10.5194/acp-8-4759-2008, <http://www.atmos-chem-phys.net/8/4759/2008/>, 2008, 2008.
- SPARC CCM Val Report on the Evaluation of Chemistry-Climate Models, V. Eyring, T. G. Shepherd, D. W. Waugh (Eds.), SPARC Report No. 5, WCRP-X, WMO/TD-No. X, 2010., <http://www.sparc-climate.org/publications/>, 2010.
- Talagrand, O. and Courtier, P.: Variational assimilation of meteorological observations with the adjoint vorticity equation, I, *Theory*, *Q. J. Roy. Meteor. Soc.*, 23, 1311–1328, DOI: [10.1002/qj.49711347812](https://doi.org/10.1002/qj.49711347812), 1987.
- Theys, N., Van Roozendael, M., Errera, Q., Hendrick, F., Daerden, F., Chabrillat, S., Dorf, M., Pfeilsticker, K., Rozanov, A., Lotz, W., Burrows, J. P., Lambert, J.-C., Goutail, F., Roscoe, H. K. and De Mazière, M.: A global stratospheric bromine monoxide climatology based on the BASCOE chemical transport model, *Atmos. Chem. Phys.*, 9, 831-848, doi: 10.5194/acp-9-831-2009, 2009.
- Thornton, H. E., Jackson, D. R., Bekki, S., Bormann, N., Errera, Q., Geer, A. J., Lahoz, W. ~~LA.~~, and ~~RhalmiliRharmili, S.~~, The ASSET intercomparison of stratosphere and lower mesosphere humidity analyses, *Atmos. Chem. Phys.*, 9, 995-~~116~~ ~~1016~~, doi:10.5194/acp-9-995-2009, 2009.
- Thubum, J., and G. Craig, GCM tests of theories for the height of the tropopause, *J. Atmos. Sci.*, 54, 869–882, ~~1997~~-doi: [10.1175/1520-0469\(1997\)054<0869:GTOTFT>2.0.CO;2](https://doi.org/10.1175/1520-0469(1997)054<0869:GTOTFT>2.0.CO;2), 1997.
- van der A, R. J., Allaart, M. A. F., and Eskes, H. J.: Extended and refined multi sensor reanalysis of total ozone for the period 1970–2012, *Atmos. Meas. Tech.*, 8, 3021-3035, doi:10.5194/amt-8-3021-2015, 2015.
- Viscardy, S., Errera, Q., Christophe, Y., Chabrillat, S. and Lambert, J. -C.: Evaluation of Ozone Analyses From UARS MLS Assimilation by BASCOE Between 1992 and 1997, *IEEE J-STARS*, 3, 190-202, doi:10.1109/JSTARS.2010.2040463, 2010.
- von Glasow, R. and Crutzen, P. J.: Tropospheric halogen chemistry, in: *The Atmosphere* (ed. R. F. Keeling), Vol. 4 *Treatise on Geochemistry* (eds. H. D. Holland and K. K. Turekian), Elsevier-Pergamon, Oxford, 2007.

- Wang, D. Y., Höpfner, M., Blom, C. E., Ward, W. E., Fischer, H., Blumenstock, T., Hase, F., Keim, C., Liu, G. Y., Mikuteit, S., Oelhaf, H., Wetzel, G., Cortesi, U., Mencaraglia, F., Bianchini, G., Redaelli, G., Pirre, M., Catoire, V., Huret, N., Vigouroux, C., De Mazière, M., Mahieu, E., Demoulin, P., Wood, S., Smale, D., Jones, N., Nakajima, H., Sugita, T., Urban, J., Murtagh, D., Boone, C. D., Bernath, P. F., Walker, K. A., Kuttippurath, J., Kleinböhl, A., Toon, G., and Piccolo, C.: Validation of MIPAS HNO₃ operational data, *Atmos. Chem. Phys.*, 7, 4905-4934, doi:10.5194/acp-7-4905-2007, 2007.
- ~~Wesely, M.L.: Parameterization of Surface Resistances to Gaseous Dry Deposition in Regional Scale Numerical Models. *Atmos. Environ.*, 23, 1293-1304, 1989.~~
- Wetzel, G., Bracher, A., Funke, B., Goutail, F., Hendrick, F., Lambert, J.-C., Mikuteit, S., Piccolo, C., Pirre, M., Bazureau, A., Belotti, C., Blumenstock, T., De Mazière, M., Fischer, H., Huret, N., Ionov, D., López-Puertas, M., Maucher, G., Oelhaf, H., Pommereau, J.-P., Ruhnke, R., Sinnhuber, M., Stiller, G., Van Roozendael, M., and Zhang, G.: Validation of MIPAS-ENVISAT NO₂ operational data, *Atmos. Chem. Phys.*, 7, 3261-3284, doi:10.5194/acp-7-3261-2007, 2007.
- Williams, J. E., Strunk, A., Huijnen, V., and van Weele, M.: The application of the Modified Band Approach for the calculation of on-line photodissociation rate constants in TM5: implications for oxidative capacity, *Geosci. Model Dev.*, 5, 15-35, doi:10.5194/gmd-5-15-2012, 2012.
- Williams, J. E., van Velthoven, P. F. J., and Brenninkmeijer, C. A. M.: Quantifying the uncertainty in simulating global tropospheric composition due to the variability in global emission estimates of Biogenic Volatile Organic Compounds, *Atmos. Chem. Phys.*, 13, 2857-2891, doi:10.5194/acp-13-2857-2013, 2013.
- Wittrock, F., A. Richter, H. Oetjen, J. P. Burrows, M. Kanakidou, S. Myriokefalitakis, R. Volkamer, S. Beirle, U. Platt, and T. Wagner, Simultaneous global observations of glyoxal and formaldehyde from space, *Geophys. Res. Lett.*, 33, L16804, doi:10.1029/2006GL026310, 2006Compounds, *Atmos. Chem. Phys.*, 13, 2857-2891, doi:10.5194/acp-13-2857-2013, 2013.
- World Meteorological Organization (WMO), *Meteorology A Three-Dimensional Science: Second Session of the Commission for Aerology*, WMO Bulletin IV(4), WMO, Geneva, 134–138, 1957.
- Yarwood, G., Rao, S., Yocke, M., and Whitten, G.: Updates to the carbon bond chemical mechanism: CB05. Final report to the US EPA, EPA Report Number: RT-0400675, available at: www.camx.com, last access: 16 July 2015, 2005.

Table 1. Trace gases relevant for the stratosphere which are constrained at the surface. The constant surface volume mixing ratios are also given.

N ₂ O	CFC11	CFC12	CFC113	CFC114	CCl ₄	CH ₃ CCl ₃
3.22E-7	2.59E-10	5.37E-10	7.93E-11	4.25E-12	1.02E-10	4.53E-11
HCFC22	HA1301	HA1211	CH ₃ Br	CHBR ₃	CH ₃ Cl	CO ₂
1.70E-10	3.30E-12	4.62E-12	9.08E-12	1.17E-12	5.44E-10	3.80E-4

5 | **Table 2.** Number of ~~tracers, trace gases, the chemistry scheme in troposphere and stratosphere, and corresponding number of~~ reactions (gas-phase / heterogeneous and photolytic), ~~and as well as specification of the circulation model and~~ computational expenses of a one-month run on T255L60 in terms of system billing units (SBU) for various C-IFS model versions. For completeness also the BASCOE-CTM system is indicated.

	C-IFS-T	C-IFS-S	C-IFS-Atmos	C-IFS-TS	BASCOE-CTM
No. tracers, trace <u>gases</u>	55	59	99	99	59
<u>Chemistry scheme in troposphere</u>	<u>CB05</u>	<u>BASCOE (P<400hPa)</u>	<u>CB05+BASCOE</u>	<u>CB05</u>	<u>BASCOE (P<400hPa)</u>
<u>Chemistry scheme in stratosphere</u>	<u>CB05/Cariolle</u>	<u>BASCOE</u>	<u>CB05+BASCOE</u>	<u>BASCOE</u>	<u>BASCOE</u>
No. reactions (gas / het / photo)	93/3/18	142/9/52	211/11/60	93/3/18 or 142/9/52	142/9/52
<u>Circulation model</u>	<u>GCM</u>	<u>GCM</u>	<u>GCM</u>	<u>GCM</u>	<u>CTM</u>
SBU	2075	2500	4563	3076	- ^a

10 | ^aBASCOE does not run on the ECMWF supercomputing facility and hence cannot be compared directly to C-IFS in terms of computational resources.

Table 3. Parameterization of photolysis rates for troposphere (CB05-based) and stratosphere (BASCOE-based)

	Troposphere (Williams et al., 2012)	Stratosphere (Errera and Fonteyn, 2001)
No. J-rates	18	52
Method	2-stream online solver, $204 < \lambda < 705 \text{nm}$	Lookup table approach, $116 < \lambda < 705 \text{nm}$
Dependencies	O ₃ overhead, pressure, solar zenith angle, cloud, aerosol, surface albedo, temperature	O ₃ overhead, pressure, solar zenith angle
terminator treatment	J>0 for $\text{sza} < 85^\circ$	J>0 for $\text{sza} < 96^\circ$, Chapman approximation

Table 4. Selection of photolytic reactions that are merged between troposphere and stratosphere. The reaction product O₂ is not shown.

Name	reaction (stratosphere)	reaction products (troposphere) ^a
J O3	$O_3 + hv \rightarrow O^1D$	
J NO2	$NO_2 + hv \rightarrow NO + O$	NO + O ₃
J H2O2	$H_2O_2 + hv \rightarrow 2OH$	
J HNO3	$HNO_3 + hv \rightarrow OH + NO_2$	
J HO2NO2	$HO_2NO_2 + hv \rightarrow HO_2 + NO_2$	
J N2O5	$N_2O_5 + hv \rightarrow NO_2 + NO_3$	
J CH2O-a	$CH_2O + hv \rightarrow HCO + H$	CO + 2HO ₂
JCH2O-b	$CH_2O + hv \rightarrow CO + H_2$	CO
J NO3-a	$NO_3 + hv \rightarrow NO_2 + O$	NO ₂ + O ₃
J NO3-b	$NO_3 + hv \rightarrow NO$	
J O2	$O_2 + hv \rightarrow 2O$	
J CH3OOH	$CH_3OOH + hv \rightarrow CH_3O + OH$	CH ₂ O + HO ₂ + OH

^a Only specified in case this is different from the stratospheric reaction.

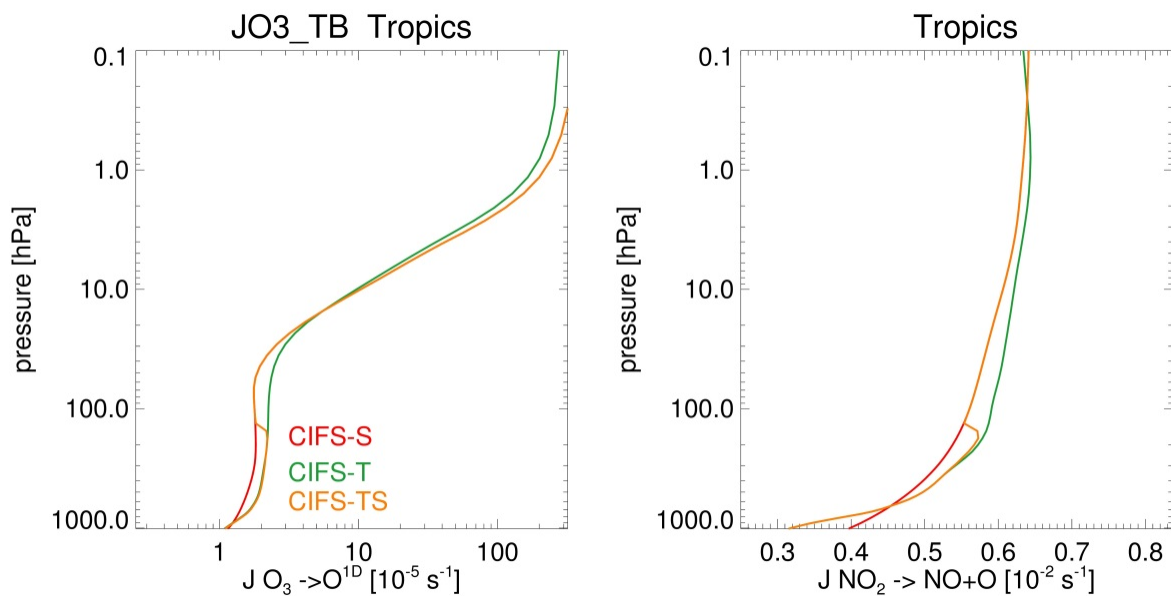


Figure 1. Illustration of the merging procedure for photolysis rates between the tropospheric and stratospheric parameterizations for the reaction $\text{O}_3 \rightarrow \text{O}^1\text{D}$ (left) and $\text{NO}_2 \rightarrow \text{NO} + \text{O}$ (right) as zonally averaged over the tropics for 1 April 2008.

5

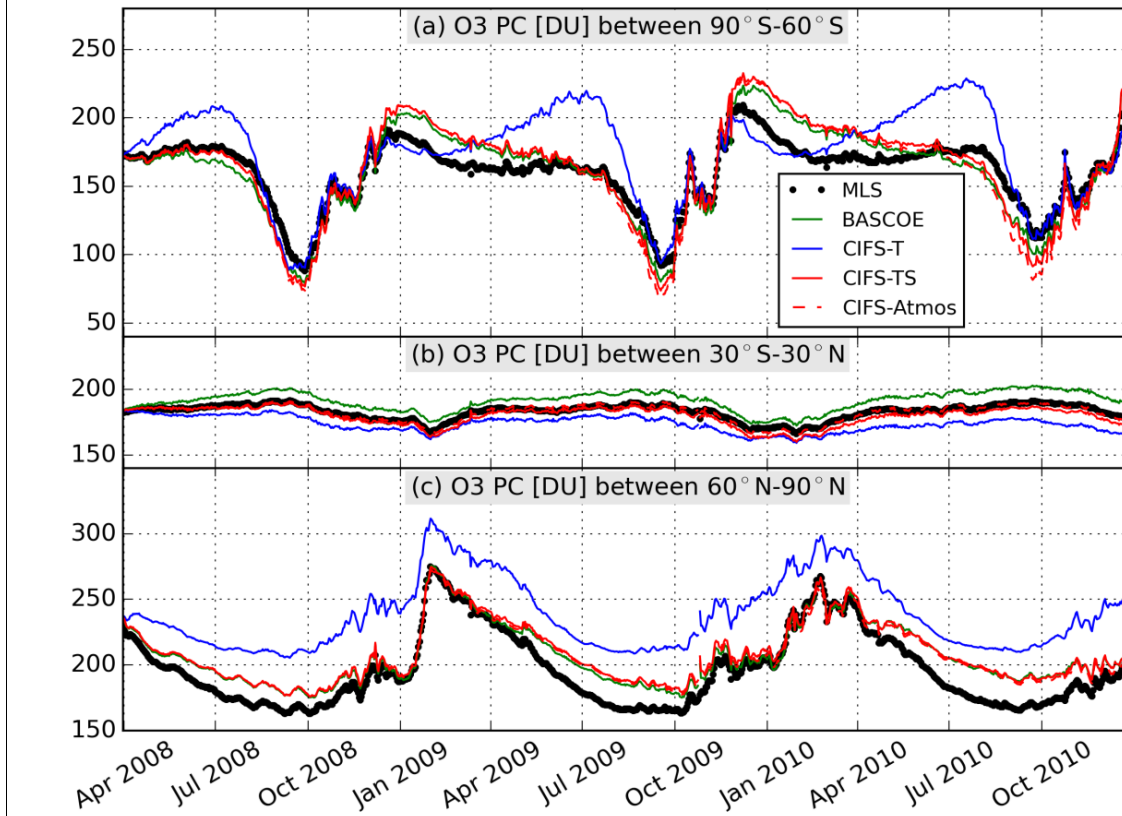


Figure 2. Evaluation of daily averages of monthly mean O_3 total partial columns in Dobson Units against the Multi Sensor Reanalysis (10-100hPa) for the Arctic ($60^\circ N$ - $90^\circ N$ - $70^\circ N$), Northern mid-latitudes ($60^\circ N$ - $30^\circ N$), tropics (Tropics ($30^\circ S$ - $30^\circ N$ - $30^\circ S$), Southern Hemisphere mid-latitudes ($30^\circ S$ - $60^\circ S$) and Antarctica ($70^\circ S$ -Antarctic ($60^\circ N$ - $90^\circ S$)-N) over the period April 2008 – December 2010. Datasets are averaged in 5-day bins and model output is interpolated to the location and time of Aura MLS v3 retrievals (black dots). Blue line: C-IFS-T; green line: BASCOE-CTM; red dashed line: C-IFS-Atmos; red solid line: C-IFS-TS.

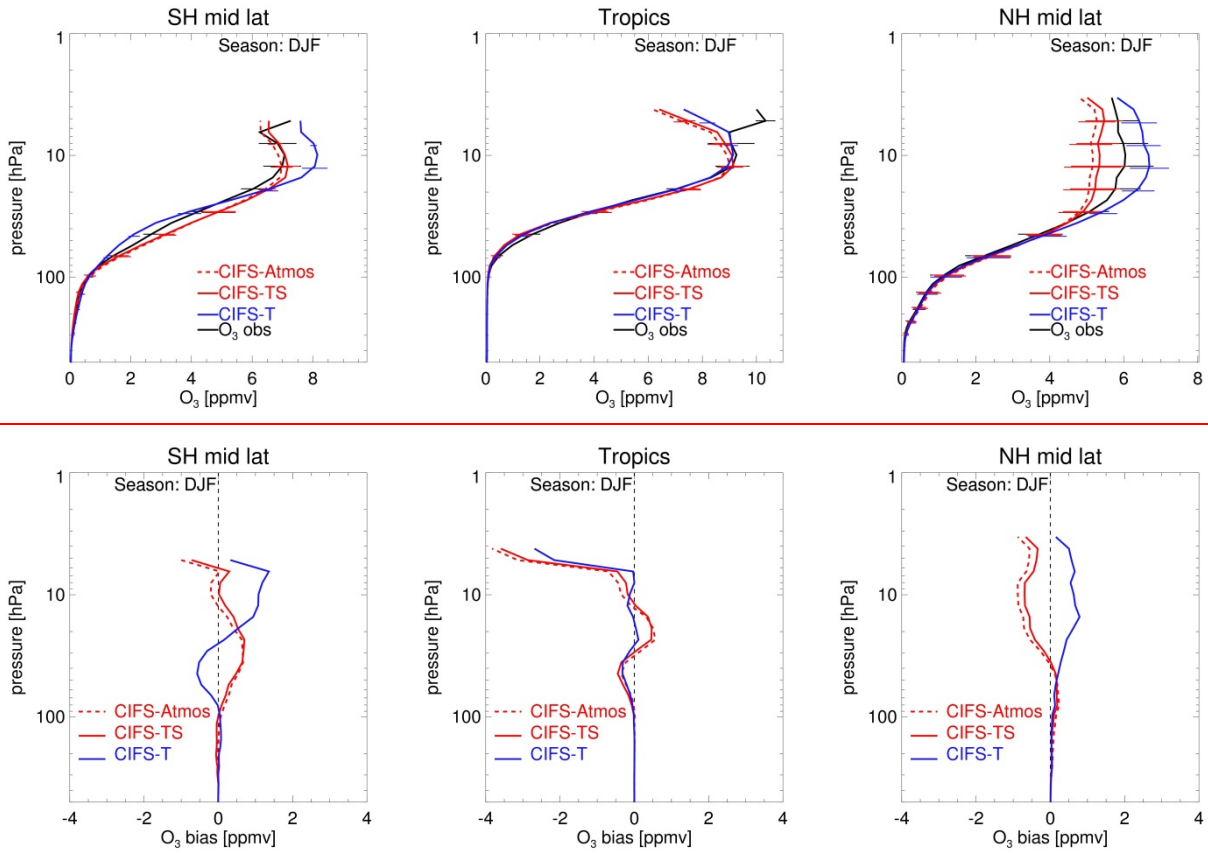


Figure 3. Top row: evaluation of ozone in units mPa against WOUDC sondes over NHSH mid-latitudes (60°N - 30°N , left), tropics (30°N - 30°S , middle) and SHNH mid-latitudes (30°S - 60°S , right) for December-January-February 2009 and 2010 in units mPappmv. Black: WOUDC observations, red dashed: C-IFS-Atmos, red solid: C-IFS-TS, blue: C-IFS-T. Error bars denote the 1-sigma spread in the models and observations. Bottom row: corresponding mean biases.

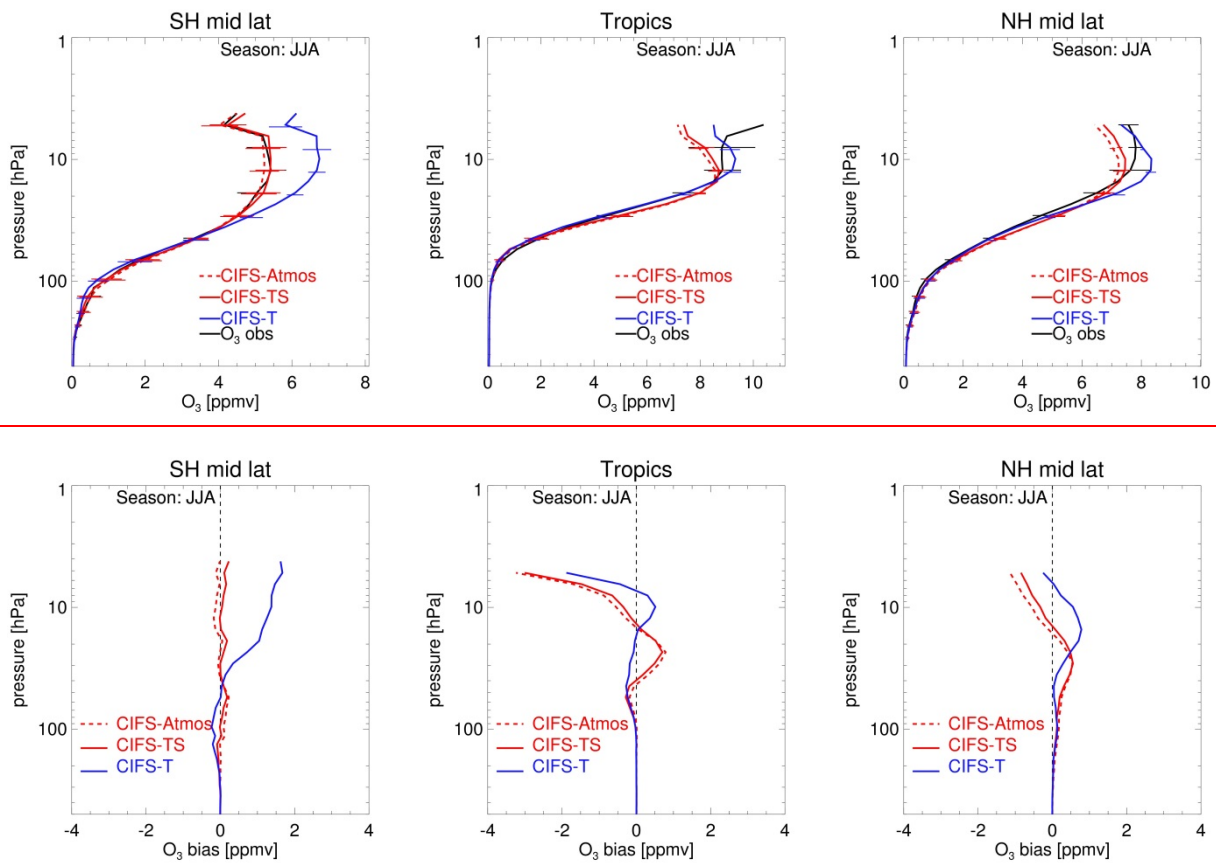


Figure 4. Same as Fig. 3, but for June-July-August 2009 and 2010.

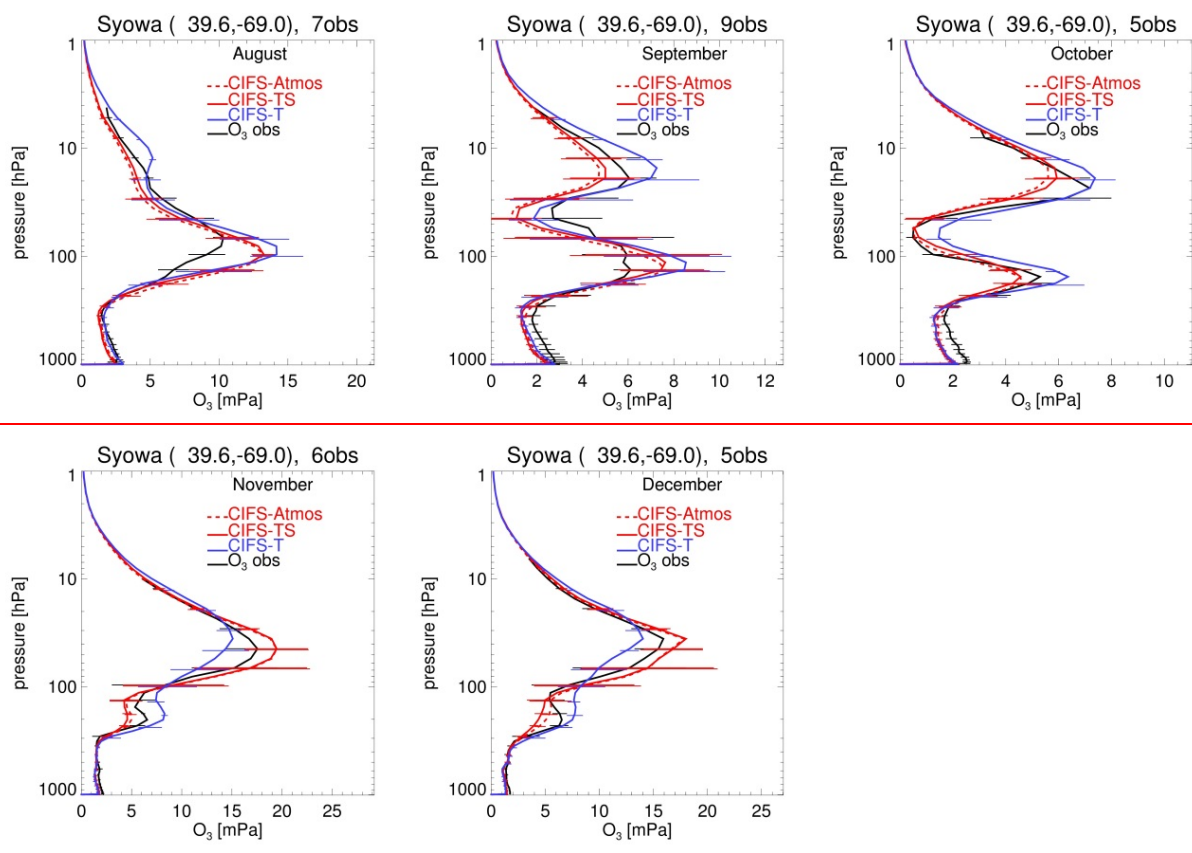
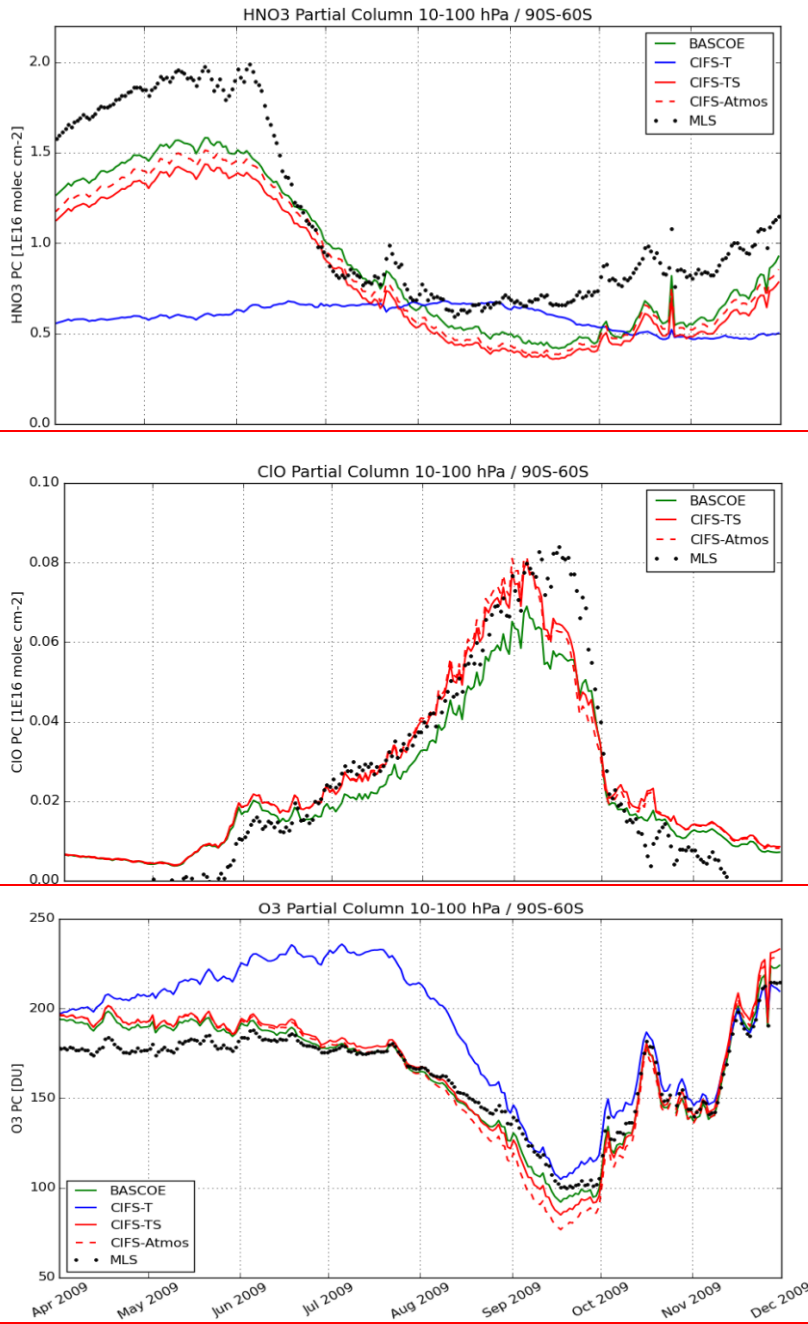


Figure 5. Evaluation of ozone in units mPa against WUDC ozone sondes at Syowa station during August-December 2009. Black: ozone sonde, ~~Red~~ red dashed: C-IFS-Atmos, red solid: C-IFS-TS, blue: C-IFS-T. Error bars denote the 1-sigma spread in the models and observations.



5

Figure 6. Daily averages of O₃ partial columns (10-100hPa) over the Antarctic (90°S-60°S), for the period April – November 2009 for HNO₃ (top), ClO (middle) and O₃ (bottom) against MLS observations.

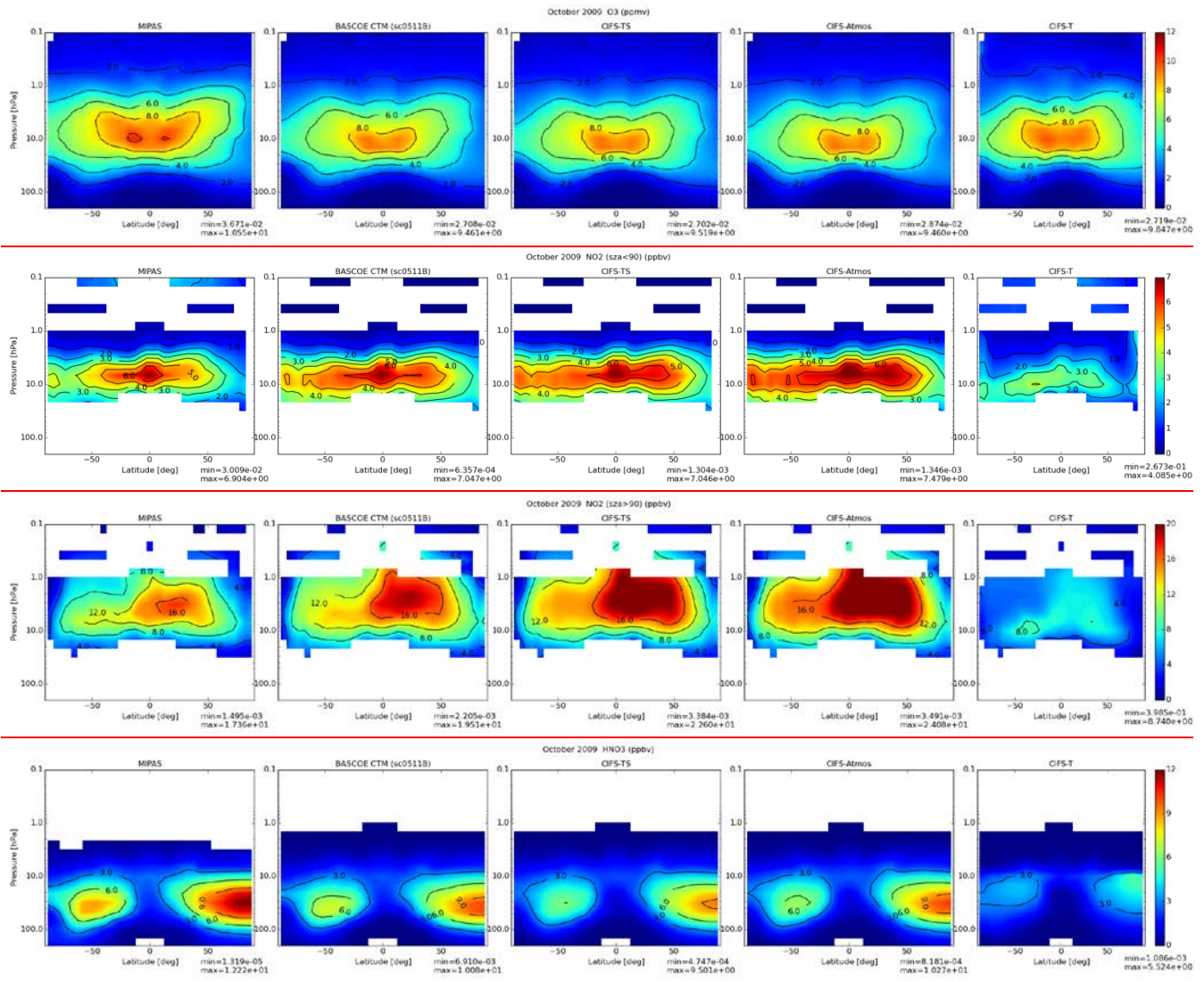
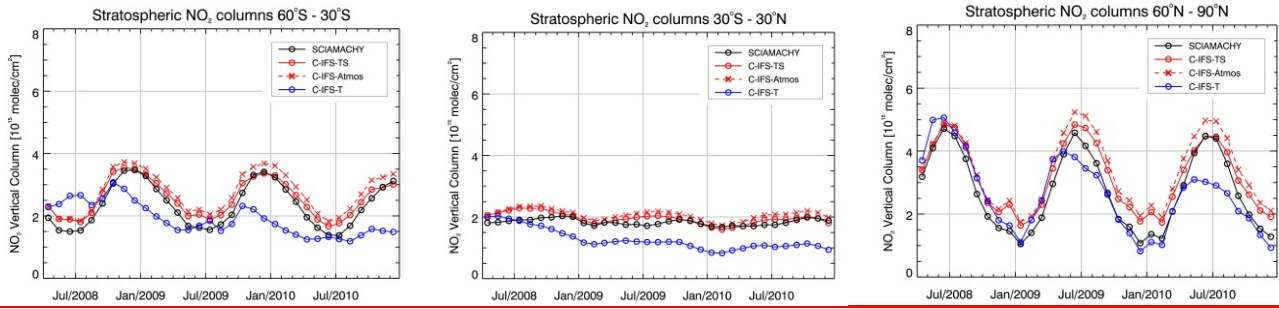
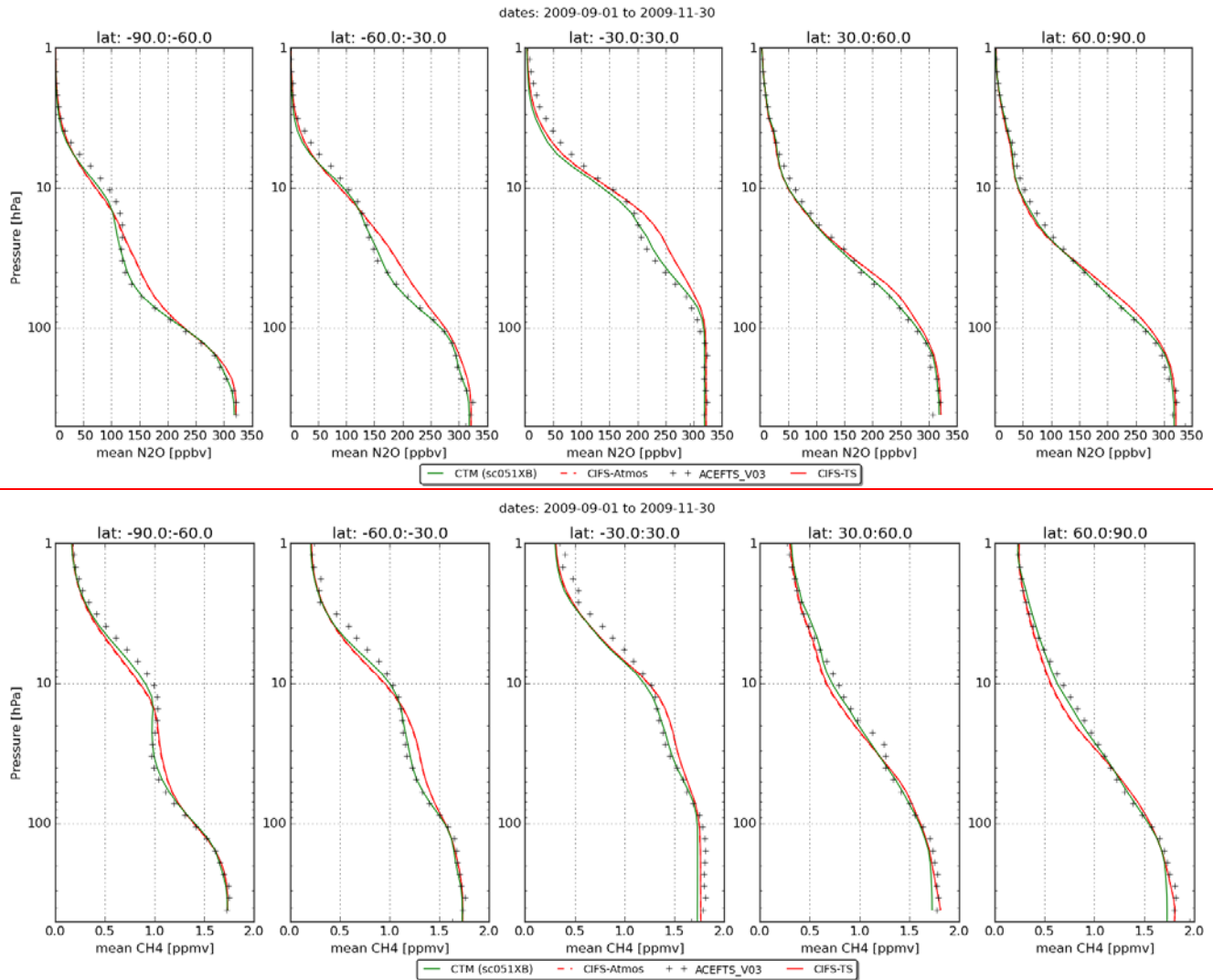


Figure 7. Zonal mean stratospheric O₃ (top row, units ppmv), daytime NO₂ (second row) and night-time NO₂ (third row) and HNO₃ (bottom row, all in units ppbv) for October 2009 using MIPAS observations (left first column) and co-located output of BASCOE-CTM (middle second), C-IFS-TS (third), C-IFS-Atmos (fourth) and C-IFS-TS (right fifth).



5 **Figure 78.** Time series of stratospheric total column NO₂ above the clean Pacific ocean (180°E-220°E) for April 2008 – Dec 2009 of C-IFS-TS against SCIAMACHY2010, in units 10¹⁵ molec cm⁻² for NH mid-latitudes (left), tropics (middle) and SH mid-latitudes (right). Black: SCIAMACHY observations, red dashed: C-IFS-Atmos, red solid: C-IFS-TS, blue: C-IFS-T.



5 **Figure 89.** Zonal mean profiles of stratospheric N₂O (top) and CH₄ (bottom) for September-October-November 2009 using ACE-FTS observations (black symbols) and co-located output of BASCOE-CTM (bluegreen lines), C-IFS-TS (red solid lines) and C-IFS-TSAtmos (red dashed lines). The zonal means are shown separately on five columns corresponding to the latitude bands 90°S-60°S, 60°S-30°S, 30°S-30°N, 30°N-60°N and 60°N-90°N, respectively.

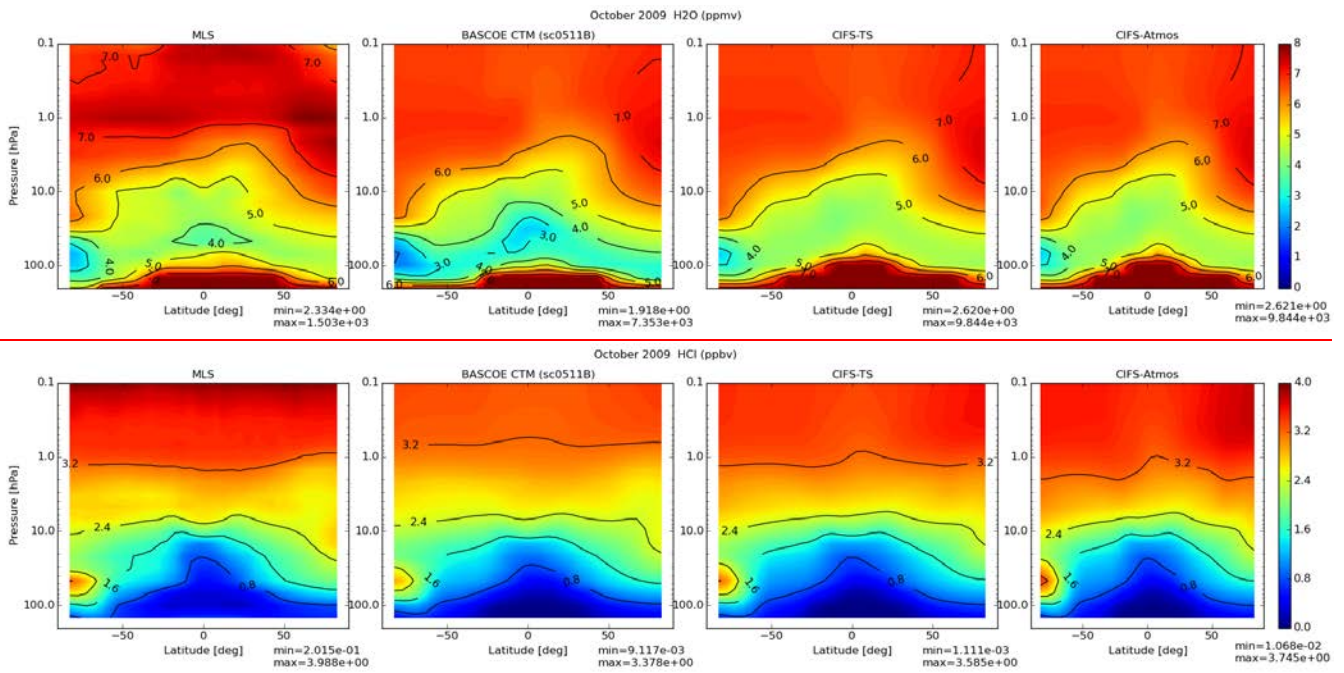


Figure 910. Zonal mean stratospheric H₂O (top, units ppmv) and HCl (bottom, units ppbv) for October 2009 using Aura/MLS observations (left first column) and co-located output of BASCOE-CTM (middle second), C-IFS-TS (third) and C-IFS-TS (right Atmos (fourth)).

5