

Brussels, 07 August 2020

Dear Editor,

I have now prepared the replies and changed the text according to the reviewers' comments.

Following the suggestions from both reviewers, we changed the structure of the paper merging the former Section 4 and Section 3, and added supplemental figures. In order to better interpret our results, we inserted new figures showing the Eliassen-Palm flux divergence across the datasets. We re-arranged the figures to follow the new structure of the text. We also improved the text by revising the structure of the introduction and scientific discussions, and by taking into account the additional references suggested by the reviewers.

Sincerely,

Daniele Minganti (on behalf of all co-authors).

Response to Reviewer#1 for: Climatological impact of the Brewer-Dobson Circulation on the N_2O budget in WACCM, a chemical reanalysis and a CTM driven by four dynamical reanalyses

Minganti et al., ACPD, 2020

We thank the reviewer for his in-depth review and useful comments. Following the reviewer's suggestion, we changed the structure of the paper and added supplemental figures. In order to better interpret our results, we inserted new figures showing the Eliassen-Palm Flux Divergence across the datasets. We also strove to improve the text by improving the structure and clarity of the Introduction and scientific discussions and by taking into account the additional references suggested by the reviewer. In our replies below the italic type is used for the reviewer's comments, the plain text for authors' answers and the bold type for the revised text in the manuscript.

Replies to general comments.

- *Although the paper contains some interesting material, which should be published, the manuscript itself could be significantly improved qualitatively in some parts (Introduction and results). Some paragraphs and sections are poor, therefore, they need to be revised by enhancing the discussion about the scientific content, the structure of results presentations as well as the wording to improve the quality of the paper.*

The Introduction was thoroughly revised and changed. We added new references (see list of references below) and enhanced the discussion by adding the relevant scientific content and removing (or reducing) when necessary. Regarding the results Section, we merged together the Section 3 with Section 4, and the manuscript was restructured in the following layout:

Section 3. Latitude pressure cross sections

Section 4. Climatological seasonal cycles

Section 4.1 Polar regions

Section 4.2 Middle latitudes

Section 4.3 Tropics

Section 5. Interannual variability of the seasonal cycles

Section 6. Summary and Conclusions

This new structure allowed to remove some purely descriptive parts in former Section 3, and add scientific content in former Section 4, i.e. the comparisons with relevant previous studies and physical interpretations of the differences/similarities in the different datasets. The layout of Figs. 5, 6 and 7 changed as well: we separated them by latitude bands (one figure for the polar regions, one figure for the surf zones and one for the tropics) in order to better follow the flow of the Section 4, and its subsections, and Section 5 of the revised manuscript. Fig. 9 was also modified according to this new structure, and it is described in the response to the comment below.

- *Particularly, the differences between WACCM and reanalyses and their possible physical causes could be significantly emphasized.*

The differences (or similarities) between WACCM and the reanalyses are better addressed now, as the pertinent studies comparing WACCM and the reanalyses or Aura MLS observations are considered and discussed. Furthermore, we expanded the Fig. 9 to include the northern and southern middle latitudes and polar regions, while the Tropics were moved in the supplement (Fig. S7). We separated it by latitude bands, i.e. one figure for the polar regions and one figure for the surf zones. We also added an additional row in each figure, showing the divergence of the Eliassen-Palm flux, as a measure of the forcing from resolved waves for all the considered datasets (BRAM2 is not shown because it uses the dynamical fields from ERAI). WACCM shows an underestimation of the divergence of the Eliassen-Palm flux, that allowed to enhance the discussion about the differences in the mid-stratospheric A_z and M_y . The former Sect. 3.3 was also improved in terms of scientific discussion thanks to the merging with the pertinent parts of Sect. 4, that were expanded and improved accordingly.

- *Appropriate references need to be used at the right places instead and properly discussed when necessary.*

Additional references were added throughout the revised manuscript. In the Introduction, we added Lin and Fu (2013); Fueglistaler et al. (2009); Birner and Bönisch (2011); Haynes et al. (1991); Rosenlof and Holton (1993); Newman and Nash (2000); Bönisch et al. (2011) for the description of the BDC. For the natural variability of the BDC we added Riese et al. (2012); Yang et al. (2014); Diallo et al. (2019, 2018); Salby and Callaghan (2005). In the part about trend studies of the BDC we added Fritsch et al. (2020). In the reanalyses and CTM description we included Gerber et al. (2010); Rao et al. (2015); Long et al. (2017); Waugh and Hall (2002); Chipperfield (2006); Monge-Sanz et al. (2012); Ménard et al. (2020). For the description of the chemical reanalysis BRAM2 we added Errera et al. (2008); Lahoz and Errera (2010). In Sect. 2 we included the suggested references In Sect. 3 we included Li et al. (2012) for the discussion of the seasonality of the BDC, we added also Roscoe et al. (2012) for the discussion of the differences in M_y above the Antarctic, and Ploeger and Birner (2016); Konopka et al. (2010) for the discussion of the lower branch of the BDC. In Sect. 4 we added Konopka et al. (2015); Gerber (2012) in connection to the divergence of the Eliassen-Palm flux, and Sato and Hirano (2019) for the discussion about A_z in the middle latitudes. In Sect. 5 we added the suggested reference Park et al. (2017) to discuss the inter-annual variability of N_2O .

Replies to Major points.

1. *The Introduction is poorly written, appropriate references are not properly used at some places, and some sentences are vague (not specific).*

The Introduction was deeply revised according to the comments of both reviewers. BDC. The description of the BDC was improved by describing its different branches, as well as how the wave breaking that leads to the BDC is quantified. The natural variability of the BDC is also discussed. The trend part was de-emphasized, as the current manuscript does not look at BDC trends (which will be the topic of a follow-up

study). The Introduction now states that it is important to study the climatological behaviour before trend studies.

CCM and WACCM. The sentences/paragraphs about CCMs and WACCM were put together into one paragraph.

Dynamical reanalyses, CTM and BASCOE. The former Introduction was too vague on these topics, therefore they were re-structured and clarified. The Introduction of dynamical reanalyses has been expanded to mention S-RIP. CTMs driven by reanalyses are described and related to BDC studies of Age of Air. Finally, the BASCOE CTM description was slightly expanded.

All the sentences about Chemical reanalyses and BRAM2 were also merged into one paragraph that starts with a general description of the added value of a chemical reanalysis, continues with the use of chemical reanalyses in TEM studies, and ends with a short description of BRAM2. We conclude the Introduction by summarizing the approach of the paper and providing its structure.

2. *It is important to show the contribution of the remaining terms such as the vertical mixing and horizontal advection in zonal mean as they are not negligible but just small than the vertical advection and horizontal mixing. This can be added as a supplement information.*

The contributions of A_y , M_z and $P - L$ for DJF and JJA are shown in the Supplement (Figs. S1 and S2) and appropriately mentioned in Section 3.

3. *As the calculation of w^* from CCM in CCMI project leads to a bias due to stratospheric shrinking (Eichinger & Shacha, 2020), this make wonder if the w^* from WACCM-CCMI calculated consistently with the w^* from BASCOE?*

The WACCM output we used includes only the basic meteorological variables, i.e. surface pressure, temperature and horizontal and vertical winds fields. w^* is re-calculated consistently across all datasets through equation 3b using the daily 3-D output of meridional and vertical wind velocity and temperature from WACCM and the dynamical reanalyses. These calculations are performed as recommended by the CCMI project (Chrysanthou et al., 2019).

4. *The scientific discussion of the figure 1 and 2 in the two paragraphs (234-239) is not clear and very poor. Differences/similarities in different terms and in different products are just omitted. All terms contributing to N_2O are not well identified and reported.*

Figures 1 and 2 are not meant for describing the differences/similarities between the datasets, rather for showing how the N_2O TEM budget and how the different terms balance each other depending on the latitude. The scientific discussion of the differences between datasets, and their possible physical causes, belongs to the following Sections. Yet, this was not stated in the manuscript, and understandably raised some confusion. We now state explicitly the purpose of these figures have shortened their description to focus on the physical meanings of the budget terms as follows:

Figs. 1 and 2 show the N_2O TEM budget terms at 15 hPa for all the datasets for the boreal winter (December-January-February, DJF mean) and summer (June-July-August, JJA mean) respectively. The 15 hPa level (around 30 km altitude) was chosen because large differences can be found between WACCM-CCMI, BRAM2, and the CTM runs at this level, and because the

dynamical reanalyses are not constrained as well by meteorological observations at higher levels (Manney et al., 2003). Figs. 1 and 2 aim to show how the dynamical and chemical terms of the budget balance each other to recover the tendency $\bar{\chi}_t$ at different latitudes. The discussion about the differences between the datasets, and their possible physical causes, are addressed in the next Sections.

The vertical advection term A_z shows how the upwelling contributes to increasing the N_2O abundances in the tropics and summertime mid-latitudes, and how polar downwelling contributes to decreasing the N_2O abundances in the winter hemisphere. The horizontal transport out of the tropics due to eddies, as represented by M_y , reduces the N_2O abundance in the tropical latitudes of the wintertime hemisphere, and increases the N_2O mixing ratio at high latitudes in the winter hemisphere. The other terms of the TEM budget are weaker than A_z and M_y : the meridional advection term A_y tends to increase the N_2O abundance in the winter subtropics and extratropics, while the vertical transport term due to eddy mixing, M_z decreases it over northern polar latitudes and the chemistry term $P - L$ shows that N_2O destruction by photodissociation and $O(1D)$ oxidation contributes to the budget in the tropics and also in the summertime hemisphere. All budget terms are weaker in the summer hemisphere than the winter hemisphere. Over the southern polar winter latitudes, the reanalyses deliver negative M_y that are balanced by large positive residuals, which implies a less robust TEM balance (Fig. 2). This is not the case with WACCM, where M_y tends to increase the N_2O abundance in the polar vortex. Such differences between the datasets are highlighted and discussed in the next sections.

5. *Why is there some differences in the vertical and horizontal mixing and residual terms in the SH between WACCM and reanalyses?*

The differences in M_y between WACCM and the reanalyses above the winter South Pole are discussed in Sects. 3 (fifth and sixth paragraphs) of the revised manuscript:

In the austral winter, over the Antarctic Polar cap and below 30 hPa, M_y agrees remarkably well in all datasets (Fig. 4). Closer to the vortex edge and above 30 hPa, the wintertime decrease of N_2O is mainly due to downwelling in WACCM-CCMI, while the reanalyses, especially BRAM2, show that the horizontal mixing plays a major role (Fig. 4). The impact of horizontal mixing on N_2O inside the wintertime polar vortex is not negligible (e.g. de la Camara et al., 2013; Abalos et al., 2016a), as Rossby waves breaking occurs there as well as in the surf zone. In contrast with the reanalyses, in WACCM-CCMI the M_y contribution is close to zero in the Antarctic vortex and maximum along the vortex edge (Fig. 4). This disagreement can be related to differences in the zonal wind: it is overestimated in WACCM above 30 km in subpolar latitudes compared to MERRA (Garcia et al., 2017) and the polar jet is not tilted equatorward as in the reanalyses (see black thin lines in Fig. 4, and Fig. 3 of Roscoe et al., 2012). Yet, the differences in M_y and A_z above the Antarctic in winter should be put into perspective with the relatively large residual terms that points to incomplete TEM budgets in the reanalyses (Fig. 4 and S4 right columns). Near the Antarctic polar vortex, the assumptions of the TEM analysis (such as small amplitude waves) are

less valid leading to larger errors in the evaluation of the mean transport and eddy fluxes (Miyazaki and Iwasaki, 2005).

Since the relative importance of the residual is considerable above the Antarctic in the reanalyses (Fig. 4), it is necessary to better understand its physical meaning. Dietmuller et al. (2017) applied the TEM continuity equation to the Age of Air (AoA) in CCM simulations. Computing the "resolved aging by mixing" (i.e. the AoA counterpart of $M_y + M_z$) as the time integral of the local mixing tendency along the residual circulation trajectories, and the "total aging by mixing" as the difference between the mean AoA and the residual circulation transit time, they defined the "aging by mixing on unresolved scales" (i.e. by diffusion) as the difference between the latter and the former. This "aging by diffusion", which can be related by construction to our residual term, arises around 60°S from the gradients due to the polar vortex edge. Even though we use a real tracer (N_2O), we find a qualitative agreement with this analysis based on AoA: our residual term is larger in regions characterized by strong gradients such as the antarctic vortex edge, and larger with dynamics constrained to a reanalysis than with a free-running CCM (see EMAC results in Fig. 1d by Dietmuller et al., 2017). We thus interpret the residual as the sum of mixing at unresolved scales and numerical errors (Abalos et al., 2017).

They are also discussed in Sect. 4.1 of the revised manuscript :

We now turn to the contribution from M_y . In the antarctic region, M_y is very different among the datasets during winter: in BRAM2 it contributes to the N_2O decrease during fall and winter, with the strongest contribution in July, but with the CTM simulations this contribution is two times weaker, while in WACCM-CCMI the horizontal mixing has almost no effect on N_2O (Fig. 6(e)). As already mentioned, the TEM analysis suffers from large residuals in the wintertime antarctic region. Yet we note that the disagreement between WACCM-CCMI and BRAM2 is significant, because in fall and winter the envelope of WACCM-CCMI realizations falls completely outside of the possible BRAM2 values when accounting for the residual. During the austral spring, the vortex breakup leads to an increased wave activity reaching the Antarctic (Randel and Newman, 1998), and mid-stratospheric M_y is in better agreement among all datasets compared to austral winter. Note that WACCM-CCMI exhibits large internal variability in this season (Fig. 6(e)).

and briefly mentioned in Sect. 2.4:

The BASCOE datasets have a coarser horizontal resolution than their input reanalyses (especially BRAM2; see Table 1). This affects the accuracy of the vertical and horizontal derivatives, with possible implications for the residual.

Again, Figs. 1 and 2 are not meant for the discussion of differences between datasets (this is left to Sects. 3 and 4), but only for showing the TEM budget and pave the way for the following discussion.

6. *So far, ERAi is the reanalysis, which shows a closer pattern changes in the last decade of trace gases closer to observations, including O_3 , HCl , etc... but it's not shown in figure*

3 and 4. A similar panel should be added in the supplement and discussed as well as the horizontal advection and vertical mixing term.

The full N_2O TEM budget obtained with ERAI and MERRA, for DJF and JJA, are now shown in Figs. S3 and S4 of the supplement.

7. *The scientific discussion of the figure 3 and 4 related to summer and winter variations of advective and mixing terms is poor and can be improved as well as linked to age spectrum/age of air published articles (Li et al., 2012, Diallo et al, 2012, Ploeger and Birner, 2016).*

The summer and winter variations are now addressed through the seasonality of the deep branch of the BDC on the TEM budget (first paragraph of the revised section 3):

Large differences arise in the dynamical terms of the budget between summer and winter for both hemispheres in the extratropics. The strong seasonality of the deep branch of the BDC and of the transport barriers are the causes of these differences, as also shown for the seasonal variations of the Age of Air spectrum (Li et al., 2012).

and through the differences between the shallow and deep branches of the BDC, which are discussed the the third paragraph of revised section 3:

In the lower stratosphere, A_z shows the contribution of the residual advection by the shallow branch of the BDC to the N_2O abundances in the winter and summer hemispheres. The two-cell structure, consisting in upwelling of N_2O in the subtropics and downwelling in the extratropics, consistently agrees across all datasets.

... and in a new paragraph at the end of section 3:

In the summertime lower stratosphere, we note a stronger contribution of M_y to the N_2O abundances above the subtropical jets in both hemispheres and for all datasets compared to higher levels in summer (Figs. 3 and 4 middle columns). This behavior is consistent with calculations of the effective diffusivity and age spectra (Haynes and Shuckburgh, 2000; Ploeger and Birner, 2016). It is due to transient Rossby waves that cannot travel further up into the stratosphere due to the presence of critical lines, i.e. where the phase velocity of the wave matches the background wind velocity, generally leading to wave breaking (Abalos et al., 2016b). In particular, above the northern tropics during the boreal summer (Figs. 4, S2 and S4), the horizontal mixing is primarily associated with the Asian monsoon anticyclone, and causes a decrease in N_2O (Konopka et al., 2010; Tweedy et al., 2017). In the lower stratosphere, the contributions from M_y combine with that from A_z in the total impact of the shallow branch of the BDC on N_2O all year round (Diallo et al., 2012).

8. *It would be very instructive to reproduce the figure 8 in Randel et al, 1994 which will compare WACCM ensemble mean versus all reanalysis means.*

We reproduced it for DJF for the WACCM ensemble and the reanalysis ensemble mean, and they are shown in the Supplement (Fig. S5 and Fig. S6 respectively). This is mentioned in the second paragraph of revised section 3:

We also reproduced the results of Randel et al. (1994, Fig. 8) for the WACCM-CCMI multi-model mean and the reanalysis mean in DJF (Figs S5 and S6 respectively). The WACCM-CCMI and the reanalysis means agree with the Community Climate Model version 2 of the early 1990's with regard to the general pattern of the TEM terms, but both deliver stronger contributions, especially the reanalyses mean.

9. *The results discussed in "climatological seasonal cycles" section is not clear. It is missing a clear structural organization and not all panels are discussed. Thus, it is very difficult to follow. One suggestion would be to organize the discussion by latitude bins and by term: "In the tropic, ...", "In the mid-latitudes, ..." and "In the polar region, ..."*

We agree with the comment from the reviewer. As stated before, the Section "climatological seasonal cycles" was merged with the Discussion section and divided in three subsections: Polar regions, middle latitudes and Tropics. This allows the structured discussion by latitude bands that the reviewer suggested.

10. *Is there any physical explanation of the spread in the tropical and mid-latitudinal N_2O vmr in figure 8? What is the contribution of different QBO representation and modulation of the upwelling to the differences?*

Regarding the tropical regions, the differences between the datasets are discussed in more detail, and in the revised section 5 we now illustrate the contribution of the QBO on WACCM and BRAM2as follows:

The inter-annual variability of the N_2O mixing ratio in both southern and northern tropics depends considerably on the dataset (Figs. 12(a) and (b)). WACCM-CCMI and the BASCOE reanalysis of Aura MLS show very similar variabilities, especially in the southern Tropics. Since the QBO is the major source of variability in the tropical stratosphere (Baldwin et al., 2001), this confirms an earlier comparison that showed a good agreement between the WACCM model and MLS observations in the middle stratosphere in terms of the inter-annual variability of N_2O due to the QBO (Park et al. 2017). Among the CTM simulations, ERAI succeeds to deliver $\sigma(\bar{X})$ as large as BRAM2 and WACCM-CCMI in the southern tropics, but not in the northern tropics.

As stated in the last paragraph of the conclusions, a detailed study of the impact of the QBO on N_2O or the TEM quantities does not belong to this paper, but to a follow-up study that will investigate inter-annual changes.

11. *The results' discussion in section 3.3 are also poor. Need to be improved.*

The Sect. 3.3 was merged with the relevant parts of Sect. 4, to become Sect. 5 in the revised manuscript. The text is less descriptive and the scientific discussion is improved, using existing and new references.

12. *The main issue of the paper is results part is poor. The scientific content of the figures are better discussed in the discussion part than in the main part of the paper. This gives to a reader the feeling that he is reading twice the same article. It would be great to put necessary elements in the main part of the manuscript when commenting the figures. This could be done by moving the information in the Discussion session to where it belongs for each figure in the main text.*

Indeed, the reviewer is right. The results part (Sect. 3) was merged with the Discussion (Sect. 4), and new subsections were created (see above). This allows to enhance the scientific discussion and cut the descriptive parts that were not necessary.

13. *The differences in the tropics, mid-latitude and high latitude need to be discuss clear by taking into account the difference in the QBO. Showing a tropical mean cross-section (5S-5N) of N_2O vmr from reanalysis means versus WACCM ensemble means as time series over the dataset period will be great for discussion and for illustration of the possible differences related to QBO (timing, amplitude, phases, ...). For insight, please see Park et al. 2017 (fig 9 and 12). In addition for the polar region discussion, it would be very instructive too related the discussion to Randel et al, 1994, where a case study of SSW have been illustrated using N_2O budget.*

As announced in the title, the scope of the paper is limited to climatologies. Time series will be investigated in a follow-up study about inter-annual changes. Thus, we decided not to show the suggested time series plot of the reanalysis mean vs WACCM mean. A reference to the work from Park et al. (2017) was added to the third paragraph of revised Section 5:

WACCM-CCMI and the BASCOE reanalysis of Aura MLS show very similar variabilities, especially in the southern Tropics. Since the QBO is the major source of variability in the tropical stratosphere (Baldwin et al., 2001), this confirms an earlier comparison that showed a good agreement between the WACCM model and MLS observations in the middle stratosphere in terms of the inter-annual variability of N_2O due to the QBO (Park et al., 2017)

as well as a connection to the SSW case study in Randel et al., 1994 for the Arctic (second paragraph of Section 5):

Above the Arctic, M_y and A_z are most variable during winter, reflecting the frequent disruptions of the northern polar vortex by sudden stratospheric warmings (SSWs, Butler et al., 2017). A case study of the effect of a SSW on the N_2O TEM budget showed that A_z and M_y contribute more to this budget during the SSW event than in the corresponding seasonal mean. Thus, the large wintertime variability of A_z and M_y is explained by the occurrence of seven major SSWs detected in the reanalyses for the 2005-2014 period (Butler et al., 2017).

Replies to minor points

1. *Page 1, line 1-2, please rephrase the sentence it sounds wrong "from the well-mixed tropical troposphere to the polar stratosphere" and "..., chemistry, ozone distribution and recovery"*

The sentence was rephrased:

The Brewer-Dobson Circulation (BDC) is a stratospheric circulation characterized by upwelling of tropospheric air in the Tropics, poleward flow in the stratosphere, and downwelling at mid and high latitudes, with important implications for chemical tracers distribution, stratospheric heat and momentum budgets and mass exchange with the troposphere.

2. Page 2, line 33-34, the BDC is the stratospheric circulation and it is not a tropospheric circulation. Please rephrase this sentence "The stratospheric circulation is mainly characterized by the Brewer Dobson Circulation.... from the troposphere..."

The sentence was rephrased:

The Brewer-Dobson Circulation (BDC, Dobson et al., 1929; Brewer, 1949; Dobson, 1956) in the stratosphere is characterized by upwelling of tropospheric air to the stratosphere in the Tropics, followed by poleward transport in the stratosphere and extratropical downwelling.

3. Page 2, line 38, please replace "The BDC is generated by Rossby waves propagating" by "The BDC is driven by Rossby wave breaking into ..."

Done.

4. Page 2, line 39, please rephrase "This departure"

The part is rephrased as follows:

...away from its radiative equilibrium. This is balanced by a meridional...

5. Page 2, line 41-43, note that the residual circulation can be split into 3 branches: transition, shallow and deep branch for more detail see Lin & Fu (2013). Please improve the discussion by including the relevant previous studies: Haynes et al., 1991, Rosenlof and Holton, 1993; Newman and Nash, 2000; and Birner and Bnich (2011). Please add also the term "breaking" after "synoptic-scale" and "Rossby" and replace "generate/generated" by "drive/driven" in the whole manuscript. The paragraph (line 38-43) is very poor and need to be improve, and also the natural variability modulations, including QBO and ENSO, of the BDC branches, trace gas transport need to be mentioned see Yang et al, 2014; Baldwin et al 2002, Tweedy et al., 2017, and Diallo et al, 2018, 2019.

The discussion has been improved and the suggested references added as follows:

The BDC is driven by tropospheric waves breaking into the stratosphere (Charney and Drazin 1961), which transfer angular momentum and force the stratosphere away from its radiative equilibrium. This is balanced by a poleward displacement of air masses, which implies tropical upwelling and extra-tropical downwelling (Holton, 2004). The residual circulation can be further separated in three branches: the transition, the shallow and the deep branch (Lin and Fu, 2013). The transition branch encompasses the upper part of the transition layer between the troposphere and the stratosphere (the tropical tropopause layer, Fueglistaler et al., 2009). The shallow branch is an all year-round lower stratospheric two-cell system driven by breaking of synoptic-scale waves, and the deep branch is driven by Rossby and gravity waves breaking in the middle and high parts of the stratosphere during winter (Plumb, 2002; Birner and Bonisch, 2011). The contributions of different wave types to the driving of the BDC branches has been quantified using the downward control principle, which states that the poleward mass flux across an isentropic surface is controlled by the Rossby or gravity waves breaking above that level (Haynes et al., 1991; Rosenlof and Holton, 1993), and using eddy heat flux calculations as an estimate of the wave activity from the troposphere (e.g., Newman and Nash, 2000).

6. *Page 2, line 50, Please rephrase this sentence "Simulations by Chemistry Climate Model (CCM)..." by "Chemistry Climate Model (CCM) simulations..."*

Done.

7. *Page 2, line 54, the references in the sentence "Observations of long-lived chemical tracers (e.g. H₂O, N₂O) are often used to derive estimates of the BDC..." is not the appropriate one. Please use the right articles, which examined BDC from H₂O, N₂O, like e.g. Hegglin et al 2014; Andrews et al. 2001; Kracher et al. 2016; Schoeberl et al, 2008 and H. K. Roscoe, 2006.*

As stated in the reply to major point 1, the part of the Introduction dealing with long-term trends was de-emphasized because this manuscript is about the climatology of the BDC, not its trends. In the revised manuscript, studies of BDC trends are introduced with one paragraph citing a few model papers and some observational papers including some of those suggested here by the reviewer.

8. *Page 2-3, line 55-56, the sentence is not correct because the balloon observation trend in the whole NH but only for the deep branch. Please be specific.*

The sentence was corrected as follows:

... but balloon-borne observations of SF₆ and CO₂ in the Northern Hemisphere (NH) middle latitudes show a non-significant trend of the deep branch of the BDC in the past decades (Engel et al., 2009, 2017).

9. *Page 3, line 58, please "Stiller et al. 2012" among the early papers using SF₆ satellite observation to estimate decadal BDC trends.*

The text was changed and the reference added.

10. *Page 3, line 59-60, please cite Diallo et al, 2012 and Monge-Sanz et al 2012 among the early papers using reanalysis and observation to assess BDC changes. Add Ploeger et al., 2019 as well.*

As the paragraph about the BDC changes was reduced, this sentence was removed.

11. *Page 3, line 59-60, the whole sentence "A number..." seems a bit off here as it is break the continuity from the previous session and mixes again reanalysis, climate model & observations while mainly talking about BDC derive from observations and its limitation.*

The reviewer is right, and the sentence was removed from the manuscript.

12. *Page 3, line 64-65, CLaMS is a Lagrangian transport model driven with reanalyses not a climate model, therefore, the citation of Ploeger et al 2019 is out of place here. Please move it to line 59-60.*

The citation to Ploeger et al., 2019 was moved to the paragraph of the Introduction that explains CTM studies about AoA:

Recent intercomparisons showed that the AoA depends to a large extent on the input reanalysis, both using the kinematic approach (Chabrillat et al., 2018) and the diabatic approach (Ploeger et al., 2019).

13. *Page 3, line 66, this "nitrous oxide (N_2O)" is already mentioned in page 2, line 53 but online define now.*

The first occurrence of the nitrous oxide formula is now at Page 1 line 5:

Since the photochemical losses of nitrous oxide (N_2O) are well-known,....

and the "nitrous oxide (N_2O)" at Page 3 line 75 is replaced by " N_2O ":

In this study we use N_2O as ...

14. *Page 3, line 77, please be specific here by replacing "from several reanalysis datasets." With "from the Chemical ObsErvation (BASCOE) Chemistry-Transport Model (CTM) driven by several reanalysis datasets (Chabrillat et al., 2018)."*

The paragraph was rearranged, we now mention the BASCOE CTM and the reanalyses used to drive it in a separate paragraph:

Here we use the same CTM as for the kinematic AoA study, i.e. the Belgian Assimilation System of Chemical ObsErvation (BASCOE) CTM. Observations of another long-lived stratospheric tracer, HCFC-22, were recently interpreted with WACCM and BASCOE CTM simulations, showing the interest of this model intercomparison (Prignon et al., 2019). In order to contribute further to the S-RIP BDC activity, four different dynamical reanalyses are used here to drive the BASCOE CTM simulations, compute the N_2O TEM budget and compare its components with the results derived from WACCM. Namely we consider: the European Centre for Medium-Range Weather Forecasts Interim Reanalysis (ERA-Interim, Dee et al., 2011), the Japanese 55-year Reanalysis (JRA55, Kobayashi et al., 2015), the Modern-Era Retrospective analysis for Research and Applications version 1 (MERRA Rienecker et al., 2011), and version 2 (MERRA2 Gelaro et al., 2017).

15. *Page 3, line 77, remove "Dynamical" and replace by "Reanalysis products"*

Done.

16. *Page 3, line 81, move "Fujiwara et al., 2017; Cameron" after "models".*

Done, and the reference to Fujiwara et al., 2017 was removed:

Reanalyses are made using different assimilation methods and forecast models (Cameron et al., 2019), and

17. *Page 3, line 86-88, please citations for each reanalysis product (e.g. Dee et al. 2011, Kobayashi et al 2015, Rienecker et al. 2011, Gelaro et al., 2017).*

The citations were added both in the Introduction (see reply to minor point 14 above) and also in a new Table 1 that provides an overview of all the datasets used in this study.

18. *Page 4, line 97-99, the description section 3.1, 3.2 and 3.3 could be combine into section 3 to avoid redundant description.*

Thanks to the new structure of the manuscript, the description of the Sections does not include subsections anymore:

In Section 3 we analyse the seasonal mean patterns of the TEM N_2O budget in each dataset and their differences. Sections 4 and 5 investigate respectively the mean annual cycle and the variability of the N_2O TEM budget terms, with

a focus on the differences between the datasets. Section 6 concludes the study with a summary of our findings and possible future research.

19. Page 4, line 102, "Data and methods". There is no "s" to "method".

Done.

20. Page 4, line 107-108, please precise what you did "ran" by yourself or "downloaded/use" existing simulations. Rephrase this sentence "We ran one realization of the public version of WACCM (hereafter WACCM4, Marsh et al., 2013), that we downloaded at https://svn-ccsm-models.cgd.ucar.edu/cesm1/release_tags/cesm1_2_2cesm1_2_2."

The sentence was rephrased as:

We ran one realization of the public version of WACCM (hereafter WACCM4, Marsh et al., 2013), with a similar setup (e.g. lower boundary conditions) as the CTM experiments; the source code of WACCM4 is available for download at https://svn-ccsm-models.cgd.ucar.edu/cesm1/release_tags/cesm1_2_2cesm1_2_2.

21. Page 4, line 104, replace "trasport (see Sect. 4)." by "transport (see Sect. 4 for detailed analysis)". The same remark for "dataset (see Sec. 2.3)".

Done.

22. Page 4, line 119, the "... (Lin, 2004)." is not correctly reported in the reference.

The reference was corrected.

23. Page 5, line 124-126, please replace the existence by these ones "In this study, the considered WACCM versions are not able to internally generate the Quasi-Biennial Oscillation (QBO, see e.g. Baldwin et al., 2001). Thus, the QBO is forcing (nudged) by a relaxation of stratospheric winds to observations in the Tropics (Matthes et al., 2010)."

Done.

24. Page 5, line 130, add coma after "In addition"

Done.

25. Page 5, line 137-138, please rephrase this sentence "The transport module requires on input only the surface pressure and horizontal wind fields from reanalyses, as it relies on mass continuity to derive vertical mass fluxes"

The sentence was rephrased:

Chabrillat et al. (2018) explain in detail the preprocessing procedure that allows the BASCOE CTM to be driven by arbitrary reanalysis datasets, and the set-up of model transport. As usual for kinematic transport modules, the FFSL scheme only needs the surface pressure and horizontal wind fields from reanalyses as input, because it is set on a coarser grid than the input reanalyses, and relies on mass continuity to derive vertical mass fluxes corresponding to its own grid.

26. Page 5, line 135, please add a comma before "which"

Done.

27. *Page 5, line 139-141, please add a comma after "but" and "In this way".*

Done.

28. *Page 5, line 147, please rephrase this sentence "For this work the BASCOE CTM provided daily mean outputs over the 2005-2014 period as for the WACCM experiment."*

The sentence was rephrased:

As for the WACCM experiment, we used the daily mean outputs from the BASCOE CTM over the 2005-2014 period.

29. *Page 5, line 150, for analogy to the tow previous model description, this part "The TEM diagnosis is also applied to N₂O" is out of place here. First describe the BRAMS2 and then...*

We do not mention the TEM N₂O budget at that stage anymore, and the sentence was rephrased:

BRAM2 is the BASCOE Reanalysis of Aura MLS, version 2, which covers the period....

30. *Page 6, line 164, please remove this "Livesey, in preparation"*

Done.

31. *Page 6, line 170, please the sentence after "temperatures," and start a new one.*

There is now a period after the temperatures (definition of $M^{(z)}$), and the new sentence starts with the definition of v^* and w^* :

$$M^{(z)} \equiv \dots$$

v^* and w^* are...

32. *Page 6 line 180, please add a comma after "Hence"*

Done.

33. *Page 7, line 195, replace "hence retaining" by "while conserving"*

The sentence was rephrased:

Before any TEM calculation all the input fields are interpolated to constant pressure levels from the hybrid-sigma coefficients, that retain the same vertical resolution as the original vertical grid of each dataset (Table 1).

34. *Page 7, line 201, please a comma before "which"*

Done.

35. *Page 7, line 202, add a comma after "Furthermore in WACCM"*

Done.

36. *Page 7, line 206, replace "timestep" by "time step"*

Done.

37. *Page 7, line 205-207, this sentence can combine to one concise sentence avoid the use of "This". Please rephrase "Finally, the daily mean fields are interpolated from their native hybrid-sigma levels to constant pressure levels prior to the TEM analysis. This could lead to numerical errors in the lower stratosphere."*

The sentence was rephrased:

The daily mean fields are interpolated from their native hybrid-sigma levels to constant pressure levels prior to the TEM analysis, leading to numerical errors in the lower stratosphere.

38. *Page 7, line 207, please add a comma after "For WACCM-CCMI"*

Done.

39. *Page 7, line 211, the term "realistic" does not fit well with second part of the sentence "but". What lead to the different representation of large-scale transport is not the fact that the temperature and winds are realistic but because the reanalyses have some differences in wind and temperature. Please see Fig. 5 in Tao et al 2019. You can rephrase the existing sentence as following "The four dynamical reanalyses used in this study provide comparable (consistent) temperature and winds in the stratosphere, but can also lead to a different representation of large-scale transport (e.g. Chabrillat et al., 2018) due to the biases in the temperature and wind fields (Kawatani et al., 2016; Tao et al., 2019). "*

The sentence was rephrased as suggested.

The four dynamical reanalyses used in this study provide overall consistent temperature and winds in the stratosphere, but can lead to a different representation of large-scale transport (e.g. Chabrillat et al., 2018) due to the biases in the temperature and wind fields (Kawatani et al., 2016; Tao et al., 2019). Note that the TEM quantities are not directly constrained by observations, especially the upwelling velocity \bar{w}^* , that can vary considerably in the dynamical reanalyses, as it is a small residual quantity (Abalos et al., 2015).

40. *Page 7, line 213, add a comma after "In the rest of the paper"*

Done.

41. *Page 7, line 214, replace "BASCOE reanalysis BRAM2" by either "BASCOE reanalysis" or "BRAM2 product"*

"BASCOE reanalysis BRAM2" was replaced with "BRAM2 product".

42. *Page 8, line 217, add a comma after "n Figs. 1 and 2"*

The sentence was rephrased:

Figs. 1 and 2 show the....

43. *Page 8, line 219, replace "the strongest" by "stronger ...". In addition DJF & JJA can be term as boreal winter and summer season.*

The whole sentence was removed from the manuscript.

44. *Page 8, line 223, regarding the Figure 1, please replace "time der" by " X_t " or "tendency" and redo the figure that the M_y (green) appear properly in all panels. The fact tendency, residual & horizontal bold line are all in black make different components hard to distinguish. Please fix it.*

"time der" was replaced by " X_t ". The y-scale was widened so M_y could appear properly in the all panels. The horizontal bold line (i.e. the zero line) was removed.

45. *Page 8, line 225, please rephrase "In the northern tropics the N_2O decrease due to horizontal mixing is clearly". Also the tendency term of WACCM-CMM is near zero in the NH. I don't see any directional sign therefore the sentence does not match what the panel is showing. Maybe for WACCM panel you can change the vertical scale and note that in the figure caption that the vertical scale of WACCM is different from the reanalyses.*

The whole sentence was indeed confusing. The discussion of Figs. 1 and 2 was reduced because it was repetitive and it aims to describe only the most important points.

46. *Page 8, line 225-226, the interpretation in this sentence is wrong "In the northern tropics ... sufficient to do so." Overall the A_y term is consistent between WACCM and the reanalyses at all latitudes.*

The discussion about Figs. 1 and 2 was changed, see our reply to major point 4.

47. *Page 8, line 226-229, please rephrase this sentence "At the higher latitudes the main terms contributing to the N_2O TEM budget are the positive horizontal mixing term in the N_2O increase, and the negative vertical advection and vertical mixing terms for the N_2O decrease in all the datasets, with negligible contributions from the other terms." It's not clear and poor.*

The discussion about Figs. 1 and 2 was changed, see comment above.

48. *Page 8, line 230-231, what about the except of MERRA where the horizontal advection is comparable to Production-lost term as well as the JRA " A_y " increase in the NH. Here also the discussion is poor.*

As mentioned before, Figs. 1 and 2 are not meant to discuss differences in the datasets (this is left to the next Sections), but only to show how the terms of the TEM budget balance each other. The discussion about Figs. 1 and 2 was changed, see comments above.

49. *Page 8, line 232, this statement is not true for the reanalysis "a general balance between the M_y and A_y " because for some reanalysis the residual and P-L term are as large as the " M_y ".*

We agree with the reviewer, but, again, we do not wish to compare the datasets at this point of the manuscript. The discussion about Figs. 1 and 2 was changed, see comments above.

50. *Page 8, line 233-234, the term " A_y " also contribute in the mid lat.*

The discussion about Figs. 1 and 2 was changed, see comments above.

51. *Page 8, line 235, please replace "is affected mostly" by "is mostly affected..."*

As the paragraph was largely changed, this is not included anymore.

52. *Page 8, line 235-239, Why their differences in the vertical and horizontal mixing and residual terms in the SH between WACCM and reanalyses is not discussed here?*

As mentioned above, Figs. 1 and 2 are meant only for illustrating the various terms of the TEM budget, and how they balance each other at different latitudes. This is now explicitly stated in the discussion of Figs. 1 and 2. The differences between datasets are discussed in detail in Sect. 3, 4 and 5 of the revised manuscript.

53. *Figure 3 and 4, it would be good to add the arrows indicating the residual mean circulation v^* and w^* as well as the zero zonal mean wind but remove the full zonal mean wind fields.*

We thank the reviewer for the comment, but we chose not to show the residual advection and not remove the full zonal wind because we think that the full zonal mean wind is useful for showing the polar jet, as it is related to the discussion of Fig. 4, and the addition of the arrows of v^* and w^* would make the panel rather difficult to interpret.

54. *Page 9, line 245, add a comma after "CCMI"*

Done.

55. *Page 9 line 250, add a comma after "During the DJF season" and before "but"*

Done, and we replaced "DJF season" by "boreal winter".

56. *Page 9 Why the colorbars in figures 3 and 4 have a different scales?*

We now use the same color scale $[-2, 2]$ ppbv/day for both the figures.

57. *Page 9 Why the differences between summer and winter term are not discussed?*

Those differences are discussed in the revised manuscript (Sect. 3). See our reply above to major point 7.

58. *Page 9, line 259, add a comma after "In the JJA season"*

Thanks to the new manuscript structure, this paragraph was removed.

59. *Page 9, line 259-267, why the large "My" term from BRAM2 is not mentioned?*

This is now discussed in the fifth paragraph of section 3:

In the austral winter, over the Antarctic Pole and below 30 hPa, M_y agrees remarkably well in all datasets (Fig. 4). Closer to the vortex edge and above 30 hPa, the wintertime decrease of N_2O in the middle stratosphere is mainly due to downwelling in WACCM-CCMI, while the reanalyses, especially BRAM2, show that the horizontal mixing also plays a major role (Fig. 4).

60. *Page 9, line 262, replace "very positive values" by "large positive values"*

With the new manuscript structure, this paragraph was removed.

61. *Page 9-10, regarding the figures 5 and 6, over the whole manuscript you have always discussed NH and then SH. Why then starting with the SH when it comes to figure 5 and 6? It would be good to keep a fix structure.*

In the revised manuscript, the discussion of the Figs. 5 and 6 (now merged into Fig. 5) is separated in subsections organized by latitude band (Polar region, middle

latitudes, Tropics), rather than by hemisphere. This allows to better describe similarities/differences between the hemispheres, and to avoid repetition whenever the patterns are similar.

62. *Page 10, line 270-271, the affirmation regarding "My" and "Az" terms showing maxima at 15hPa is wrong because the "Az" terms maximum is around 5 hPa for WACCM-JRA55 and a bit high for the others reanalyses in both seasons DJF & JJA figures. Your previous argument was that it's level of better assimilation of meteorological observations according to Manney et al. 2003. Please correct that.*

The sentence was removed as the same statement was already in Sect. 2.4.

63. *Page 9, line 274, add a comma after "For WACCM-CCMI"*

Done.

64. *Page 9, line 275-281, this information should move to the caption. In addition, BRAM2 is a BASCOE reanalysis, while the other reanalysis products (ERAi, JRA55, MERRA) use well-established assimilation system constrained with observations. I don't see why BRAM2 is consider here as the "truth"?*

The part from "The color codes..." until "remain cautious" was moved to the caption of Fig. 5 of the revised manuscript. Regarding BRAM2, it is constrained by N_2O observations, which is not the case for the CTM nor for any of its 4 driving reanalyses. We do not consider BRAM2 as the "truth" more than we would consider an observational dataset to be the "truth". A whole paragraph explains this in section 2.4, both in the ACPD and revised versions, with the revised version stating:

In the rest of the paper, we will assume that the BRAM2 product provides the best available approximation of the TEM budget for N_2O , at least where the residual is smaller than the vertical advection and horizontal mixing terms. This assumption relies on the combination in BRAM2 of dynamical constraints from ERA-Interim with chemical constraints from MLS (Errera et al., 2019)

Furthermore the caption of Figure 6 in the revised manuscript states:

BRAM2 is depicted with a black line and symbols, as usually done for observations, because it is constrained by both dynamical and chemical observations.

65. *Page 9, line 282, replace "We first investigate" by "First, we investigate..."*

Done and moved to Sect. 4.1 page 13 line 385.

First, we investigate the N_2O mixing ratio...

66. *Pages 9-10, line 283-285, Is there any possible physical explanation of ERAi underestimation in tropics? Is there any link to the upwelling or extent of the tropical pipe? Or just a different location of the maximum for ERAi compare to JRA-WACMM?*

The physical reason behind the underestimation of N_2O in ERAI compared to JRA55 is the faster upwelling in JRA55 (evaluated by Chabrillat et al., 2018 through mean AoA) compared to ERAI (because of the inverse relationship between N_2O and mean Age of Air). Unfortunately, we did not have mean AoA output from WACCM to draw similar conclusions for the CCM. This is discussed in Sect. 4.3 of the revised manuscript:

In the tropical regions, the N_2O mixing ratios in WACCM-CCMI agrees well with the reanalysis of Aura MLS, while the CTM results show large differences in the N_2O abundances depending on the input reanalysis (Fig. 9(a) and 9(b)). In regions where the AoA is less than 4.5 years and N_2O is greater than 150 ppb, i.e. in the tropical regions and lower stratospheric middle latitudes (Strahan et al., 2011), the N_2O mixing ratio is inversely proportional to the mAoA, because faster upwelling (younger air) implies more N_2O transported from lower levels, decreasing its residence time and resulting in a limited chemical destruction (Hall et al., 1999; Galytska et al., 2019). The dynamical reanalyses also produce large differences in mAoA at 15 hPa: MERRA delivers the oldest mAoA and MERRA2, ERAI and JRA55 progressively show younger mAoA (Fig. 4(b) in Chabrillat et al., 2018). Hence the large discrepancies in N_2O mixing ratio can be explained by the large differences in AoA, while M_y and A_z contribute to rates of change of N_2O .

67. Page 9-10, line 283-287, the discussion is not clear and very hard to follow. Why "the subtropics 40-60" is just not mentioned in the N_2O vmr? All panels in the figure have to be discussed, if not please do not show them. It will be clearer and easier to follow if the discussion is done by latitude band e.g. "In the tropic, ...", "In the mid-latitudes, ..." and "In the polar region, ..."

Indeed, the structure was confusing. As mentioned before, we changed the layout of the manuscript, merging the Sections 3 and 4. In the revised manuscript, the Sect. 4 "Climatological seasonal cycles" is divided in three subsections by latitude bands: Polar regions, middle latitudes and Tropics.

68. Page 10, line 289, replace "We then investigate" by "Second, we investigate..."

The sentence was re-written as follows:

We continue by investigating the contribution from A_z .

69. Page 10, line 322-323, the sentence is not clear and can be split into 2 sentences and formulated clearly.

As a result of the structure of the manuscript, this sentence does not mention anymore the middle latitudes:

In both the tropical regions, A_z is positive all year round showing the effect of tropical upwelling, and agrees very well in the reanalyses (Figs. 9(c) and (d)), as a result of the good agreement in the tropical upwelling velocity at 15 hPa (Fig. S7 bottom row), and also as depicted by mAoA diagnostics (Fig. 4(d) in Chabrillat et al., 2018).

70. Page 10, line 326, add a comma after "Finally". Same after "In the Tropics from November to April (Fig. 6(g))", same after "In the middle latitudes (Fig. 6(h))", same after "In the arctic region (Fig. 6(i))"

With the new manuscript structure, these parts were removed, or moved to the correct places and corrected.

71. In this section 3.2, differences are reported but there is no physically explained attempt.

As mentioned before, we merged the Sections 3 and 4 to address this problem. We reduced the purely descriptive parts, and we moved (and enhanced where possible) the relevant scientific discussion to where it belongs for each figure.

72. *Page 11, line replace "After reporting on the climatological annual cycles, it is desirable to estimate their inter-annual variability. To this end," by " To analyse the inter-annual variability of the annual cycle, we..."*

Done.

73. *Redo panel f) and i) of figure 6 in order to get the quantities shown properly. It is not necessary to keep the same y-axis scaling identical for "Az" and "My" terms.*

Done.

74. *Page 11, line 341, replace "We first consider" by "First, we consider"*

Done.

75. *Page 11, line 342-343, in the [0 , 20] at 15hPa, BRAMS N₂O mixing ratio is more closer to the reanalyses at the first half of the year.*

The sentence was rephrased for the [0 , 20] latitudinal band:

WACCM-CCMI and the BASCOE reanalysis of Aura MLS show very similar variabilities, especially in the southern tropics.

76. *Page 11, line 344, add a comma after "In the northern mid-latitudes (Fig. 7(d))"*

Done. The sentence was modified as follows:

In the northern mid-latitudes, the interannual variability of the N₂O mixing ratio increases in late winter across all the datasets, as a response to the increased wintertime variability of the surf zone (Fig. 11(b)).

77. *Redo panel a) and b) of figure 8.*

Done.

78. *Page 11, line 345-346, why there is no attempt of physical explanation or to link of the spread to differences in upwelling or tropical pipe in the dataset?*

In the revised manuscript, the Sect. 3.3 was merged with the relevant parts of Sect. 4 and the scientific discussion was improved, while some purely descriptive parts were removed.

79. *Page 11, line 347, add a comma after "In the middle latitudes (Figs. 7(e) and 7(h))"*

In the revised manuscript this part was expanded as follows:

The inter-annual variabilities of A_z and M_y in the southern mid-latitudes are shown in Figs. 11(c) and 11(e) respectively. As their mean value, A_z and M_y are most variable during austral spring and late summer in the reanalyses, while WACCM simulates an earlier peak during winter in the inter-annual variabilities of A_z and M_y compared to the reanalyses. In the northern mid-latitudes, the inter-annual variabilities of A_z and M_y peak in winter, as their mean values, and WACCM simulates smaller variabilities compared to the reanalyses (Fig. 11(d) and 11(f)).

80. Page 11, line 348, add a comma after "In the antarctic region (Fig. 8(c))"

This part was removed from the revised manuscript.

81. Page 11, line 348-350, what is the physical explanation of the hemispheric differences in the A_z and M_y ? The strength of the polar? Sudden stratospheric warming?

The differences between the Arctic and Antarctic are discussed in Sect. 5 of the revised manuscript:

We now look at the interannual variability of A_z and M_y in the polar regions. Above the Antarctic, the inter-annual variability of A_z and M_y is maximum during spring (Figs. 10(c) and (e)), due to the large inter-annual variability in vortex breakup dates (Strahan et al., 2015). While the maximum variability of M_y is consistently reached in October in all the reanalyses, WACCM-CCMI simulates an earlier maximum (September) that does not correspond with the maximum in its mean values. The lower wintertime variability of both A_z and M_y would increase if a longer period was considered to include the exceptional Antarctic vortices of 2002 (Newman and Nash, 2005) and 2019 (Yamazaki et al., 2019). Above the Arctic, M_y and A_z are most variable during winter, reflecting the frequent disruptions of the northern polar vortex by sudden stratospheric warmings (SSWs, Butler et al., 2017). A case study of the effect of a SSW on the N_2O TEM budget showed that A_z and M_y contribute more to this budget during the SSW event than in the seasonal mean (Randel et al., 1994). Thus, the large wintertime variability of A_z and M_y is explained by the occurrence of seven major SSWs detected in the reanalyses for the 2005-2014 period (Butler et al., 2017).

82. Page 11, line 349, replace "the vortex break-up," by "the breaking vortex period"

Done. The sentence was also rephrased:

The variability of the N_2O mixing ratio increases in October i.e. during the breaking vortex period that is highly variable in time (Strahan et al., 2015).

83. Page 12, line 350-351, replace "We now move to the variability of the horizontal mixing term M_y starting from the Tropics (Figs. 7(j) and 7(k)). In the southern tropics (Fig. 7(j))" by "Regarding the variability of the horizontal mixing in the southern tropics (Figs. 7(j, k)), M_y term shows... In the northern tropics (Fig. 7(k)), M_y"

This part was rephrased after the structure change of the manuscript.

The variability of M_y in the tropical regions is small compared to the extratropical regions (Figs. 12(e) and 12(f)), in agreement with calculations of standard deviations of the effective diffusivity within the tropical pipe (Abalos et al., 2016a). The reanalyses deliver a larger inter-annual variability in the northern tropics during boreal winter, while in the southern tropics the variability of M_y presents a much weaker annual cycle. WACCM-CCMI does not reproduce this hemispheric asymmetry, with a rather flat profile in both hemispheres and a clear underestimation in the northern tropics, as shown for its mean values.

84. Page 12, line 355, add a comma after "In the mid-latitudes"

This sentence was removed and the mid-latitudes are better discussed now. See response to minor comment 79.

85. *Page 12, line 338, add a comma after "In the antarctic region (Fig. 8(e))".*

This descriptive sentence was removed, and the polar regions are better discussed now, see response to minor comment 81.

86. *Page 12, line 360 add a comma after "The Arctic (Fig. 8(f)) "*

This descriptive sentence was removed, see response to minor comment 81.

87. *Page 12, line 360 add a comma after "Among the reanalyses"*

This descriptive sentence was removed.

88. *Page 12, line 370, please don't oversell the agreement. Replace "excellent agreement" by "fairly good" and complete the sentence "but some differences also occur at ...". In addition this part of the sentence "while the CTM delivers overall smaller variabilities." is not true as the reanalysis also show spread in the tropics.*

This generic part of the Discussion was removed.

89. *Page 12, line 376, add a comma after " Above the Arctic in the middle stratosphere"*

Done, and "in the middle stratosphere" was removed.

90. *Page 13, line 408, add a comma after "During the SH spring"*

Done and "SH" was replaced by "austral".

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Response to Reviewer#2 for: Climatological impact of the Brewer-Dobson Circulation on the N_2O budget in WACCM, a chemical reanalysis and a CTM driven by four dynamical reanalyses

Minganti et al., ACPD, 2020

We thank the reviewer for his/her useful comments. In our replies below the italic type is used for the reviewer's comments, the plain text for authors' answers and the bold type for the revised text in the manuscript.

Replies to general comments.

However the manuscript should be highly improved in structure and wording! I have the feeling that in some sections the text lacks an organized structure. E.g. when describing the figures, the text jumps from one figure panel to another and it is really hard to follow. I recommend publication after carefully reading over the text again and rephrasing where it is necessary

As recommended by both reviewers, the structure of the manuscript was changed: the Sect. 3 was merged with Sect. 4, and the manuscript was restructured as follows:

Section 1. Introduction

Section 2. Data and Method

Section 3. Latitude pressure cross sections

Section 4. Climatological seasonal cycles

Subsection 4.1 Polar regions

Subsection 4.2 Middle latitudes

Subsection 4.3 Tropics

Section 5. Interannual variability of the seasonal cycles

Section 6. Summary and Conclusions

This new structure allowed to remove some purely descriptive parts in Sect. 3, and to better follow the text by latitude band when discussing the figures (especially for Fig. 5 and 6). The change in the manuscript structure led to a change of the layout of the figures as well. We separated them by latitude bands of each hemisphere (one figure for the polar regions, one figure for the surf zones and one for the tropics) in order to better follow the flow of the Section 4 and its subsections and Section 5 of the revised manuscript.

The Introduction was revised as well. Every major concept now gets his own paragraph(s), and some of them were improved, e.g. reanalyses and CTMs, while the paragraph about long-term trends of the BDC was de-emphasized, because this manuscript investigates only climatologies and inter-annual variabilities but not long-term changes.

All these structure changes, together with the reduction of the descriptive parts, intend to improve the wording/phrasing of the manuscript.

Specific comments/questions.

1. -page 1, line 2: reword: " ... from well-mixed tropical troposphere to polar stratosphere....": This is a bit too short, here one has the impression, that tracers are transported directly from trop. troposphere to the polar region.

The sentence was reworded as follows:

The Brewer-Dobson Circulation (BDC) is a stratospheric circulation characterized by upwelling of tropospheric air in the Tropics, poleward flow in the stratosphere, and downwelling at mid and high latitudes, with important implications for chemical tracers distribution, stratospheric heat and momentum budgets and mass exchange with the troposphere.

2. -page 1, line 7: insert "in" – > in a chemical reanalysis

Done.

3. -page 1, line 10: have not been compared before.

Done.

4. - page 1, line 14: Please clarify, I do not understand the sentence: "...reflecting the large diversity in mean AoA obtained with the same experiments." The present study does not look at AoA with CTM experiments.

Here we referred to the study from Chabrilat et al., (2018). They used the same configuration of the BASCOE CTM as for the current manuscript to do Age of Air calculations. Anyway, the sentence was not clear and it is rephrased:

....reflecting the large diversity in the mean Age of Air obtained with the same CTM experiments in a previous study.

5. - page 2, line 27: include that you compare interannual variability between the different datasets.

Done, the sentence was modified as follows:

We also compare the inter-annual variability in the horizontal mixing and the vertical advection terms between the different datasets.

6. - page 2, line 33: reword and clarify this sentence to e.g. "The Brewer Dobson Circulation is characterized by upwelling of tropospheric air to the stratosphere in the tropics, followed by". Note however that the BDC includes both residual circulation (net mass transport) and two-way mixing. Moreover the downwelling takes not only place in the high, but also in the mid-latitudes (change to – > extratropical downwelling) and not only in wintertime, although in the respective winter hemisphere it is much stronger.

The sentence was re-written as follows:

The Brewer-Dobson Circulation (BDC, Dobson et al., 1929; Brewer, 1949; Dobson, 1956) in the stratosphere is characterized by upwelling of tropospheric air to the stratosphere in the Tropics, followed by poleward transport in the stratosphere and extratropical downwelling. For tracer-transport purposes the BDC is often divided into an advective component, the residual mean meridional circulation (hereafter residual circulation), and a quasi-horizontal two-way mixing which causes net transport of tracers, not of mass (Butchart, 2014).

7. - page 2, line 46: Why should mixing be limited to a specific latitudinal region of the winter stratosphere? In the surf zone mixing is only stronger. (see e.g. Fig. 1 in Bnisch et al. 2011)

The sentence was modified and the reference was added:

The two-way mixing is stronger in a specific latitudinal region of the winter stratosphere, the "surf zone" (McIntyre and Palmer, 1983), and in the subtropical lower stratosphere all year round (e.g. Fig.1 of Bnisch et al., 2011).

8. - page 2, line 51: change to: "... due to the increase in well mixed greenhouse gases (e.g. Butchart et al 2014,...) and due to increased ozone depleting substances (e.g. Polvani et al. 2018 ...) "

Done.

....due to the increase in well-mixed greenhouse gases (Butchart et al., 2010; Hardiman et al., 2014; Palmeiro et al., 2014) and ozone-depleting substances (Polvani et al., 2018),....

9. -page 3, line 56 and line 63: Here the study of Fritch et al. 2019 (<https://www.atmos-chem-phys-discuss.net/acp-2019-974>) is interesting.

The mentioned study is now included in the manuscript:

The difficulty to derive observational trends in the BDC can be partly attributed to the spatial and temporal sparseness of the observations, together with its large dynamical variability and the uncertainty of trends derived from non-linearly increasing tracers (Garcia et al.; 2011, Hardiman et al., 2017; Fritsch et al., 2020).

10. - page 3, line 60: ... observational trends in the ...

Done.

11. - page 3, line 65: Say why is it important to do this separation?

This sentence and the previous one ("Furthermore the observational datasets cannot discriminate....") were removed from the revised manuscript as this paragraph was de-emphasized.

12. - page 3, line 72: Could you write more about the study of Tweedy et al. 2017, as they are also looking at the N₂O TEM continuity equation in GEOSCCM!

A sentence about Tweedy et al., 2017 was added:

In the tropical lower stratosphere, the distinction between vertical and horizontal transport is important, as they impact differently the seasonality of N₂O in the northern and southern Tropics (Tweedy et al., 2017).

13. - page 3, line 75: In Abalos et al. 2013 the stratospheric N₂O buget isn't shown.

The reference to Abalos et al. (2013) was removed.

14. - page 3, line 85: change to: ...four different dynamical reanalyses are used here to drive simulations

The paragraphs about the reanalyses and the CTM were changed. Now the mentioned part states:

In order to contribute further to the S-RIP BDC activity, four different dynamical reanalyses are used here to drive the BASCOE CTM simulations,

compute the N_2O TEM budget and compare its components with the results derived from WACCM. Namely we consider:.....

15. - page 3, line 88: Please clarify: Is only WACCM4 compared to BRAM2?

Both WACCM and the CTM experiments are compared to BRAM2, this is now explicitly stated:

WACCM and the CTM experiments are also compared....

16. - page 4, line 93: Are there studies with CTMs driven by reanalyses that studied tracer transport in TEM framework?

To our knowledge, a few studies were performed using CTM in the TEM framework, but they used dynamical fields obtained from CCMs and not from reanalyses (e.g. Strahan et al., 1996). Hence they were not deemed relevant to this work and we did not include them in the manuscript.

17. - page 4, line 107-118: You explain the differences of WACCM-4 and WACCM-CCMI by model development. But are there also differences in the setup of the simulations (e.g. different SSTs,)

The model setup of WACCM4 was as similar as possible to the CTM experiments, to allow fair comparison. This is now stated in the manuscript:

We ran one realization of the public version of WACCM (hereafter WACCM4, Marsh et al., 2013), with a similar setup (e.g. lower boundary conditions) as the CTM experiments;....

18. - page 4, section 2: I recommend to include a table to give an overview over the different simulations (CCM, CTM with diff. reanalysis).

The table is now included (Table 1).

Dataset name	Reference	Dynamical Reanalysis	Chemical reanalysis of	Model grid	Top level
WACCM4	Marsh et al., (2013)	none	none	2.5°x1.9°, L66	5.1x10 ⁻⁶ hPa
WACCM-CCMI	Garcia et al., (2017)	none	none	2.5°x1.9°, L66	5.1x10 ⁻⁶ hPa
ERA1	Chabrillat et al., (2018)	ERA-Interim (Dee et al., 2011)	none	2.5°x2°, L60	0.1 hPa
JRA55	Chabrillat et al., (2018)	JRA-55 (Kobayashi et al., 2015)	none	2.5°x2°, L60	0.1 hPa
MERRA	Chabrillat et al., (2018)	MERRA (Rienecker et al., 2011)	none	2.5°x2°, L72	0.01 hPa
MERRA2	Chabrillat et al., (2018)	MERRA2 (Gelaro et al. 2017)	none	2.5°x2°, L72	0.01 hPa
BRAM2	Errera et al., (2019)	ERA-Interim (Dee et al., 2011)	MLS (Livesey et al., 2015)	3.75°x2.5°, L37	0.1 hPa

Table 1: Overview of the datasets used in this study.

19. - page 5, line 132: WACM – > WACCM

Done.

20. - page 5, line 137: ... as input...

The sentence was changed:

Chabrillat et al. (2018) explain in detail the preprocessing procedure that allows the BASCOE CTM to be driven by arbitrary reanalysis datasets, and the set-up of model transport.

21. - page 6, line 161: What do you mean with situation of interest?

”Situation of interest” was indeed misleading, a more appropriate wording would be ”regions of interest”. BRAM2 has been evaluated in several regions of interest in the

middle atmosphere as defined in the BRAM2 paper (Errera et al., 2019): the middle stratosphere (MS) the tropical tropopause layer (TTL), the lower stratospheric polar vortex (LSPV) and the upper stratosphere polar vortex (USPV). The chemical species were only evaluated in some relevant regions, and BRAM2 N_2O was evaluated in MS, LSPV and USPV. The text was rewritten more clearly:

BRAM2 N_2O has been validated between 3 and 68 hPa against several instruments with a general agreement between 15 % depending on the instrument and the atmospheric region (the middle stratosphere or the polar vortex, see Errera et al., 2019).

22. - page 6, line 182: " N_2O balance" – > *In this section you use tracer X to explain the TEM diagnostics, but here you change back to N_2O . Perhaps you use N_2O instead of X in the entire section?*

We now use χ in all the formulas, and " N_2O balance" was changed to "tracer balance". Furthermore, we stated explicitly that χ represents the N_2O concentrations in the revised manuscript:

...where χ is the volume mixing ratio of N_2O ,...

23. - page 7, line 200: *Can you be a bit clearer, please: You are giving the causes of the non-zero residual for WACCM, but what about the residuals in the CTM, and the chemical reanalysis? Is it only the timestep in BASCOE?*

Regarding the CTM experiments and BRAM2, the reason for the large residual could be the coarser resolution compared to their input reanalyses (especially for BRAM2), impacting the numerical errors in the the horizontal and vertical derivatives that are involved in the TEM analysis. For this reason, a new reanalysis of Aura MLS is planned (BRAM3) with the same horizontal and vertical resolution as in the CTM. The unresolved mixing can also play a large role, as discussed in Sect. 3 of the revised manuscript. Taking into account these two factors, the text was rewritten:

The BASCOE datasets have a coarser horizontal resolution than their input reanalyses (especially BRAM2; see Table 1). This affects the accuracy of the vertical and horizontal derivatives, with possible implications for the residual. The possible causes of the residual in the five reanalyses are discussed in more detail in Sect. 3

24. -page 7, line 205: "...while ..." – > "...even though ..."

Done.

25. - page 7, line 209: *Note that Tweedy et al. 2017 looked at N_2O TEM buget at 85 hPa in the tropics.*

In the revised manuscript, it is stated more clearly that they looked in the tropical lower stratosphere:

In order to validate our N_2O TEM budget, we reproduced the findings reported in Tweedy et al. (2017, Fig. 7) with WACCM-CCMI in the tropical lower stratosphere, and we noticed similar results (not shown).

26. - page 7, line 213: *Why does w^* vary in reanalyes data? Perhaps you can add one sentence more about Abalos et al. 2015.*

The sentence was slightly modified to include the main physical reason of the disagreement:

The upwelling velocity \bar{w}^* can vary considerably in the dynamical reanalyses, , as it is a small residual quantity (Abalos et al., 2015).

27. -page 8, line 219: delete "the" – > are strongest ...

The sentence was removed from the revised manuscript.

28. - page 8, line 220: *You motivate the choice of the 15 hPa level with large differences between the CCM and CTM simulations in this region. Where do you see this? I suppose in Figs. 3 +4. And why isn't it interesting to see what is going on in the lower stratosphere?*

Indeed those differences can be seen from Figures 3 and 4. We didn't look at the lower stratosphere because the vertical range of validity for BRAM2 is limited to 3-68 hPa (Errera et al., 2019).

29. - page 8, line 16: *The terms, "vertical advection", "horizontal mixing" and their abbreviations A_y and M_y are mixed within the manuscript, even between one sentence these terms are mixed (e.g. page 8, line 225). Can you please use the terms consistently?*

In the description of Figs. 1 and 2, we kept using the full names and their abbreviations (e.g. the vertical advection term A_z) as we explain the methodology in that section. In the rest of the manuscript we use the abbreviations A_z and M_y .

30. -page 8, line 226: *"higher latitudes" – > I can see this mainly in the northern higher latitudes.*

The description of Figs. 1 and 2 was largely reduced in order to remove purely descriptive sentences such as this one (lines 226-229).

31. page 8, line 232 (and also line 229): *"... especially in the reanalyses A_z and the residual play a minor role": I wouldn't say, that this effect is "minor"!*

The whole paragraph was re-written, see comment above. Figures 1 and 2 are now described and discussed as follows:

Figs. 1 and 2 show the N_2O TEM budget terms at 15 hPa for all the datasets for the boreal winter (December-January-February, DJF mean) and summer (June-July-August, JJA mean) respectively. The 15 hPa level (around 30 km altitude) was chosen because large differences can be found between WACCM-CCMI, BRAM2, and the CTM runs at this level, and because the dynamical reanalyses are not constrained as well by meteorological observations at higher levels (Manney et al., 2003). Figs. 1 and 2 aim to show how the dynamical and chemical terms of the budget balance each other to recover the tendency $\bar{\chi}_t$ at different latitudes. The discussion about the differences between the datasets, and their possible physical causes, are addressed in the next Sections.

The vertical advection term A_z shows how the upwelling contributes to increasing the N_2O abundances in the tropics and summertime mid-latitudes, and how polar downwelling contributes to decreasing the N_2O abundances in the winter hemisphere. The horizontal transport out of the tropics due

to eddies, as represented by M_y , reduces the N_2O abundance in the tropical latitudes of the wintertime hemisphere, and increases the N_2O mixing ratio at high latitudes in the winter hemisphere. The other terms of the TEM budget are weaker than A_z and M_y : the meridional advection term A_y tends to increase the N_2O abundance in the winter subtropics and extratropics, while the vertical transport term due to eddy mixing, M_z decreases it over northern polar latitudes and the chemistry term $P - L$ shows that N_2O destruction by photodissociation and $O(1D)$ oxidation contributes to the budget in the tropics and also in the summertime hemisphere. All budget terms are weaker in the summer hemisphere than the winter hemisphere. Over the southern polar winter latitudes, the reanalyses deliver negative M_y that are balanced by large positive residuals, which implies a less robust TEM balance (Fig. 2). This is not the case with WACCM, where M_y tends to increase the N_2O abundance in the polar vortex. Such differences between the datasets are highlighted and discussed in the next sections.

32. - page 8 line 238: spelling: reanalyses

Done.

33. - page 9, line 253: You only show thee reanalyses here, not four.

"...in the four reanalyses" was replaced by "...in the other reanalyses".

34. -page 9, line 266: middle stratospheric – > middle stratosphere

Done.

35. -page 9, line 257: "(Fig. 3(f), (i), (l))" – > right columns of Fig. 3

Done.

36. -page 9, line 269: Motivate why you are choosing a single level in the middle stratosphere (15 hPa). What about the lower stratosphere?

We tried several levels in the middle stratosphere and found that the differences between the datasets were most visible at 15 hPa while other levels did not bring added value to the intercomparison. With respect to the lower stratosphere, see reply 28 above.

37. -page 9-11, description of the climatological seasonal cycles: In my opinion this section is very hard to read, as the SH and NH are separated into two pictures. I recommend to merge Fig.5 and 6 to one Figure and then describe first the tropical, mid-latitude and polar N_2O (upper row), second the vertical advection A_z (middle row) and third horizontal mixing M_y (bottom row). Thus it is easier to see the differences in NH and SH, the text is better structured and you do not have to repeat patterns that are similar.

In order to follow this comment and another major comment by the first reviewer, Figs. 5 and 6 were re-organized into three figures, each of them covering both hemispheres. The revised Figs. 6, 8 and 9 show respectively the polar regions, mid-latitudes and tropics and are discussed in sections 4.1, 4.2 and 4.3 respectively. This new structure avoids any repetition while showing simultaneously, for each latitude band, the N_2O cycle and the two main terms contributing to its TEM budget. Fig. 9 was also split into latitude regions and inserted as revised Figs. 5 and 7, to contribute to the interpretation of our results in the polar regions and mid-latitudes. The tropical regions of Fig. 9 were moved to the Supplement.

38. - page 9, line 278-281: *What do you mean with uncertainty - the 1 sigma standard deviation?*

Yes indeed, as stated in Errera et al., (2019). This sentence was moved to the caption of Fig. 6 following a comment from Reviewer 1.

39. - page 9, line 282: *" We first investigate the N_2O mixing ratio in the SH. In the tropic (Fig 5c and 6a)...." – > Fig. 6a is not in the SH!*

After the rearrangement of the sections explained above, this sentence is not limited to the SH any more:

In the tropical regions, the N_2O mixing ratio in WACCM-CCMI agrees well with the reanalysis of Aura MLS, while the CTM results show large differences in the N_2O abundances depending on the input reanalysis (Figs. 9(a) and 9(b)).

40. -page 9, line 283: *Please point out here more clearly, that BRAM2 is used as reference, and that this is the case for the entire section.*

This is pointed out more clearly after the structure rearrangement:

In the following, we will consider BRAM2 as the reference when comparing N_2O mixing ratios between datasets, because its dynamics and chemistry are both constrained to observational datasets.

41. -page 10, line 286: *change to: ...is smaller than in BRAMS in all simulations.*

Done.

42. -page 10, line 284-288: *You missed to describe the mid-latitudes....*

With the new manuscript structure, the middle latitudes are now discussed in the dedicated Sect. 4.2.

43. -page 10, line 287: *You wanted to talk about N_2O , not about A_z and M_y ...*

That paragraph was confusing indeed. Now the discussion of the middle latitudes is put together in Sect 4.2. It starts with the N_2O mixing ratio in both hemispheres, and continues with A_z and M_y for each hemisphere.

44. -page 10, line 300: *"...expect for JRA55" – > expect JRA55*

Done.

45. -page 10, line 305: *"It is yet comparable..." – > What? The uncertainty.*

The sentence was removed from the revised manuscript, as it did not add any relevant scientific point.

46. -page 10, line 311: *Replace differ to different.*

Done.

47. -page 11, line 337: *Do you use the 1-sigma standard deviation?*

Yes indeed. The text could be more precise, as implicitly suggested by the reviewer. The revised sentence now states:

... we compute for each month the 1-sigma standard deviations of the N_2O mixing ratio, M_y and A_z across the ten simulated years.

48. -page 11, line 335-340: *I think it is easier for the reader if you plot the standard deviation the same way as in Fig. 5+6. I do not see a real advantage of plotting the results in this order. And as recommended before it would be nice to have Fig. 7+8 in one plot and restructure the text accordingly.*

Indeed, we now plot the standard deviations as the previous figures. We separated former Fig. 7 by latitude bands, and, in the revised manuscript, Fig. 10 (former Fig. 8) shows the polar regions, Fig. 11 the middle latitudes, and Fig. 12 the Tropics. The text was restructured accordingly, and according to the new sections layout.

49. -page 11, line 343: *Why does the variability in WACCM-CCMI strongly depends on the considered realization? Shouldn't the internal variability between these ensemble simulations be similar?*

This was a surprising result, as in the other latitude bands the internal variability of WACCM does not play a major role. Strong differences between ensemble members with respect to inter-annual variability indicate that the considered period is not long enough to explore the inter-annual variability in the northern mid-latitudes, and that the mean variability from this ensemble (with only 3 members) would not be representative of the internal variability of WACCM. Fortunately, our study did not investigate the ensemble mean but showed instead the full range from the 3 WACCM realizations. This will be stated in the revised manuscript.

50. -page 12, subsection "polar regions": *The structure of this subsection was not clear to me during reading: you first write about the wintertime North Pole, then about the wintertime South, then you jump to the SH spring and to Antarctic and Arctic inter-annual variability. Perhaps you can give an introducing sentence of what you will discuss in this section.*

With the new structure of the manuscript mentioned above, this subsection was merged with Sect. 3, and all the information (wintertime North Pole, wintertime South,...) were moved to the right places when describing the figures.

51. -page 12, line 375: *What do you mean with "Above the Arctic in the middle stratosphere ... (Fig.6)"? Do you refer to the 15 hPa level in Fig. 6?*

Yes. This paragraph was moved to Sect. 4.1 of the revised manuscript.

52. -page 12, line 376: *I cannot see that N_2O abundance in polar regions (Fig. 6c) are in good agreement in WACCM and BRAMS in the wintertime ...*

The reviewer is right, and sentence was modified:

Above the Arctic, the N_2O abundances simulated by WACCM agree with the BRAM2 reanalysis, except in December and January, and....

53. -page 12, line 379: *Compared to which reanalysis? To all? Before you were comparing with BRAMS.*

Yes, we consider here all the reanalyses. The text was modified accordingly:

Compared to the dynamical reanalyses and BRAM2,....

54. -page 12, line 381: *Replace "Fig. 6 bottom row", to Fig. 6 g+h. And why are you talking about tropics and mid-latitudes here? In this chapter you wanted to discuss the polar regions.*

The references to Tropics and mid-latitudes were removed as a consequence of the manuscript structure. The sentence was re-written:

Compared to the dynamical reanalyses and BRAM2, WACCM shows in the Arctic a 2-fold underestimation of the N_2O changes due to horizontal mixing during winter.

55. -page 13, line 383: *Do you mean the aging by mixing term in the polar regions of Fig. 2 in Dietmüller et al. 2018? Moreover reword "Note that ..." This is a poor transition between the two sentences.*

Yes, we mean aging by mixing. The sentence was modified for clarity:

It should also be emphasized that WACCM is among the CCM models with the lowest contribution of aging by mixing to Age of Air (Fig. 2 in Dietmüller et al., 2018).

56. -page 13, line 386: *Include that TEM AoA budget was done in CCM simulations.*

Done.

Dietmüller et al. (2017) applied the TEM continuity equation to the Age of Air (AoA) in CCM simulations.

57. -page 13, line 391: *Can you explain, why the TEM formulation is different in this study?*

Our phrasing was misleading. The differences arise only from the different nature of AoA and N_2O : AoA does not have chemical sources nor sinks in the stratosphere, while N_2O is destroyed in the tropical higher stratosphere. Since the definition of the dynamical TEM terms does not change, we removed "with a different TEM formulation", and the sentence now reads:

Even though we use a real tracer (N_2O), we find a qualitative agreement with this analysis based on AoA: our residual term is larger in regions characterized by strong gradients such as the antarctic vortex edge, and larger with dynamics constrained to a reanalysis than with a free-running CCM (see EMAC results in Fig. 1d by Dietmüller et al., 2017).

58. -page 13, line 392: *"... agreement: our residual term is larger ..." But you are listing the differences here.*

The second difference ("with a different TEM formulation") was removed. The point of this paragraph is that we find qualitative agreement between their "aging by diffusion" and our residual term, since both are computed as the remaining of the respective TEM budgets. We hope that the revised sentence makes this clearer (see previous comment).

59. -page 13, line 396: *Perhaps change to "...SH winter". (Also in other parts of the paper)*

The sentence was re-written:

In the austral winter, over the Antarctic Polar cap and below 30 hPa, M_y agrees remarkably well in all datasets (Fig. 4).

60. -page 13, line 397: *Again: What do you mean with "above 30 hPa"? Do you mean the 15 hPa level (latitude band 60-80S), as you are referring to Fig. 5?*

We referred to Fig. 4 of the ACPD manuscript. This sentence was moved and adapted to Sect. 3 in the revised manuscript, where it still refers to Fig. 4; this is now clearer because Fig. 5 is introduced only in the next section.

61. -page 13, line 399: *You are talking about Fig. 4, not about Fig 5!*

Same reply as for the previous comment.

62. -page 13, line 401: *Are these studies are giving an explanation for the mixing inside the vortex. If yes, can you please give the explanation here.*

De la Camara et al., 2013 states that the Rossby waves breaking can contribute to the tracer mixing inside the polar vortex and occasionally across its edge. The sentence was re-written as:

The impact of horizontal mixing on N_2O inside the wintertime polar vortex is not negligible (e.g. de la Camara et al., 2013; Abalos et al., 2016a), as Rossby waves breaking occurs there as well as in the surf zone.

63. -page 13, line 403: *Make clear, that it is overestimated in WACCM ... (and overestimated according to what?)*

Garcia et al. (2017) compared the winds simulated by WACCM to the winds from MERRA. This is stated more precisely in the revised manuscript:

This disagreement can be related to differences in the zonal wind: it is overestimated in WACCM above 30 km in subpolar latitudes compared to MERRA (Garcia et al., 2017) and the polar jet is not tilted equatorward as in the reanalyses (see black thin lines in Fig. 4, and Fig. 3 of Roscoe et al., 2012).

64. -page 13, line 404: *Change to : ... (see black thin lines in Fig. 4).*

Done.

65. -page 13, line 405: *You do not show the residual terms in Fig. 5.*

The sentence refers to Fig. 4, as the residual terms were not shown in Fig. 5. The sentence was moved to Sect. 3 of the revised manuscript, and changed as follows:

Yet, the differences in M_y and A_z above the Antarctic in winter should be put into perspective with the large residual term that points to an incomplete TEM budget (Fig. 4 right column).

66. -page 13, line 408: *Say, why you are now looking at SH spring.*

Indeed, that change to SH spring was confusing as the new paragraph was not properly introduced. After the change in the structure of the manuscript, this part was moved to Sect. 4.1 of the revised manuscript, and now it follows the discussion of the wintertime M_y at 15 hPa over the antarctic.

67. -page 13, line 409: *... better agreement ...” Better compared to what?*

Here we mean compared to austral winter. After the change in the manuscript structure, this sentence was moved to Sect. 4.1 of the revised manuscript for the description of Fig. 6 and changed as follows:

During the austral spring, the vortex breakup leads to an increased wave activity reaching the Antarctic (Randel and Newman, 1998), and mid-stratospheric M_y is in better agreement among all datasets compared to austral winter.

68. -page 14, line 418: Replace "reanalyses" with dynamical reanalyses. And why is BRAM2 not included in this comparison?

The word "reanalyses" was replaced by "dynamical reanalyses". BRAM2 is not included in this comparison because it is dynamically constrained to the winds from the ERA-Interim reanalysis, and its results are nearly identical with those of the CTM simulation driven by ERA-Interim, i.e. these differences are only due to the coarser resolution of BRAM2 and they are negligible.

69. -page 14, line 434: Please explain critical lines.

This is explained in the revised manuscript as follows:

It is due to transient Rossby waves that cannot travel further up into the stratosphere due to the presence of critical lines, i.e. where the phase velocity of the wave matches the background wind velocity, generally leading to wave breaking (Abalos et al., 2016b).

70. -page 14, line 448: $vmr > mixing\ ratio$

Done.

Replies to comments to the Figures:

71. - Fig. 1+2: Can you please replace "time der" to dN_2O/dt in the legend.

Done.

72. - You are showing different colorbars in Fig. 3 and 4!

We now use the same color scale [-2,2] ppbv/day for both figures.

73. -Fig 5+6, y-axis: Replace X with N_2O .

Done.

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Climatological impact of the Brewer-Dobson Circulation on the N₂O budget in WACCM, a chemical reanalysis and a CTM driven by four dynamical reanalyses

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Abstract.

The Brewer-Dobson Circulation (BDC) ~~transports chemical tracers from the well-mixed tropical troposphere to the polar stratosphere, with many~~ is a stratospheric circulation characterized by upwelling of tropospheric air in the Tropics, poleward flow in the stratosphere, and downwelling at mid and high latitudes, with important implications for ~~climate, chemistry, ozone~~ distribution and recovery chemical tracers distribution, stratospheric heat and momentum budgets and mass exchange with the troposphere. Since the photochemical losses of nitrous oxide (N₂O) are well-known, model differences in its rate of change are due to transport processes that can be separated in the mean residual advection and the isentropic mixing terms in the Transformed Eulerian Mean (TEM) framework. Here the climatological impact of the stratospheric BDC on the long-lived tracer N₂O is evaluated through a comparison of its TEM budget in the Whole Atmosphere Community Climate Model (WACCM), in a chemical reanalysis of the Aura Microwave Limb Sounder version 2 (BRAM2) and in a Chemistry-Transport Model (CTM) driven by four modern reanalyses (ERA-Interim, JRA-55, MERRA and MERRA2). The effects of stratospheric transport on the N₂O rate of change, as depicted in this study, have not been compared before across this variety of datasets and never investigated in a modern chemical reanalysis. We focus on the seasonal means and climatological annual cycles of the two main contributions to the N₂O TEM budget: the vertical residual advection and the horizontal mixing terms.

The N₂O mixing ratio in the CTM experiments has a spread of approximately ~ 20% in the middle stratosphere, reflecting the large diversity in the mean Age of Air obtained with the same ~~experiments~~ CTM experiments in a previous study. In all datasets the TEM budget is well-closed and the agreement between the vertical advection terms is qualitatively very good in the Northern Hemisphere, and good in the Southern Hemisphere except above the Antarctic region. The datasets do not agree as well with respect to the horizontal mixing term, especially in the Northern Hemisphere where horizontal mixing has a smaller contribution in WACCM than in the reanalyses. WACCM is investigated through three model realizations and a sensitivity test ~~where gravity waves are forced differently in the Southern Hemisphere~~ using the previous version of gravity waves parameterization. The internal variability of the horizontal mixing in WACCM is large in the polar regions, and comparable to

the differences between the dynamical reanalyses. The sensitivity test has a relatively small impact on the horizontal mixing term, but significantly changes the vertical advection term and produces a less realistic N₂O annual cycle above the Antarctic.

25 In this region, all reanalyses show a large wintertime N₂O decrease, which is mainly due to horizontal mixing. This is not seen with WACCM, where the horizontal mixing term barely contributes to the TEM budget. While we must use caution in the interpretation of the differences in this region, where the reanalyses show large residuals of the TEM budget, they could be due to the fact that the polar jet is stronger and not tilted equatorward in WACCM compared with the reanalyses.

We also compare the inter-annual variability in the horizontal mixing and the vertical advection terms between the different datasets. As expected, the horizontal mixing term presents a large variability during austral fall and boreal winter in the polar regions. In the Tropiestropics, the inter-annual variability of the vertical advection term is much smaller in WACCM and JRA-55 than in the other experiments. The large residual in the reanalyses and the disagreement between WACCM and the reanalyses in the Antarctic region highlight the need for further investigations on the modeling of transport in this region of the stratosphere.

35 1 Introduction

The ~~stratospheric circulation is mainly characterized by~~ Brewer-Dobson Circulation (BDC, Dobson et al., 1929; Brewer, 1949; Dobson, 19
the stratosphere is characterized by upwelling of tropospheric air to the stratosphere in the ~~Brewer-Dobson Circulation (BDC, Dobson et al.,~~
~~consisting in a slow upwelling in the Tropics from the troposphere into the stratosphere~~ Tropics, followed by poleward trans-
port ~~and downwelling at higher latitudes in wintertime~~ in the stratosphere and extratropical downwelling. For tracer-transport
40 purposes the BDC is often divided into an advective component, the residual mean meridional circulation (hereafter residual circulation), and a quasi-horizontal two-way mixing which causes net transport of tracers, not of mass (Butchart, 2014).

The BDC is ~~generated by Rossby waves propagating into the winter~~ driven by tropospheric waves breaking into the
stratosphere (Charney and Drazin, 1961), which transfer angular momentum and force the stratosphere away from its ra-
diative equilibrium. This ~~departure from radiative equilibrium~~ is balanced by a ~~meridional (poleward)~~ poleward displace-
45 ment of air masses, which implies tropical upwelling and extra-tropical downwelling (Holton, 2004). The residual cir-
culation can be further separated in ~~a shallow branch, i. e.~~ three branches: the transition, the shallow and the deep branch
(Lin and Fu, 2013). The transition branch encompasses the upper part of the transition layer between the troposphere and
the stratosphere (the tropical tropopause layer, Fueglistaler et al., 2009). The shallow branch is an all year-round lower strato-
spheric two-cell system ~~generated by~~ driven by breaking of synoptic-scale waves, and ~~a winter-time deep branch in the higher~~
50 ~~part~~ the deep branch is driven by Rossby and gravity waves breaking in the middle and high parts of the stratosphere ~~generated~~
~~by Rossby waves (Plumb, 2002)~~ during winter (Plumb, 2002; Birner and Bönisch, 2011). The contributions of different wave
types to the driving of the BDC branches has been quantified using the downward control principle, which states that the
poleward mass flux across an isentropic surface is controlled by the Rossby or gravity waves breaking above that level
(Haynes et al., 1991; Rosenlof and Holton, 1993), and using eddy heat flux calculations as an estimate of the wave activity
55 from the troposphere (e.g., Newman and Nash, 2000).

The quasi-horizontal two-way mixing is generated by two-way transport due to the adiabatic motion of Rossby waves. In the stratosphere this motion is ultimately combined with the molecular diffusion which makes the total process irreversible (Shepherd, 2007). The two-way mixing is ~~limited to~~ stronger in a specific latitudinal region of the winter stratosphere, the 'surf zone' (McIntyre and Palmer, 1983), and in the subtropical lower stratosphere all year round (e.g. Fig. 1 of Bönisch et al., 2011).

60 The mixing process homogenizes the tracer concentration in the surf zone and creates sharp tracer and Potential Vorticity (PV) gradients on its edges (in the ~~tropics-subtropics~~ and at the polar vortex edge), indicating an inhibition of mixing. For this reason the ~~tropics-subtropics~~ and the polar vortex edge are often called transport barriers (Shepherd, 2007).

~~Simulations by Chemistry Climate Model (CCM) predict a global BDC acceleration~~ The BDC plays major roles in controlling the spatial and temporal distributions of chemical tracers such as ozone, water vapor, aerosols, and greenhouse gases, as well in
65 coupling stratospheric processes with the climate system (Riese et al., 2012; Butchart, 2014; Tweedy et al., 2017). The natural variability of the atmosphere largely influences the BDC (Hardiman et al., 2017). All three branches of the BDC are affected by changes in sea surface temperatures and El Niño Southern Oscillation (Yang et al., 2014; Diallo et al., 2019), as well as the phase of the Quasi Biennial Oscillation (QBO, Diallo et al., 2018), and the Arctic oscillation (Salby and Callaghan, 2005).

Modeling studies predict an acceleration of the BDC over the last decades and the twenty-first century due to ~~global changes~~
70 ~~in the abundances of ozone-depleting substances and greenhouse gases (Butchart et al., 2010; Hardiman et al., 2014; Palmeiro et al., 2014; increase in well-mixed greenhouse gases (Butchart et al., 2010; Hardiman et al., 2014; Palmeiro et al., 2014) and ozone-depleting substances (Polvani et al., 2018), but these results cannot be evaluated easily because the BDC cannot be observed directly (Butchart, 2014). Observations of long-lived chemical tracers (e.g.,) are often used to derive estimates of the BDC (Butchart, 2014, and references therein). Yet, attempts to confirm the models' predictions with observation-derived datasets give contrasting results. Balloon-borne~~
75 Observational studies over short periods (typically 2003-2012) show significant evidence of a changing BDC in the boreal lower stratosphere (Schoeberl et al., 2008; Stiller et al., 2012; Hegglin et al., 2014; Mahieu et al., 2014; Haenel et al., 2015), but balloon-borne observations of SF₆ and CO₂ in the ~~Northern Hemisphere (NH) middle latitudes northern mid-latitudes~~ show a non-significant trend of the deep branch of the BDC in the past decades (Engel et al., 2009, 2017). ~~Studies over shorter periods and using other tracers or temperature observations show significant evidence of a changing BDC in the boreal lower stratosphere (Ray et al., 2014; Hegglin et al., 2014; Haenel et al., 2015).~~

~~A number of studies on the possible BDC changes compared observations, reanalyses and climate models (Mahieu et al., 2014; Garfinkel et al., 2014). The difficulty to derive~~ significant observational trends in the BDC can be partly attributed to the spatial and temporal sparseness of the observations, together with its large dynamical variability and the uncertainty of trends derived from non-linearly increasing tracers ~~(Garcia et al., 2011; Hardiman et al., 2017). Furthermore the observational datasets cannot discriminate between~~
85 ~~the separate effects of residual circulation and mixing. This separation turns out to be important in the BDC change studies in climate models (Garny et al., 2014; Ploeger et al., 2015; Eichinger et al., 2019) (Garcia et al., 2011; Hardiman et al., 2017; Fritsch et al., 2017).~~ Before investigating multi-decadal changes of the BDC, it is important to perform an accurate evaluation of its climatological state and inter-annual variability, which is the aim of this paper.

In this study we use ~~nitrous oxide (N₂O)~~ as a tracer to study the BDC. N₂O is continuously emitted in the troposphere
90 (with larger abundances in the Northern Hemisphere, NH), and transported into the stratosphere where it is destroyed by

photodissociation and, to a lesser extent, by reaction with $O(^1D)$. The estimated lifetime of N_2O is approximately 120 years, which makes it an excellent long-lived tracer for transport studies in the middle atmosphere (Brasseur and Solomon, 2006; Seinfeld and Pandis, 2016).

We use the Transformed Eulerian Mean (~~TEM~~) (~~Andrews et al., 1987~~) (TEM, Andrews et al., 1987) analysis to separate the local rates of change of N_2O due to transport and chemistry (Randel et al., 1994). The transport term can be further separated into the contribution of isentropic mixing and residual advection (~~Abalos et al., 2013; Tweedy et al., 2017~~) as done previously for O_3 and CO (Abalos et al., 2013). The isentropic mixing and the residual advection can be additionally separated in their horizontal and vertical contributions. ~~Here the horizontal mixing and the vertical advection are investigated~~ In the tropical lower stratosphere, the distinction between vertical and horizontal transport is important, as they impact differently the seasonality of N_2O in the northern and southern tropics (Tweedy et al., 2017). We choose to focus our study on the horizontal mixing and vertical advection, because their ~~magnitude is~~ magnitudes are larger than the vertical mixing and the meridional residual advection in most of the ~~stratospheric budget (Abalos et al., 2013).~~ stratosphere.

Chemistry Climate Models (CCMs) include the full representation of dynamical, radiative, and chemical processes in the atmosphere and their interactions. In particular, they combine the feedbacks of the chemical tracers on the heat budget and dynamics, that ultimately affects tracer transport. We use the Whole Atmosphere Community Climate Model version 4 (WACCM, Garcia et al., 2017) to simulate the N_2O TEM budget in the stratosphere for the 2005-2014 period, ~~and we compare the results with those obtained from several reanalysis datasets.~~ WACCM has been widely used for studies of tracers transport in the stratosphere and upper troposphere based on the TEM analysis (e.g. Abalos et al., 2017). WACCM simulations of the climatological N_2O over the 2005-2014 period have also been evaluated favourably with satellite observations in the stratosphere (Froidevaux et al., 2019).

~~Dynamical reanalyses~~ In order to assess their representation of the atmospheric processes, CCMs are often compared to reanalyses (e.g. Gerber et al., 2010). Reanalysis products merge dynamical atmospheric observations (e.g. surface pressure, wind, temperature) with a global forecast model using an assimilation scheme to offer the best reproduction of the past climate. They provide a multivariate, consistent record of the global atmospheric state. Reanalyses are made using different assimilation methods and forecast models (Cameron et al., 2019), and they are often compared among each other and with CCMs (~~Fujiwara et al., 2017; Cameron et al., 2019~~). ~~While dynamical reanalyses do not assimilate observations of chemical compounds, chemical reanalyses achieve this step, and can be used to evaluate CCMs or study differences between instruments using the reanalysis as a transfer tool (Inness et al., 2013; Davis et al., 2016)~~ (Rao et al., 2015). The SPARC (Stratosphere-troposphere Processes And their Role in Climate) Reanalysis Intercomparison Project (S-RIP) coordinates the intercomparison of all major global atmospheric reanalyses and provides reports to document these results (Fujiwara et al., 2017; Long et al., 2017).

~~The dynamical reanalyses are used here to drive 4 different simulations of the~~ Meteorological fields from reanalyses are often used to drive Chemistry-Transport Model (CTM) in order to study the BDC through a common diagnostic, namely the Age of Air (AoA, Waugh and Hall, 2002), and simulate realistic distributions of chemical tracers (Monge-Sanz et al., 2012; Ménard et al., 2020). Thanks to their simplicity, CTMs are useful to compare different reanalyses within the same transport framework, thereby contributing to the study of the BDC in S-RIP (chapter 5; see Fig. 1 in Fujiwara et al., 2017). CTMs may use either sigma-pressure

levels with a kinematic transport scheme and vertical velocities simply derived from mass conservation, or isentropic levels with a diabatic transport scheme (Chipperfield, 2006). Recent intercomparisons showed that the AoA depends to a large extent on the input reanalysis, both using the kinematic approach (Chabrillat et al., 2018) and the diabatic approach (Ploeger et al., 2019).

Here we use the same CTM as for the kinematic AoA study, i.e. the Belgian Assimilation System ~~for~~ of Chemical Ob-
servation (BASCOE) ~~Chemistry-Transport Model (CTM) (Chabrillat et al., 2018).~~CTM. Observations of another long-lived
stratospheric tracer, HCFC-22, were recently interpreted with WACCM and BASCOE CTM simulations, showing the interest
of this model intercomparison (Prignon et al., 2019). In order to contribute further to the S-RIP BDC activity, four different
dynamical reanalyses are used here to drive the BASCOE CTM simulations, compute the N_2O TEM budget and compare its
components with the results derived from WACCM. Namely we consider: the European Centre for Medium-Range Weather
Forecasts Interim Reanalysis (~~ERA-Interim~~)(ERA-Interim, Dee et al., 2011), the Japanese 55-year Reanalysis (~~JRA55~~)(JRA55, Kobayashi
the Modern-Era Retrospective analysis for Research and Applications version 1 (~~MERRA~~)(MERRA Rienecker et al., 2011),
and version 2 (~~MERRA2~~). WACCM4 is also compared to a chemical reanalysis of Aura Microwave Limb Sounder (MLS) using
the BASCOE assimilation system driven by the ERA-Interim reanalysis (BRAM2, Errera et al., 2019). (~~MERRA2 Gelaro et al., 2017~~).

~~WACCM has been widely used for studies of tracers transport in the stratosphere and upper troposphere based on the
TEM analysis (e.g. Abalos et al., 2013, 2017). CTMs driven by dynamical reanalyses are often used to investigate Age of Air
(e.g. Chabrillat et al., 2018; Ploeger et al., 2019). Chemical reanalyses~~ While dynamical reanalyses do not assimilate observations
of chemical compounds, chemical reanalyses achieve this step, and can be used to evaluate CCMs or study differences between
instruments using the reanalysis as a transfer tool (Errera et al., 2008; Lahoz and Errera, 2010; Davis et al., 2016). Chemical
reanalyses driven by meteorological fields from modern dynamical reanalyses have not been used to study ~~stratospheric~~ tracer
transport in the stratosphere using the TEM framework to our knowledge. ~~In WACCM and the CTM experiments are compared
with a chemical reanalysis of Aura Microwave Limb Sounder (MLS) using the BASCOE Data Assimilation System (DAS)
driven by the ERA-Interim reanalysis (BRAM2, Errera et al., 2019).~~

To summarize, in this study we analyze ~~and compare the effect of transport on the stratospheric using the state-of-the-art
CCM WACCM, together the representation of the BDC in WACCM through an analysis of the TEM budget of N_2O , and
we evaluate the simulation of this budget through comparisons~~ with the BASCOE CTM ~~driven by a variety of dynamical
reanalyses, (driven by four dynamical reanalyses)~~ and the BRAM2 chemical reanalysis.

In Section 2 we describe the datasets used in the study and the TEM analysis of N_2O . In Section 3.1 we analyse the seasonal
mean patterns of the TEM ~~budget in the different datasets~~ N_2O ~~budget in each dataset~~ and their differences. ~~In~~ Sections 4 and 5
~~respectively, investigate respectively~~ the mean annual cycle and ~~the~~ variability of the N_2O TEM budget terms ~~are studied~~, with
a focus on the differences between the datasets. Section ~~4 discusses the results pointing to the possible causes of disagreements
and section~~ 6 concludes the study with a summary of our findings and possible future research.

2 Data and ~~methods~~method

This work uses seven datasets that were generated by WACCM, the BASCOE CTM and the BASCOE DAS. Table 1 provides an overview of these datasets and their main differences, and the next three subsections provide details about the models and systems that generated them.

2.1 WACCM

WACCM (Garcia et al., 2017) is the atmospheric component of the Community Earth System Model version 1.2.2 (Hurrell et al., 2013), which has been developed by the U.S. National Center of Atmospheric Research. It is the extended (whole atmosphere) version of the Community Atmosphere Model version 4 (CAM4, Neale et al., 2013).

We ran one realization of the public version of WACCM (hereafter WACCM4, Marsh et al., 2013), that we downloaded with a similar setup (e.g. lower boundary conditions) as the CTM experiments; the source code of WACCM4 is available for download at https://svn-ccsm-models.cgd.ucar.edu/cesm1/release_tags/cesm1_2_2cesm1_2_2. In this study we also use 3 realizations of the REF-C1 simulation used in the SPARC (Stratosphere-troposphere Processes And their Role in Climate) Chemistry-Climate Model Initiative (CCMI, Morgenstern et al., 2017). The CCMI experiments, hereafter WACCM-CCMI, differ from WACCM4 for the modified gravity waves parameterization and the updated heterogeneous chemistry (Garcia et al., 2017). The inclusion of WACCM4 allows us to make a sensitivity test for the impact of the modified gravity waves parameterization on the simulation of the N₂O transport (see Sect. 4.4 for detailed analysis). We consider use three-dimensional daily-mean output over the 2005-2014 period to allow a fair comparison with the BRAM2 dataset (see Sec. 2.3 for detailed analysis). WACCM has a longitude-latitude grid of 2.5°x1.9° and 66 vertical levels ranging from the surface to about 140 km altitude. The vertical coordinate is hybrid-pressure, i.e. terrain-following below 100 hPa and purely isobaric above. The vertical resolution depends on the height: it is approximately 3.5 km above 65 km, 1.75 km around the stratopause (50 km), 1.1-1.4 km in the lower stratosphere (below 30 km), and 1.1 km in the troposphere. The time step for the physics in the model is 30 minutes.

The physics of WACCM is the same as CAM4 and the dynamical core is a finite volume with a horizontal discretization based on a conservative flux-form semi Lagrangian (FFSL) scheme (Lin, 2004). The gravity wave parameterization accounts for momentum and heat deposition separating orographic and non-orographic sources. The orographic waves are modified according to Garcia et al. (2017), while non-orographic waves are parameterized depending on the convection and the frontogenesis occurrence in the model (Richter et al., 2010).

~~The WACCM versions considered here~~ In this study, the considered WACCM versions are not able to internally generate the Quasi Biennial Oscillation (QBO, see e.g. Baldwin et al., 2001) internally, forcing it instead. Thus, the QBO is nudged by a relaxation of stratospheric winds to observations in the Tropics (Matthes et al., 2010). The solar forcing uses the Lean et al. (2005) approach.

WACCM includes a detailed coupled chemistry module for the middle atmosphere based on the Model for Ozone and Related Chemical Tracers, version 3 (MOZART-3) (Kinnison et al., 2007; Marsh et al., 2013). The species included within this mechanism are contained within the O_x, NO_x, HO_x, ClO_x and BrO_x chemical families, along with CH₄ and its degradation products. In addition, 20 primary non-methane hydrocarbons and related oxygenated organic compounds are represented along with their surface emission. There is a total of 183 species and 472 chemical reactions; this includes 17 heterogeneous reactions

on multiple aerosol types, i.e. sulfate, nitric acid trihydrate, and water-ice. In ~~WACM-CCMI~~ WACCM-CCMI the heterogeneous chemistry is updated by Solomon et al. (2015).

195 2.2 BASCOE CTM

The BASCOE data assimilation system (Errera et al., 2019) is built on a Chemistry-Transport Model, which consists in a kinematic transport module with the FFSL advection scheme (Lin and Rood, 1996) and an explicit solver for stratospheric chemistry, comprising 65 species and 243 reactions (Prignon et al., 2019). ~~The transport module requires on input only~~ Chabrillat et al. (2018) explain in detail the pre-processing procedure that allows the BASCOE CTM to be driven by arbitrary
200 reanalysis datasets, and the set-up of model transport. As usual for kinematic transport modules, the FFSL scheme only needs
the surface pressure and horizontal wind fields from reanalyses ~~, as it as input, because it is set on a coarser grid than the input~~
reanalyses, and relies on mass continuity to derive vertical mass fluxes corresponding to its own grid. Similar to Chabrillat et al.
(2018), the model is driven by four different reanalysis datasets on a common, low-resolution latitude-longitude grid ($2.5^\circ \times 2^\circ$),
but keeping their native vertical grids. In this way, we avoid any vertical regridding and the intercomparison explicitly accounts
205 for the different vertical resolutions.

The four input reanalyses are part of the SPARC Reanalysis Intercomparison Project (S-RIP) which is a coordinated inter-comparison of all major global atmospheric reanalyses. They are described in Fujiwara et al. (2017): the European Centre for Medium-Range Weather Forecasts Interim Reanalysis (ERA-Interim, hereafter ERAI; Dee et al., 2011), the Japanese 55-year Reanalysis (JRA55; Kobayashi et al., 2015), the Modern-Era Retrospective analysis for Research and Applications (MERRA;
210 Rienecker et al., 2011) and its version 2 (MERRA2; Gelaro et al., 2017). ERAI and JRA55 have 60 levels up to 0.1 hPa while MERRA and MERRA2 have 72 levels up to 0.01 hPa. The CTM time step is set to 30 minutes. ~~For this work the BASCOE~~
~~CTM provided~~ As for the WACCM experiment, we used the daily mean outputs from the BASCOE CTM over the 2005-2014
period ~~as for the WACCM experiment~~.

2.3 BASCOE Reanalysis

215 ~~The TEM diagnosis is also applied to assimilated fields from~~ BRAM2 is the BASCOE Reanalysis of Aura MLS, version
2 ~~(BRAM2, Errera et al., 2019)~~, which covers the period August 2004-August ~~2019~~ 2019 (Errera et al., 2019). For BRAM2,
BASCOE is driven by dynamical fields from ERA-Interim, with a horizontal resolution of $3.75^\circ \times 2.5^\circ$ longitude-latitude. The
vertical grid is represented by 37 hybrid-pressure levels which are a subset of the ERA-Interim 60 levels.

In BRAM2, N_2O profiles from the MLS version 4 standard product ~~has~~ have been assimilated within the 0.46-68 hPa
220 pressure ranges (Livesey et al., 2015). This ~~product dataset~~ is retrieved from the MLS 190 GHz radiometer instead of the 640
GHz radiometer in earlier MLS version. The 640 GHz radiometer, which provided a slightly better quality retrieval down to
100 hPa, ceased to be delivered after August 2013 because of instrumental degradation in the band used for that retrieval. To
avoid any artificial discontinuity due to switching from one product to the other in August 2013, BRAM2 has assimilated the
190 GHz N_2O during the whole reanalysis period.

225 BRAM2 N₂O has been validated between 3 and 68 hPa against several instruments with a general agreement between 15 % depending on the instrument and the situation of interest and between 3 and 68 hPa (see Errera et al., 2019) atmospheric region (the middle stratosphere or the polar vortex, see Errera et al., 2019). It is not recommended to use BRAM2 N₂O reanalysis outside ~~these pressure ranges. BRAM this pressure range.~~ BRAM2 N₂O is also affected by a small drift of around -4 % between 2005 and 2015 (~~see also Froidevaux et al., 2019; ?~~) (see also Froidevaux et al., 2019).

230 2.4 TEM diagnostics

For atmospheric tracers the TEM analysis (Andrews et al., 1987) allows to separate the local change of a tracer with volume mixing ratio χ in terms due to transport and chemistry (Eq. (1)).

$$\bar{\chi}_t = -\bar{v}^* \bar{\chi}_y - \bar{w}^* \bar{\chi}_z + e^{z/H} \nabla \cdot \mathbf{M} + \bar{S}, \quad (1)$$

where χ is the volume mixig ratio of N₂O, and \mathbf{M} is the eddy flux vector, defined as:

$$235 \quad M^{(y)} \equiv -e^{-z/H} (\overline{v' \chi'} - \overline{v' \theta'} \bar{\chi}_z / \bar{\theta}_z), \quad (2a)$$

$$M^{(z)} \equiv -e^{-z/H} (\overline{w' \chi'} + \overline{v' \theta'} \bar{\chi}_y / \bar{\theta}_z). \quad (2b)$$

~~and~~ v^* and w^* are the meridional and vertical components of the residual mean meridional circulation, and defined respectively as:

$$\bar{v}^* \equiv \bar{v} - e^{z/H} (e^{-z/H} \overline{v' \theta'} / \bar{\theta}_z)_z, \quad (3a)$$

$$240 \quad \bar{w}^* \equiv \bar{w} + (a \cos \phi)^{-1} (\cos \phi \overline{v' \theta'} / \bar{\theta}_z)_\phi. \quad (3b)$$

Where \bar{v} , \bar{w} and $\bar{\theta}$ are respectively the Eulerian zonal-mean meridional and vertical velocities and the potential ~~temperatures~~ temperature, ϕ is the latitude, and S is the net rate of change due to chemistry i.e. $\bar{S} = \bar{P} - \bar{L}$, where \bar{P} and \bar{L} are respectively the zonal-mean chemical production and loss rates. Overbar quantities represent zonal mean fields, primed quantities the departures from the zonal mean, and subscripts denote derivatives. Meridional derivatives are evaluated in spherical coordinates and vertical
245 derivatives with respect to log-pressure altitude $z \equiv -H \log_e(p/p_s)$, with $p_s = 10^5 Pa$ and $H = 7 km$.

Hence, transport is separated into ~~the~~ advection due to the residual circulation (first 2 terms on the right-hand side (RHS) of Eq. (1)) and ~~the~~ irrevlesible quasi-horizontal isentropic eddy mixing, $e^{z/H} \nabla \cdot \mathbf{M}$ (~~third term on the RHS of Eq. (1)~~).

In order to better understand the role of each term in the tracer balance it is useful to separate the components of the vector \mathbf{M} and rearrange the terms of Eq. (1):

$$250 \quad \bar{\chi}_t = A_y + M_y + A_z + M_z + (\bar{P} - \bar{L}) + \bar{\epsilon}, \quad (4)$$

where:

$$A_y = -\bar{v}^* \bar{\chi}_y, \quad (5a)$$

$$M_y = e^{z/H} \cos\phi^{-1} (M^{(y)} \cos\phi)_y, \quad (5b)$$

$$A_z = -\bar{w}^* \bar{\chi}_z, \quad (5c)$$

$$255 \quad M_z = e^{z/H} (M^{(z)})_z, \quad (5d)$$

with A_y representing the meridional residual advection, M_y the horizontal transport due to eddy mixing, A_z the vertical residual advection and M_z the vertical eddy mixing (all expressed in ppbv day⁻¹). It is important to note that the total mixing term ($M_y + M_z$) includes not only the effects of irreversible mixing, but also some effects of the advective transport which are not resolved by the residual advection (Andrews et al., 1987; Holton, 2004).

260 Before any TEM calculation all the input fields are interpolated to constant pressure levels from the hybrid-sigma coefficients, ~~hence retaining the that retain the same vertical resolution as the~~ original vertical grid of each dataset (Table 1). Each derivative is computed using a centered differences method.

In addition to the physical TEM terms (Eq. (1)), it is necessary to include an additional term on the RHS of Eq. (4): the residual term ϵ . It is the difference between the actual rate of change of χ (LHS of Eq. (4)) and the sum of all the transport and
265 chemical terms of the TEM budget.

~~The This~~ non-zero residual has several causes (Abalos et al., 2017). The TEM calculations for WACCM rely on ~~a diagnostic variable the diagnostic variable~~ w , which is not used to advect the tracers, because the model is based on a Finite Volume dynamical core (Lin, 2004). Furthermore in WACCM, an implicit numerical diffusion is added to the transport scheme in order to balance the small-scale noise without altering the large-scale. This numerical diffusion is not included in the TEM budget
270 and is larger in regions with large small-scale features, i.e. regions where gradients are ~~larger/~~stronger (Conley et al., 2012). All TEM calculations are done using daily mean data, ~~while even though~~ WACCM and BASCOE both run with a much smaller ~~timestep time step~~ of 30 minutes. ~~Finally, the The~~ daily mean fields are interpolated from their native hybrid-sigma levels to constant pressure levels prior to the TEM analysis. ~~This could lead, leading~~ to numerical errors in the lower stratosphere. ~~The BASCOE datasets have a coarser horizontal resolution than their input reanalyses (especially BRAM2; see Table 1). This affects the accuracy of the vertical and horizontal derivatives, with possible implications for the residual. The possible causes of the residual in the five reanalysis datasets are discussed in more detail in Sect. 3.1.~~ For WACCM-CCMI, the TEM budget is computed for each realization, allowing the examination of both the ~~multi-model ensemble~~ mean (e.g. for seasonal means) or the model envelope (e.g. for line plots). In order to validate our N₂O TEM budget, we reproduced the findings reported in Tweedy et al. (2017, Fig. 7) with WACCM-CCMI ~~in the tropical lower stratosphere~~, and we noticed similar results (not shown).
280 ~~Dynamical reanalyses provide realistic~~ ~~In order to interpret the TEM analysis of the N₂O budget, we also compute the Eliassen-Palm Flux Divergence (EPFD). The Eliassen-Palm flux is a 2-D vector defined as $\mathbf{F} \equiv (F^{(\phi)}, F^{(z)})$ (Andrews et al., 1987),~~

with its meridional and vertical components given respectively by:

$$F^{(\phi)} \equiv e^{-z/H} \text{acos}\phi (\overline{u_z v' \theta'} / \overline{\theta_z} - \overline{v' u'}), \quad (6a)$$

$$F^{(z)} \equiv e^{-z/H} \text{acos}\phi \{ [f - (\text{acos}\phi)^{-1} (\overline{u \cos\phi})_\phi] \overline{v' \theta'} / \overline{\theta_z} - \overline{w' u'} \}. \quad (6b)$$

285 The EPFD reflects the magnitude of the eddy processes, and provides a direct measure of the dynamical forcing of the zonal-mean state by the resolved eddies (Edmon et al., 1980).

The four dynamical reanalyses used in this study provide overall consistent temperature and winds in the stratosphere, but can lead to a different representation of large-scale transport (e.g. Chabrilat et al., 2018) due to the biases in the temperature and wind fields (Kawatani et al., 2016; Tao et al., 2019). Note that the TEM quantities are not directly constrained by observations.
290 The-, especially the upwelling velocity \bar{w}^* , that can vary considerably in the dynamical reanalyses (Abalos et al., 2015)-, as it is a small residual quantity (Abalos et al., 2015).

In the rest of the paper, we will assume that the BASCOE reanalysis-BRAM2 product provides the best available approximation of the TEM budget for N_2O , at least where the residual is smaller than the vertical advection and horizontal mixing terms. This assumption relies on the combination in BRAM2 of dynamical constraints from ERA-Interim with chemical constraints
295 from MLS (Errera et al., 2019).

In Figs. 1 and 2 we show the N_2O TEM budget terms at 15 hPa for all the datasets for the December-January-February (DJF) and June-July-August (JJA) means respectively. It is important to make this seasonal distinction because the TEM quantities are the strongest in the winter hemisphere (see Sect. 1).

The choice of the boreal winter (December-January-February, DJF mean) and summer (June-July-August, JJA mean)
300 respectively. The 15 hPa level (around 30 km) level is due to the large differences that altitude) was chosen because large differences can be found between WACCM-CCMI, BRAM2, and the CTM runs -, and the better assimilation of at this level, and because the dynamical reanalyses are not constrained as well by meteorological observations at this altitude with respect to higher levels (Manney et al., 2003).

Figure 1 shows the Figs. 1 and 2 aim to show how the dynamical and chemical terms of the budget balance each other to
305 recover the tendency $\bar{\chi}_t$ at different latitudes. The discussion about the differences between the datasets, and their possible physical causes, are addressed in the next Sections.

The vertical advection term A_z shows how the upwelling contributes to increasing the N_2O TEM budget in DJF at 15 hPa for the considered datasets. The Tropics are characterized by a abundances in the tropics and summertime mid-latitudes, and how polar downwelling contributes to decreasing the N_2O increase due to the upwelling -, balanced by a decrease mostly due to
310 the chemical loss and -, abundances in the winter hemisphere. The horizontal transport out of the tropics due to a smaller extent, meridional advection. In the northern tropics the eddies, as represented by M_y , reduces the N_2O decrease due to horizontal mixing is clearly compensated by A_y in WACCM-CCMI (Fig. 1(a)), while for the reanalysis datasets a positive residual term arises because A_y is not sufficient to do so. At the higher latitudes abundance in the tropical latitudes of the wintertime hemisphere, and increases the main terms contributing to the N_2O mixing ratio at high latitudes in the winter hemisphere. The

315 other terms of the TEM budget are weaker than A_z and M_y ; the meridional advection term A_y tends to increase the positive horizontal mixing term in the increase, and the negative vertical advection and vertical mixing terms for the N_2O decrease in all the datasets, with negligible contributions from the other terms (except for the residual term which plays a minor role especially in the reanalyses).

Figure 2 is the same as Fig. 1 but for the JJA season. In the northern Tropics the main terms contributing to the budget are the increase due to the vertical advection and abundance in the loss due to chemistry. In the southern tropics the pattern is noisier, with a general balance between the winter subtropics and extratropics, while the vertical transport term due to eddy mixing, M_z , decreases the N_2O mixing ratio over the northern polar latitudes, and the chemistry term $P - L$ shows that N_2O destruction by photodissociation and $\text{O}(^1\text{D})$ oxidation contributes to the budget in the tropics and also in the summertime hemisphere. All budget terms are weaker in the summer hemisphere than the winter hemisphere. Over the southern polar winter latitudes, the reanalyses deliver negative M_y and A_y , while, especially in the reanalyses, A_z and the residual terms play a minor role. In the southern mid-latitudes a large contribution is due to the negative A_z and the positive M_y ; the residual term plays a role in the balance of M_y in the reanalyses, especially in BRAM2 that are balanced by large positive residuals, which implies a less robust TEM balance (Fig. 2(f)). In the Southern polar region the. This is not the case with WACCM, where M_y tends to increase the N_2O distribution is affected mostly by a decrease due to the horizontal mixing and to the vertical advection to a smaller extent. M_z plays a role in decreasing mostly around 60 S in the reanalyses, while it increases in the WACCM-CCMI simulations. South of 60 S the WACCM-CCMI simulations do not show large contribution to the budget for any of the terms, while the reanalyses show a consistent decrease due to the horizontal mixing and balanced by a positive residual term. Such differences in the importance of M_y and the large residual term make the TEM analysis less robust in the antarctic region. abundance in the polar vortex. Such differences between the datasets are highlighted and discussed in the next sections.

335 3 TEM balances Latitude pressure cross sections

3.1 Cross sections

Figures 3 and 4 show respectively the DJF and JJA means of three contributions to the N_2O TEM budget, namely horizontal mixing M_y , vertical advection A_z and residual term ϵ , for WACCM-CCMI, JRA55, MERRA2 and BRAM2. The CTM results For those datasets, the remaining terms of the TEM budget (A_y , M_z and $P - L$) for DJF and JJA are shown respectively in supplemental figures S1 and S2. The full N_2O TEM budgets obtained with MERRA and ERAI are not shown because they are analogous to those obtained with MERRA2. for DJF and JJA are shown respectively in Figs. S3 and S4. In the case of WACCM-CCMI, the seasonal means were computed separately for each realization and we verified that the multi-model ensemble means show the same features as the individual realizations. Large differences arise in the dynamical terms of the TEM budget between summer and winter for both hemispheres in the extratropics. The strong seasonality of the deep branch of the BDC and of the transport barriers are the causes of these differences, as for the seasonal variations of the Age of Air spectrum (Li et al., 2012).

We also reproduced the results of Randel et al. (1994, Fig. 8) for the WACCM-CCMI multi-model mean and the reanalysis mean in DJF (Figs S5 and S6 respectively). The WACCM-CCMI and the reanalysis means agree with the Community Climate Model version 2 of the early 1990's with regard to the general pattern of the TEM terms, but both deliver stronger contributions, especially the reanalyses mean.

We first compare the contribution of ~~vertical advection~~ A_z across the datasets ~~in Figs. 3 and 4~~. The tropical upwelling increases the abundance of N_2O mostly in the mid-high stratosphere (between 1 and 15 hPa) with the maximum contribution in the summer tropics, while the downwelling decreases it mostly in the wintertime extratropics in the middle and low stratosphere (between 5 and 100 hPa). This reflects the path followed by the deep branch of the BDC (Birner and Bönisch, 2011). During ~~the DJF season~~ ~~boreal winter~~, these features are very similar across all datasets (Fig. 3), but noticeable differences appear during the ~~JJA season~~ ~~austral winter~~ (Fig. 4): the tropical upwelling has a ~~clearer~~ ~~larger~~ secondary maximum in the ~~reanalyses~~ (e.g. southern tropics with JRA55 and MERRA2) ~~than in WACCM-CCMI~~ ~~than with the other datasets~~, and the extra-tropical downwelling extends to the South Pole in WACCM-CCMI and JRA55 while it is mostly confined to the mid-latitudinal surf zone in the ~~four~~ other reanalyses. In the lower stratosphere, A_z shows the contribution of the residual advection by the shallow branch of the BDC to the N_2O abundances in the winter and summer hemispheres. The two-cell structure, consisting in upwelling of N_2O in the subtropics and downwelling in the extratropics, consistently agrees across all datasets. The meridional residual advection term A_y contributes to the poleward transport of air masses in the middle stratosphere, mostly during the winter, and its contribution to the N_2O TEM budget is weaker than A_z . A_y agrees well among the datasets in boreal winter (Figs. S1 and S3), while during austral winter WACCM-CCMI overestimates it around 30° S compared to the reanalyses (Figs. S2 and S4).

~~This~~ We move now to the mixing contributions to the N_2O budget. The horizontal mixing is the predominant contribution to the poleward tracer transport in the middle and lower stratosphere (Abalos et al., 2013), as it flattens the tracer gradients generated by the the residual advection. In the N_2O TEM budget during boreal winter, M_y mostly balances the extratropical downwelling and part of the the tropical upwelling (Figs 3 and S4). The surf zone is ~~also~~ characterized by strong ~~mixing~~ horizontal mixing, depicted here as large positive M_y contributions, and delimited by transport barriers which appear ~~here~~ as intense gradients of M_y in the winter ~~hemisphere~~ ~~hemispheres~~ (middle columns of Figs. 3 and 4). In the wintertime NH, the patterns of M_y are similar in all datasets (Fig. 3), but the effect of ~~irreversible~~ ~~horizontal~~ eddy mixing on N_2O is stronger in the reanalyses than in WACCM-CCMI. ~~The residual term~~ In Sect. 4 we analyze quantitatively the differences of the mid-stratospheric M_y between datasets. The residual terms in the reanalyses (right column of Fig. 3(f),(i),(l)) ~~presents larger values in correspondence to the transport barriers in the middle stratosphere, that tend to cancel the~~ are largest in the middle stratosphere at the latitudes of the transport barriers, and their signs are opposite to M_y contribution.

~~In the JJA season there is an important disagreement in~~ In the austral winter, over the Antarctic Polar cap and below 30 hPa, M_y ~~between~~ agrees remarkably well in all datasets (Fig. 4). Closer to the vortex edge and above 30 hPa, the wintertime decrease of N_2O is mainly due to downwelling in WACCM-CCMI ~~and the reanalyses~~ (middle column of), while the reanalyses, especially BRAM2, show that the horizontal mixing plays a major role (Fig. 4). The ~~transport barrier at the polar vortex edge (strong vertical gradients of zonal wind) can be clearly seen in the reanalyses as weak mixing (grey vertical lines)~~

(Haynes and Shuckburgh, 2000a), but impact of horizontal mixing on N_2O inside the wintertime polar vortex is not negligible (e.g. de la Cámara et al., 2013; Abalos et al., 2016a), as Rossby waves breaking occurs there as well as in the surf zone. In contrast with the reanalyses, in WACCM-CCMI has strong mixing there with very positive values of the M_y . Inside the Antarctic vortex and above 20 hPa, the reanalyses show a negative contribution of horizontal mixing to the budget whereas this contribution is very small in WACCM-CCMI. A physical interpretation of these patterns is not straightforward because the TEM budget is not fully closed in the SH polar regions in contribution is close to zero in the Antarctic vortex and maximum along the vortex edge (Fig. 4). This disagreement can be related to differences in the zonal wind: it is overestimated in WACCM above 30 km in subpolar latitudes compared to MERRA (Garcia et al., 2017) and the polar jet is not tilted equatorward as in the reanalyses (see black thin lines in Fig. 4, and Fig. 3 of Roscoe et al., 2012). Yet, the differences in M_y and A_z above the Antarctic in winter should be put into perspective with the relatively large residual terms that points to incomplete TEM budgets in the reanalyses (Fig. 4 and S4 right columns). Near the Antarctic polar vortex, the reanalyses as assumptions of the residual term is large (right column of Fig.4). The largest residual is encountered with BRAM2 in TEM analysis (such as small amplitude waves) are less valid leading to larger errors in the evaluation of the mean transport and eddy fluxes (Miyazaki and Iwasaki, 2005).

Since the relative importance of the outer part of the antarctic vortex in residual is considerable above the Antarctic in the next section we focus on a single level in the middle stratospheric to investigate quantitatively the disagreement between WACCM-CCMI and the reanalyses, accounting for the largest residual term (reanalyses (Fig. 4), it is necessary to better understand its physical meaning. Dietmüller et al. (2017) applied the TEM continuity equation to the Age of Air in CCM simulations. Computing the "resolved aging by mixing" (i.e. the AoA counterpart of $M_u + M_z$) as the time integral of the local mixing tendency along the residual circulation trajectories, and the "total aging by mixing" as the difference between the mean AoA (mAoA) and the residual circulation transit time, they defined the "aging by mixing on unresolved scales" (i.e. by diffusion) as the difference between the latter and the former. This "aging by diffusion", which can be related by construction to our residual term, arises around 60° S from the gradients due to the polar vortex edge. Even though we use a real tracer (N_2O), we find a qualitative agreement with this analysis based on AoA: our residual term is larger in regions characterized by strong gradients such as the antarctic vortex edge, and larger with dynamics constrained to a reanalysis than with a free-running CCM (see EMAC results in Fig. 1d by Dietmüller et al., 2017). We thus interpret the residual as the sum of mixing at unresolved scales and numerical errors (Abalos et al., 2017).

In the summertime lower stratosphere, we note a stronger contribution of M_u to the N_2O abundances above the subtropical jets in both hemispheres and for all datasets compared to higher levels in summer (Figs. 3 and 4 middle columns). This behavior is consistent with calculations of the effective diffusivity and age spectra (Haynes and Shuckburgh, 2000b; Ploeger and Birner, 2016). It is due to transient Rossby waves that cannot travel further up into the stratosphere due to the presence of critical lines, i.e. BRAM2).

3.1 Climatological seasonal cycles

415 ~~In this section we show the monthly mean climatological annual cycles of the~~ where the phase velocity of the wave matches
the background wind velocity, generally leading to wave breaking (Abalos et al., 2016b). In particular, above the northern
tropics during the boreal summer (Figs. 4, S2 and S4), the horizontal mixing is primarily associated with the Asian monsoon
anticyclone, and causes a decrease in N_2O ~~mixing ratio, and two transport terms that contribute to its time derivative:~~ (Konopka et al., 2010;
In the lower stratosphere, the contributions from M_y combine with that from A_z in the total impact of the shallow branch of
420 the BDC on N_2O all year round (Diallo et al., 2012).

The vertical mixing contribution M_z is very small during boreal winter, except in the middle and lower stratosphere poleward
of 60° N, where it tends to balance the M_y contribution (Figs. S1 and S3). In austral winter, there is a strong disagreement
between WACCM-CCMI and the reanalyses around 60° S between 5 and A_z , ~~for all the datasets at 15 hPa (around 30 km, Figs.~~
~~5 and 6).~~ As shown in the previous figures this level corresponds to the maximum values of both Figs. S2). WACCM-CCMI
425 simulates a strong M_z contribution at the polar jet core, that decreases the N_2O abundances and tends to balance M_y and,
while in the reanalyses M_z is weaker and increases N_2O in the higher stratosphere.

In the next section we focus on a single level in the middle stratosphere to study quantitatively the disagreement between
WACCM-CCMI and the reanalyses.

4 Climatological seasonal cycles

430 After investigating the seasonal means of A_z and M_y , it is interesting to examine their climatological mean annual cycles in
order to study the month-to-month variations over the year and their dependance on the latitude in the middle stratosphere.
The cycles are shown ~~separately in each hemisphere~~ for three latitude bands in each hemisphere corresponding to the Tropics
tropics (0° - 20°), the surf zones (40° - 60°) and the polar regions (60° - 80°). ~~The subtropical barriers are not shown because~~
~~of the large latitudinal gradients of M_y and A_z in these regions that would hinder the interpretation of their means.~~ For
435 WACCM-CCMI ~~we show,~~ we examine the envelope of the three model realizations in order to evaluate the role of the internal
variability and its relative importance for each month and latitude band. ~~The color codes for the four CTM simulations follow~~
~~the conventions of S-SRIP (Fujiwara et al., 2017). BRAM2 is depicted with a black line and symbols, as usually done for~~
~~observations, because it is constrained by both dynamical and chemical observations. Since the~~ In the following, we will
consider BRAM2 as the reference when comparing N_2O mixing ~~ratio in BRAM2 has been evaluated with a ratios between~~
440 datasets, because its dynamics and chemistry are both constrained to observational datasets.

4.1 Polar regions

The EPFD is often used to quantify the forcing of the wave drag due to resolved (planetary) waves (e.g. Gerber, 2012; Konopka et al., 2015).
We first show the monthly mean climatological annual cycles of EPFD averaged between 3 and 50 hPa, and the residual vertical
velocity w^* at 15 % uncertainty at 15 hPa (Errera et al., 2019), this is highlighted by a dark grey regions in top rows of Figs. 5
445 and 6). The light grey shading around the BRAM2 cycles represents the uncertainty arising from the residual term in the TEM

budget, i. e. it is entirely interpreted first as an uncertainty on A_z and then as an uncertainty on M_y hPa for the polar regions (60°-80° S and N, Fig. 5). We arbitrarily average the EPFD between 3 and 50 hPa in order to remain cautious.

identify the wave forcing for the deep branch of the BDC (Plumb, 2002; Konopka et al., 2015). However, the qualitative results do not depend on the choice of the lower boundary level. We first investigate the mixing ratio in the SH. In the Tropics (Fig. 5(e) and 6(a)), using the BRAM2 reanalysis of Aura MLS as reference, JRA55 and WACCM-CCMI agree very well, while ERAI, MERRA2 and MERRA underestimate the mixing ratio. WACCM-CCMI exhibits nearly no annual cycle, which is in clear disagreement. We also show one realization of the earlier version WACCM4 which suffered from a larger cold bias above the Antarctic (see Sect. 2.1). In WACCM-CCMI, the parameterization of gravity waves was adjusted in order to reduce this issue while not significantly changing the dynamics in the NH, that results in an enhanced polar downwelling above the southern polar region (Garcia et al., 2017). Above the Antarctic, the forcing from resolved waves peaks in October in the reanalyses, as a result of the vortex breakup that allows an enhanced wave activity compared to austral winter (Randel and Newman, 1998). The WACCM simulations miss this strong springtime peak, and they are in good agreement with the reanalyses. In the antarctic region in the rest of the year (Fig. 55(a)) the annual mean agrees well among all the datasets, but the springtime increase is smaller in all the simulations than in BRAM2. In all the latitude bands the WACCM-CCMI simulations are not outliers. The residual vertical velocity w^* above the Antarctic is shown in Fig. 5(c). This comparison between the WACCM versions was already shown in Garcia et al. (2017, Fig. 10), we repeat it here adding the dynamical reanalyses. In November-December the weaker downwelling in WACCM-CCMI agrees well with the reanalyses. Throughout the rest of the year WACCM-CCMI simulates a stronger downwelling than all reanalyses (also at lower levels, not shown). This difference raises the question whether the residual vertical velocity is correctly represented in WACCM-CCMI or in the dynamical reanalyses. Above the Arctic, the WACCM simulations underestimate the EPFD contribution during boreal winter compared to the reanalyses, while, regarding A_z and M_y , they differ with (Fig. 5(b)), and the downwelling velocities simulated by WACCM are weaker than the reanalyses in that period, with no significant differences between the WACCM versions (Fig. 5(d)). The differences between WACCM and the reanalyses in EPFD and w^* in the reanalyses, especially in the polar regions. This will be discussed in Section 4 will help the interpretation of the differences in A_z and M_y .

We then investigate the contribution from vertical advection in the SH, starting from the Tropics. Figure 6 shows the monthly mean climatological annual cycle of the N_2O mixing ratio, A_z in the 20, M_y for the polar regions (60°-0-80° S latitudinal band and N) at 15 hPa for all the datasets. First, we investigate the N_2O mixing ratio in the Antarctic region (Fig. 5(f)) is positive all year round showing the effect of tropical upwelling. As expected, the largest values are in the boreal late fall and winter (Seviour et al., 2012). The datasets show a general agreement, but 6(a)). During winter, the N_2O abundances are smaller than the rest of the year, because of the suppressed transport from the lower latitudes caused by the onset of the polar barrier. After the vortex breakup, the N_2O increase during spring and early summer is smaller in all the simulations than in BRAM2. In WACCM-CCMI, the modification of the parameterization of gravity waves results also in a shift towards earlier vortex breakup dates in the austral spring compared to WACCM4 (Garcia et al., 2017). The earlier vortex breakup in WACCM-CCMI underestimates A_z by up to $\sim 20\%$ in January and JRA55 overestimates it up to $\sim 50\%$ in November. In the Southern mid-latitudes (Fig. 5(e)) A_z is negative in all seasons except during summer and there is again a good agreement among the

datasets except for WACCM-CCMI and JRA55. These two datasets appear to have a purely annual cycle in this region, while the other four show a semi-annual component. The strongest contributions are reached in September for all reanalyses, with JRA55 almost twice more negative than the other ones. WACCM-CCMI, on the other hand, reaches its strongest contribution three months earlier (June) with A_z twice larger than obtained with BRAM2. allows the transport of N_2O -rich air from lower latitudes for a longer period compared to WACCM4, resulting in larger and more realistic simulations of the N_2O mixing ratios during austral spring and early summer (Fig. 6(a)).

In the antarctic region (Fig. 5(d)) A_z shows the effect of downwelling on, the downwelling decreases N_2O during most of the year. Here again (A_z term in Fig. 6(c)). Here, JRA55 and WACCM-CCMI are outliers: both present stronger A_z contributions in fall and winter, especially WACCM-CCMI reaching values three times lower stronger than BRAM2 in early winter, as a result of the stronger downwelling velocity simulated by WACCM-CCMI in that region. While this strong disagreement is questioned by the large residuals, we note that all the reanalyses confirm it except for JRA55. During fall and summer, A_z is stronger in WACCM-CCMI than in WACCM4, as a consequence of the stronger downwelling in WACCM-CCMI resulting from the modification of the gravity waves parameterization.

We now turn to the contribution from the horizontal mixing in the same hemisphere (Fig. 5 bottom row). In the Tropics (Fig. 5(i)) We now turn to the contribution from M_y , shows a decrease from May to October (when is transported to the middle latitudes), and a near-zero contribution in the rest of the year, generally common in all the considered datasets. The BRAM2 uncertainty is smaller than for the polar region (Fig. 5(g)) and middle latitudes (Fig. 5(h)) confirming the better performances of the TEM analysis outside the high latitudes. It is yet comparable with. In the antarctic region, M_y , because its contribution is quite small in this region, where the largest dynamical term is A_z (Fig. 5(f)). In the southern mid-latitudes (Fig. 5(h)) M_y increases throughout the winter, reflecting the mixing associated to the surf zone, and peaks in the early spring (September). During summer and early fall M_y does not contribute significantly to the TEM budget, and in November M_y reaches negative values which are comparable to the residual term. In WACCM-CCMI M_y starts increasing in February, i.e. two months earlier than the reanalyses, and the values remain twice larger during fall and winter, but they stop increasing in August, i.e. one month earlier than the reanalyses. In the antarctic region (Fig. 5(g)) M_y is very different among the datasets during winter: in BRAM2 it contributes to the N_2O decrease during fall and winter, with the strongest contribution in July, but with the CTM simulations this contribution is twice two times weaker, while in WACCM-CCMI the horizontal mixing has almost no effect on N_2O (Fig. 6(e)). During spring all the datasets show similarly positive values for M_y , with WACCM-CCMI presenting a large internal variability. As already mentioned, the TEM analysis suffers from large residuals in the wintertime antarctic region. Yet, we note that the disagreement between WACCM-CCMI and BRAM2 is significant, because in fall and winter the envelope of WACCM-CCMI realizations falls completely outside of the possible BRAM2 values when accounting for the residual.

In the NH (Fig. 6) the vertical range for A_z During the austral spring, the vortex breakup leads to an increased wave activity reaching the Antarctic, and M_y is extended with respect to the SH because of the larger values of the TEM terms above the Arctic, and the x axis is shifted by six months to better show the boreal winter. With regard to the mixing ratio, (Fig. 6 upper row) WACCM-CCMI and the CTM driven by JRA55 are in good agreement with BRAM2, while ERAI, MERRA2 and MERRA underestimate it.

In the tropics, A_z (Fig. 6(d)) shows the effect of the upwelling that transports from lower levels all year round, and in the middle latitudes (Fig. 6(e)) the wintertime downwelling to the lower stratosphere. The agreement is very good among the datasets in both latitude bands. The arctic region (Fig. 6(f)) is also characterized by the wintertime downwelling, which peaks in January, with JRA55 and ERAI showing a larger contribution ($\sim 30\%$ difference) than the other datasets.

520 Finally we consider the contribution of the horizontal mixing in the NH (Fig. 5 bottom row). In the Tropics from November to April (Fig. 6(g)) M_y is negative and presents a marked seasonality in the reanalyses that is much weaker in WACCM-CCMI. In the middle latitudes (Fig. 6(h)) the strong horizontal mixing in the surf zone tends to increase during winter. The reanalyses show a large spread, with values reaching ~ 1.5 in BRAM2 and ~ 0.9 in the MERRA runs, while WACCM-CCMI presents a large underestimation with respect to the reanalyses. In the arctic region (Fig. 6(i)) we note a seasonal cycle twice stronger than
525 in the mid-latitudes. Large discrepancies characterize this region: BRAM2 delivers the largest value better agreement among all datasets, the CTMs agree for a smaller contribution in late winter and spring, and compared to austral winter. Note that WACCM-CCMI significantly underestimates M_y as evaluated by all reanalyses. We also note that M_y in WACCM-CCMI has a larger exhibits large internal variability in the Arctic than in the other regions.

4.2 Interannual variability of the seasonal cycles

530 After reporting on the climatological annual cycles, it is desirable to estimate their inter-annual variability. To this end, we compute for each month the standard deviation of the mixing ratio, M_y and A_z across the ten simulated years. Figures 7 and 8 show the annual cycles of these standard deviations for each dataset, at the same pressure level and latitude bands as in previous figures, and using the same color code. The polar regions this season (Fig. 8) are separated from the other latitude bands (Fig. 7) because they are characterized by much larger inter-annual variability for M_y and A_z 6(e).

535 We first consider the variability of the mixing ratio (Figs. 7 and 8 top rows). The WACCM-CCMI simulations agree remarkably well with BRAM2, with the exception of the southern middle latitudes (Fig. 7(a)). In the northern mid-latitudes (Fig. 7(d)) the variability in WACCM-CCMI strongly depends on the considered realization. The CTM experiments generally underestimate the inter-annual variability of compared to BRAM2, with the exception of ERAI in the southern Tropics (Fig. 7(b)). Figures 7(f) and 7(g) show the inter-annual variability of A_z in the northern and southern Tropics respectively. The datasets disagree
540 more in these regions than in the other latitude bands, with WACCM-CCMI and JRA55 showing the smallest variabilities and BRAM2 and ERAI the largest. In the middle latitudes (Figs. 7(e) and 7(h)) A_z has the largest variability in winter and the summertime values are close to zero every year. In the antarctic region (Fig. 8(e)) the contribution of the upwelling to the abundances does not change much from year to year except in October during the vortex break-up, while in the arctic region (Fig. 8(d)) the wintertime inter-annual variability is larger.

545 We now move to the variability of the horizontal mixing term M_y starting from the Tropics (Figs. 7(j) and 7(k)). In the southern tropics (Fig. 7(j)) M_y shows generally a small inter-annual variability with larger values in the second part of the year in the reanalyses but not in WACCM-CCMI. In the northern tropics (Fig. 7(k)) M_y is variable mostly from November until May with a very good agreement among the reanalyses, except in January when it is much more variable in BRAM2. The variability of M_y in the northern tropics is clearly underestimated in WACCM-CCMI. In the mid-latitudes the variability

550 of M_y peaks in winter/spring in both hemispheres (Fig. 7(i) and (l)). WACCM-CCMI finds the same amount of variability in both hemispheres, while according to the reanalyses it is larger in the NH where the BRAM2 dataset is much more variable than all the others. In the antarctic region (Fig. 8(e)) M_y is highly variable in late winter and spring with a peak in October. All reanalyses agree, while in WACCM-CCMI the peak is reached one month earlier and the variability strongly depends on the model realization. The Arctic (Fig. 8(f)) is characterized by a very large wintertime variability of M_y . Among the reanalyses
 555 BRAM2 presenting the largest variability and MERRA the smallest. The variability with WACCM-CCMI is similar to the dynamical reanalyses and again depends on the model realization.

5 Discussion

We have described and compared the impact of the BDC on the rate of change in the chemical reanalysis BRAM2, three realizations and one sensitivity test with WACCM, and the BASCOE CTM driven by four different dynamical reanalyses.
 560 In summary, the present study reveals a good agreement at 15 hPa between WACCM-CCMI and BRAM2 in terms of the mid-stratospheric mixing ratio, while the CTM runs show a large spread (approximately 20%), especially in the Tropics (Figs. 5 and 6 top rows). The vertical advection term A_z agrees well across the datasets, except in the southern mid-latitudes and polar regions (Figs. 4 and 5 middle row). Large differences arise in the representation of M_y , in particular in the wintertime polar regions (Figs. 3 and 4). The inter-annual variability of the mixing ratio is in excellent agreement between WACCM-CCMI and
 565 BRAM2, while the CTM delivers overall smaller variabilities. We now discuss some possible causes of these results in light of the current literature about the modeling of the BDC.

4.1 Polar regions

It is interesting to highlight the differences between the wintertime ~~North Pole and South Pole~~ Arctic and Antarctic regions, because the hemispheric differences in wave activity (Scaife and James, 2000; Kidston et al., 2015) play a crucial role in
 570 the N_2O abundances and TEM budget. Above the Arctic ~~in the middle stratosphere the~~, the N_2O abundances simulated by WACCM agree with the BRAM2 reanalysis ~~and the advection term~~, except in December and January, and the CTM experiments driven by MERRA and MERRA2 deliver smaller N_2O mixing ratios compared to BRAM2 (Figs. 6(b)). A_z is also in good agreement between the datasets above the Arctic, with the exception of ERAI and JRA55 that provide stronger contributions (Fig. 6). ~~This region~~ 6(d)). The Arctic is characterized by a very variable polar vortex with a shorter life span than the antarctic
 575 vortex (Randel and Newman, 1998; Waugh and Randel, 1999), resulting in an enhanced contribution of the horizontal mixing to the N_2O budget during winter compared to the Antarctic (Fig. ~~6(i)~~ 6(f)). Compared to the ~~reanalyses~~ dynamical reanalyses and BRAM2, WACCM shows in ~~this region the Arctic~~ a 2-fold underestimation of the N_2O changes due to horizontal mixing during winter; ~~this can also be seen in the northern Tropics and middle latitudes (Fig. 6 bottom row).~~ We consider that this disagreement between WACCM and the reanalyses. Note that the Arctic extended winter presents the largest internal variability compared
 580 to the other regions, as shown by the spread in the WACCM realizations. The weaker contribution from M_y in WACCM is

meaningful because the relative importance of the residual term is small in the NH. ~~Note that WACCM is among the CCMI models with the lowest contribution of mixing to Age of Air (Fig. 2 in Dietmüller et al., 2018).~~

~~Above the Antarctic in the reanalyses, the relative importance of the residual is considerable (Figs. 4 and Fig. 5(g)) as it may cancel out most of the~~ The horizontal mixing is predominately influenced by the forcing from breaking of resolved (planetary) waves (Plumb, 2002; Dietmüller et al., 2018). In the Arctic region, WACCM underestimates the forcing from resolved waves compared to the dynamical reanalyses in the middle stratosphere (see Fig. 5(d)). This discrepancy in the resolved wave driving could contribute to the large differences in the wintertime M_y contribution. Hence it is necessary to better understand its physical meaning. Dietmüller et al. (2017) applied the TEM continuity equation to the Age of Air (AoA). Computing the "resolved aging by mixing" (i.e. the AoA counterpart of $M_y + M_z$) as the time integral of the local mixing tendency along the residual circulation trajectories, and the "total aging by mixing" as the difference between the mean AoA and between the CCM simulations and the CTM experiments above the Arctic. On the other hand, the residual circulation transit time, they defined the "role of different waves driving on mixing processes is an ongoing research topic, and additional data and sensitivity tests are needed in order to establish a clear separation of the waves contribution (e.g. gravity waves parameterization, spatial resolution, etc., Dietmüller et al., 2017). It should also be emphasized that WACCM is among the CCMI models with the lowest contribution of aging by mixing on unresolved scales" (i.e. by diffusion) as the difference between the latter and the former. This "aging by diffusion", which can be related by construction to our residual term, arises around 60 to Age of Air (Fig. 2 in Dietmüller et al., 2018).

4.1 Middle latitudes

Figure 7 shows the monthly mean climatological annual cycle of w^* at 15 hPa and EPFD averaged between 3 and 50 hPa over the surf zones (40° - 60° S from the gradients due to the polar vortex edge. Even though we use a real tracer (and N), and Fig. 8 shows the monthly mean climatological annual cycle of the N_2O) with a different TEM formulation, we find a qualitative agreement with this analysis based on AoA: our residual term is larger in regions characterized by strong gradients such as the Antarctic vortex edge, and larger with dynamics constrained to a reanalysis than with a free-running CCM (see EMAC results in Fig. 1d by Dietmüller et al., 2017). We propose to interpret the residual as the sum of mixing at unresolved scales and spurious numerical errors (Abalos et al., 2017) mixing ratio, A_z and M_y at 15 hPa averaged over the same latitudes. The subtropical barriers are not shown because M_y and A_z change sign in these regions, and averaging across them would hinder the interpretation of their means.

Over the Antarctic Pole and below 30 hPa, M_y agrees remarkably well in all datasets during winter (Fig. 4). Closer to the vortex edge and above 30 hPa, the wintertime decrease of in the middle stratosphere is mainly due to vertical advection in WACCM-CCM. In the southern mid-latitudes, the EPFD peaks in austral spring in the reanalyses, because of the enhanced wave activity in the Southern Hemisphere (SH) during austral spring compared to winter (Konopka et al., 2015), while the reanalyses, especially BRAM2, show that the horizontal mixing also plays a major role. WACCM simulations deliver an earlier and weaker peak during austral winter (Fig. 5). The impact of horizontal mixing on inside the wintertime polar vortex in the reanalyses should not be surprising: de la Cámara et al. (2013) and Abalos et al. (2016a) showed, using observations and reanalyses respectively, that the isentropic mixing is not negligible inside the vortex. In contrast with the reanalyses,

615 in WACCM-CCMI the M_y contribution is close to zero in 7(a)). The downwelling velocity w^* shows a similar pattern as the EPFD (Fig. 7(c)), as it is also driven by the breaking of resolved waves (Abalos et al., 2015). In the antarctic vortex and maximum along the vortex edge (Fig. 4). This disagreement can be related to differences in the zonal wind: it is overestimated above 30 km in subpolar latitudes (Garcia et al., 2017) and northern mid-latitudes, the polar jet is not tilted equatorward as in the EPFD peaks in winter in all the datasets, reflecting the stronger wave forcing in the surf zone in this season, and WACCM simulates lower EPFD values compared to the reanalyses (Fig. 4). Yet, the differences in 7(b)), that leads to a weaker downwelling velocity in the WACCM simulations (Fig. 7(d)). As for the polar regions, the differences in EPFD and w^* between the WACCM simulations and the reanalyses will help interpreting of the differences in A_z and M_y and A_z above the Antarctic in winter should be put into perspective with the large residual term that points to an incomplete TEM budget (Figs. 4 right column and 5 left column). Near the Antarctic polar vortex, the assumptions of the TEM analysis (such as small amplitude waves) are less valid leading to larger errors in the evaluation of the mean transport and eddy fluxes (Miyazaki and Iwasaki, 2005).

During the SH spring the vortex breakup leads to an increased wave activity reaching the Antarctic (Randel and Newman, 1998), and mid-stratospheric M_y is in better agreement among all datasets and WACCM-CCMI exhibits a larger internal variability. With regard to the N_2O mixing ratio in both hemispheres, the CTM driven by JRA55 and ERAI are in good agreement with BRAM2, while MERRA2 and MERRA underestimate it (Fig. 5(g)). Figure 5 also shows one realization of the earlier version WACCM4 which suffered from a larger cold bias above the Antarctic (see Sect. 2.1). In 8(a) and 8(b)). The WACCM-CCMI simulations agree well with the chemical reanalysis BRAM2, confirming the results obtained through the direct comparison with MLS observations (Froidevaux et al., 2019).

We now investigate the contribution from A_z and M_y . In the southern mid-latitudes, the parameterization of gravity waves was adjusted in order to reduce this issue while not significantly changing the dynamics in the NH. This results in an enhanced polar downwelling compared to WACCM4 (Garcia et al., 2017). Above the Antarctic the modification of the gravity waves in WACCM-CCMI results in a more realistic simulation of the mixing ratio in the austral spring compared to WACCM4 (Fig. 5(a)). While M_y does not show a significant impact of the gravity waves modification (Fig. 5(g)), A_z is stronger in negative in all seasons except during summer and there is again a good agreement among the datasets except for WACCM-CCMI than in WACCM4 (Fig. 5(d)), this is evidently due to the stronger downwelling in WACCM-CCMI. Figure 9(a) shows the climatological mean of the monthly residual vertical velocities (see Eq. (3b)) at 15 hPa over the Antarctic. This comparison between the WACCM versions was already shown in Garcia et al. (2017, Fig. 10), we repeat it here adding the reanalyses. In November-December the weaker downwelling in WACCM-CCMI agrees well with the reanalyses. Throughout the rest of the year WACCM-CCMI simulates a stronger downwelling than all reanalyses (also at lower levels, not shown). This difference raises the question whether the residual vertical velocity is correctly represented in WACCM-CCMI or in the dynamical reanalyses.

Above the Antarctic inter-annual variability of the vertical advection and the horizontal mixing terms is maximum during spring (Fig. 8) and JRA55 (Fig. 8(c)). 8), due to the large inter-annual variability in vortex breakup dates (Strahan et al., 2015). The lower wintertime variability would increase if a longer period was considered to include the exceptional Antarctic vortices of 2002 (Newman and Nash, 2005) and 2019 (Yamazaki et al., 2019). Above the Arctic the horizontal mixing and the vertical

650 ~~advection terms are most variable during~~ These two datasets appear to have a purely annual cycle in this region, while the other four show a semi-annual component. The peak in the A_z contribution in the reanalyses in September results from the increased forcing from the resolved waves (see Fig. 7(a)) and from the stronger contribution from gravity waves to the mass flux during spring (Sato and Hirano, 2019, Fig. 11). In the same region, M_y increases throughout the winter, reflecting the frequent disruptions of the northern polar vortex by sudden stratospheric warmings (Butler et al., 2017).

655 4.2 Middle latitudes

In both hemispheres the residual term is smaller than in the polar regions, i.e. the TEM budget is well-closed. The location of the subtropical transport barriers (around 30°N and S) and the extension of the northern surf zone compare well between WACCM-CCMI and mixing associated to the surf zone, and also peaks in early spring in the reanalyses (Figs. 3 and 4). Fig. 8(e)). During summer and early fall, M_y does not contribute significantly to the TEM budget, and in November M_y reaches negative values which are comparable to the residual term. In the summertime lower stratosphere, we note increases due to horizontal mixing above the subtropical jets in both hemispheres and for all datasets (Figs. 3 and 4 middle columns). This behavior is consistent with early calculations of the effective diffusivity (Haynes and Shuckburgh, 2000b). It is due to transient Rossby waves that cannot travel further up into the stratosphere due to the presence of critical lines (Abalos et al., 2016b).

In the mid-stratospheric SH both A_z and M_y peak in mid-winter in the WACCM-CCMI simulations, while in the reanalyses these maxima are reached three months later (Fig. 5(e)). This difference is related to the earlier minimum in the downwelling velocity \bar{w}^* simulated by WACCM-CCMI (Fig. 9(b) see Fig. 7(c)), that affects the vertical advection term directly affects A_z (Fig. 8(c)) and, by compensation, the horizontal mixing term M_y (Fig. 5(h) 8(e)). In the southern mid-latitudes Among the reanalyses, the compensating contributions of A_z and M_y are stronger for JRA55 than for the other reanalyses (up to twice larger in September, see Fig. 5(e), (h) 8(c) and 8(e)). This reflects the more intense BDC in JRA55 that resulted in the youngest mean AoA (mAoA) in the whole stratosphere (Chabrillat et al., 2018).

In the northern middle latitudes, A_z shows the effect of the wintertime downwelling to lower levels on N_2O , with the WACCM experiments simulating a slightly weaker contribution than the reanalyses (Fig. 8(d)). Such disagreement mostly originates from the weaker downwelling velocity in the CCM compared to the reanalyses showed in Fig. 7(b). In the northern mid-latitudes, the strong M_y contribution tends to increase the N_2O abundances in the surf zone during winter (Fig. 8(f)). The reanalyses show a large spread, with values reaching ~ 1.5 ppbv day $^{-1}$ in BRAM2 and ~ 0.9 ppbv day $^{-1}$ in the MERRA runs, and WACCM-CCMI presents a large underestimation with respect to the reanalyses. While the spread across the reanalyses cannot be explained by the forcing from the resolved waves, the weaker M_y contribution simulated by WACCM could be partly attributed to the weaker EPFD in the CCM compared to the reanalyses (see Fig. 7(d)).

4.2 Tropics

680 Figure 9 shows the climatological annual cycle for the N_2O mixing ratio, A_z and M_y for the southern and northern tropics (0°-20° S and N) at 15 hPa across all the datasets. The same latitude bands for the cycles of w^* and EPFD are shown in the Supplement (Fig. S7). In the tropical regions the, the N_2O mixing ratio in WACCM-CCMI agrees well with the reanalysis

of Aura MLS, while the CTM results show large differences in the N_2O ~~mixing-ratio-abundances~~ depending on the input reanalysis (Figs. 5(e) and 6(a) 9(a) and 9(b)). In regions where the ~~mAoA-AoA~~ is less than 4.5 years and N_2O is greater than 150 ppb, i.e. in the tropical regions and lower stratospheric middle latitudes (Strahan et al., 2011), the N_2O mixing ratio is inversely proportional to the mAoA, because faster upwelling (younger air) implies more N_2O transported from lower levels, decreasing its residence time and resulting in a limited chemical destruction (Hall et al., 1999; Galytska et al., 2019). The dynamical reanalyses also produce large differences in mAoA at 15 hPa: MERRA delivers the oldest mAoA and MERRA2, ERAI and JRA55 progressively show younger mAoA (Fig. 4(b) in Chabrillat et al., 2018). Hence, the large discrepancies in N_2O ~~vmr-mixing ratio~~ can be explained by the large differences in mAoA, while M_y and A_z contribute to rates of change of N_2O .

~~As in the middle latitudes, the TEM budget is well-closed in the tropical regions. The~~ We continue by investigating the contribution from A_z . In both the tropical regions, the upwelling term A_z is positive all year round showing the effect of tropical upwelling, and agrees very well in the reanalyses (Figs. 5(f) and 6(c) and 9(d)). ~~This can be related to,~~ as a result of the good agreement in the tropical upwelling velocity at 15 hPa according to (Fig. S7 bottom row), and also as depicted by mAoA diagnostics (Fig. 4(d) in Chabrillat et al., 2018). ~~In the southern tropics Large inter-hemispheric differences arise in the seasonality of A_z has a pronounced annual cycle in the middle stratosphere that is not present in the~~ between the tropical regions. The largest values of A_z in the southern tropics are in the boreal late-fall and winter (Fig. 9(c)), while no large seasonal variations can be detected in the annual cycle of the A_z in the northern tropics (Fig 9(d)). This is the result of the more pronounced seasonality of the upwelling velocity in the southern tropics compared to the northern tropics (Figs. 5(f) and 6(d) Fig. S7 bottom row).

We now turn to the contribution from M_y . In the southern tropics, M_y causes a decrease of the N_2O abundances from May to October (when N_2O is transported to the middle latitudes), and has a near-zero contribution in the rest of the year, generally common in all the considered datasets (Fig. 9(e)). ~~As found previously with GEOSCCM (Tweedy et al., 2017), no such asymmetry can be seen in the lower stratosphere (not shown)~~ The BRAM2 uncertainty is smaller than for the polar region and middle latitudes indicating a better performances of the TEM analysis outside the high latitudes. In the northern tropics, M_y is negative from November to April and presents a marked seasonality in the reanalyses that is much weaker in WACCM-CCMI (Fig. 9(f)). With respect to inter-hemispheric differences in M_y , WACCM disagrees with the reanalyses: according to WACCM, M_y has a larger impact in the southern tropics than in the northern tropics, but according to the reanalyses M_y has a much larger impact in the northern tropics (Figs. 5(i) and 6(e) 9(e) and 9(f)). These inter-hemispheric differences in the M_y contributions can be partly attributed to different forcings from the resolved waves between northern and southern tropics. The EPFD presents a stronger seasonality in the northern tropics than in the southern tropics in all the datasets (Fig. S7 top row), that could partly explain the differences in the seasonality of M_y in the reanalyses, but it does not impact the M_y simulated by WACCM.

5 Interannual variability of the seasonal cycles

715 To analyse the inter-annual variability of the annual cycle, we compute for each month the 1-sigma standard deviations of the N_2O mixing ratio, M_y and A_z across the ten simulated years. Figure 10 shows the annual cycles of these standard deviations for each dataset in the polar regions (60° - 80° S and N) at 15 hPa for both hemispheres. First, we consider the variabilities of the N_2O mixing ratio. In the Antarctic, during austral winter and early spring the year-to-year change of the N_2O abundances is very small (Fig. 10(a)), because duration, extension and strength of the polar vortex are very stable in a climatological sense, isolating air masses in the vortex from the highly variable mid-latitudes (Vaugh and Randel, 1999). The variability of the N_2O mixing ratio increases in October i.e. during the breaking vortex period that is highly variable in time (Strahan et al., 2015). Furthermore, the mid-latitude air masses, which have more variable composition, become free to reach the higher latitudes during this period. In the arctic region, the inter-annual variability of the N_2O mixing ratio is also largest during springtime but this is spread over a longer period, i.e. from February to June (Fig. 10(b)), reflecting the large interannual variability in the duration, extension and zonal asymmetry of the Arctic polar vortex (Vaugh and Randel, 1999). In both polar regions, WACCM-CCMI agrees well with BRAM2 while the CTM experiments simulate a smaller variability.

We now look at the interannual variability of A_z and M_y in the polar regions. Above the Antarctic, the inter-annual variabilities of A_z and M_y are maximum during spring (Figs. 10(c) and 10(e)), due to the large inter-annual variability in vortex breakup dates (Strahan et al., 2015). While the maximum variability of M_y is consistently reached in October in all the reanalyses, WACCM-CCMI simulates an earlier maximum (September) that does not correspond with the maximum in its mean values. The lower wintertime variability of both A_z and M_y would increase if a longer period was considered to include the exceptional Antarctic vortices of 2002 (Newman and Nash, 2005) and 2019 (Yamazaki et al., 2019). Above the Arctic, M_y and A_z are most variable during winter, reflecting the frequent disruptions of the northern polar vortex by sudden stratospheric warmings (SSWs, Butler et al., 2017). A case study of the effect of a SSW on the N_2O TEM budget showed that A_z and M_y contribute more to this budget during the SSW event than in the seasonal mean (Randel et al., 1994). Thus, the large wintertime variabilities of A_z and M_y are explained by the occurrence of seven major SSWs detected in the reanalyses for the 2005-2014 period (Butler et al., 2017).

~~In the Tropics the~~ In Fig. 11 we show the inter-annual variabilities of the N_2O mixing ratio, M_y and A_z for each dataset in the surf zones (40° - 60° S and N) at 15 hPa for both hemispheres. Regarding the N_2O mixing ratio, the inter-annual variability in the southern middle latitudes reaches the lowest values during austral winter. The datasets deliver very diverse values, with WACCM showing the largest variability and JRA55 the lowest across the climatological year (Fig. 11(a)). In the northern mid-latitudes, the inter-annual variability of the N_2O mixing ratio ~~depends~~ increases in late winter across all the datasets, as a response to the increased wintertime variability of the surf zone (Fig. 11(b)). The variability of WACCM-CCMI largely depends on the considered realization, except in October and November. Strong differences between ensemble members with respect to inter-annual variability indicate that the considered period is not long enough to explore the inter-annual variability in the northern mid-latitudes, and that the mean variability from this ensemble (with only three members) would not be representative of the internal variability of WACCM. The inter-annual variabilities of A_z and M_y in the southern mid-latitudes are shown in Figs. 11(c) and 11(e) respectively. As their mean value, A_z and M_y are most variable during austral spring and late summer in the reanalyses, while WACCM simulates an earlier peak during winter in the inter-annual variabilities of A_z and M_y compared

750 to the reanalyses. In the northern mid-latitudes, the inter-annual variabilities of A_z and M_y peak in winter, as their mean values, and WACCM simulates smaller variabilities compared to the reanalyses (Fig. 11(d) and 11(f)).

Figure 12 shows the annual cycles of the standard deviations of the N_2O mixing ratio, M_y and A_z for each dataset in the tropical regions (0° - 20° S and N) at 15 hPa for both hemispheres. The inter-annual variability of the N_2O mixing ratio in both southern and northern tropics depends considerably on the dataset (Figs. 7(b) and 7(e), 12(a) and 12(b)). WACCM-CCMI and the BASCOE reanalysis of Aura MLS show very similar variabilities, especially in the southern tropics. Since the QBO is the major source of variability in the tropical stratosphere (Baldwin et al., 2001), this confirms an earlier comparison that showed a good agreement between the WACCM model and MLS observations in the middle stratosphere in terms of the inter-annual variability of N_2O due to the QBO (Park et al., 2017). Among the CTM simulations, ERAI succeeds to deliver $\sigma(\bar{X})$ as large as BRAM2 and WACCM-CCMI in the southern tropics, but not in the northern tropics.

760 The inter-annual variability of A_z (Figs. 7(f) and 7(g)) in both hemispheres can be related to the impact of the QBO on the tropical upwelling (Flury et al., 2013). Among MERRA, ERAI and JRA55 the fraction of variance in deseasonalized tropical upwelling \bar{w}^* that is associated with the QBO is the largest with ERAI (Abalos et al., 2015). Our findings support this conclusion since the largest $\sigma(\bar{A}_z)$ among the reanalyses is again found with ERAI (Fig. 7). Figs. 12(c) and 12(d). However, a detailed analysis of the impact of the QBO on the BDC as illustrated here goes beyond the scope of this study. The variability of M_y in the tropical regions is small compared to the extratropical regions (Figs. 12(e) and 12(f)), in agreement with calculations of effective diffusivity that show small variabilities within the tropical pipe (Abalos et al., 2016a). The reanalyses deliver a larger inter-annual variability in the northern tropics during boreal winter, while in the southern tropics the variability of M_y presents a much weaker annual cycle. WACCM-CCMI does not reproduce this hemispheric asymmetry, with a rather flat profile in both hemispheres and a clear underestimation in the northern tropics, as shown for its mean values. In the tropical regions, both the variabilities of M_y and A_z fail to explain the good agreement in the variability of N_2O between WACCM and BRAM2, as well as their disagreement with the dynamical reanalyses, because M_y and A_z directly contribute to the N_2O tendency rather than its mixing ratio.

6 Summary and Conclusions

We have evaluated the climatological (2005-2014) N_2O transport processes in the stratosphere using the tracer continuity equation in the TEM formalism. In particular we ~~considered-emphasized~~ the horizontal mixing and the vertical advection terms (M_y and A_z respectively). The upwelling term A_z reduces the N_2O concentrations in the ~~Tropics and increases it~~ tropics and increases them in the extratropics, while M_y tends to reduce the meridional gradients of N_2O and presents large hemispheric differences. Since M_y or A_z contribute to the local ~~and instantaneous~~ rates of change of N_2O , this analysis is complementary to time-integrated diagnostics such as mAoA ~~or the mixing ratio itself~~. The comparison investigates a variety of datasets, from a free-running chemistry-climate model to a reanalysis where dynamics and chemistry are both constrained. The former comprises three realizations of the CCMI REF-C1 experiment by WACCM, and the latter is the chemical reanalysis of

Aura MLS driven by ERA-Interim: BRAM2. The intercomparison also includes the BASCOE CTM driven by four dynamical reanalyses: ERAI, JRA55, MERRA and MERRA2 in order to contribute to the S-RIP.

Considering the N_2O mixing ratio in the middle stratosphere, all datasets agree in the annual cycle, with the large spread
785 in the N_2O abundances of the CTM experiments ($\sim 20\%$) reflecting the diversity of mAoA obtained with the same model
(Chabrillat et al., 2018). The upwelling term A_z also agrees among the datasets, especially in the NH where WACCM follows
closely the reanalyses. ~~With respect to the~~ The horizontal mixing term M_y , in the NH ~~WACCM simulates a weaker impact is~~
~~weaker in WACCM~~ compared to the reanalyses. In the ~~southern tropics and middle latitudes the wintertime M_y is stronger in~~
~~WACCM than in extratropics, this could be attributed to the weaker forcing from the planetary waves in WACCM compared~~
790 ~~to~~ the reanalyses. The differences in M_y become striking in the wintertime Antarctic, where the polar vortex has a major role.
According to the reanalyses, the horizontal mixing plays an important role in that region, but that is not found by WACCM ~~and~~
~~However,~~ this large wintertime M_y in the reanalyses is challenged by a nearly as large residual term. It should be noted that
the residual term also includes effects from mixing by diffusion. An additional WACCM run with different gravity waves in
the SH is used as a sensitivity test. ~~This Over the Antarctic, this~~ test has small impact on ~~the horizontal mixing term M_y ,~~ but
795 significantly modifies ~~the vertical advection term A_z and the~~ ~~in the austral fall and winter due to the enhanced downwelling,~~
~~and the~~ N_2O mixing ratio ~~in the Antarctic during spring as a consequence of a more realistic timing of the vortex breakup.~~

The inter-annual variability of the mid-stratospheric horizontal mixing term M_y is largest in the polar regions. In the Antarc-
tic it is related to the ~~vortex breakup variability in the vortex breakup dates~~ during spring, while in the Arctic it is related to
the ~~very highly~~ variable polar vortex in winter. The inter-annual variability of A_z is characterized by a large spread in the
800 mid-stratospheric tropical regions where WACCM-CCMI and JRA55 deliver a smaller contribution than the other reanalyses.
This variability reflects the impact of the QBO on the tropical upwelling (Abalos et al., 2015).

The application of the TEM framework to tracer transport with reanalyses suffers from a poor closure of the budget in
the polar regions. We chose to analyse these regions nonetheless because the differences in M_y between WACCM and the
reanalyses are larger than the residual term, but it remains important to better understand the causes of these large uncertainties.
805 To this end, detailed studies of transport in the polar stratosphere are needed, e.g. comparing the residual circulations with
indirect estimates derived from momentum and thermodynamic balances, and evaluating the effective diffusivity in each dataset
(Abalos et al., 2015, 2016a).

The next step of this reasearch consists in the analysis of the inter-annual variations of the BDC, including the impact of
the QBO and the El-Nino Southern Oscillation. Further extensions of this work would include the addition of new reanalysis
810 products such as ERA5 and an intercomparison of several CCMs as already done for the residual circulation itself (Chrysanthou
et al., 2019).

Data availability. The 9 monthly climatologies of the N_2O mixing ratios and TEM budget terms are freely available at the BIRA-IASB
repository (<http://repository.aeronomie.be>) under <https://doi.org/10.18758/71021057>.

815 *Author contributions.* DM, SC and EM designed the study. YC provided support in installing and running the models. QE provided the chemical reanalysis BRAM2 and helped in its interpretation. MP ran the CTM experiments. DK provided the WACCM-CCMI realizations and helped in the interpretation of the WACCM datasets. DM wrote and ran the software tools to compute the TEM budgets and realized all the figures. DM, MA and SC analyzed the TEM budgets. DM and SC wrote the text. All co-authors contributed to the interpretation of the results and the reviews of the draft manuscripts.

Competing interests. The authors declare that they have no conflict of interest.

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 825 Rolando Garcia in the discussion of the paper specifically for the model results of WACCM.

<u>Dataset name</u>	<u>Reference</u>	<u>Dynamical Reanalysis</u>	<u>Chemical reanalysis of</u>	<u>Model grid</u>	<u>Top level</u>
<u>WACCM4</u>	<u>Marsh et al. (2013)</u>	<u>none</u>	<u>none</u>	<u>2.5°x1.9°, L66</u>	<u>5.1x10⁻⁶ hPa</u>
<u>WACCM-CCMI</u>	<u>Garcia et al. (2017)</u>	<u>none</u>	<u>none</u>	<u>2.5°x1.9°, L66</u>	<u>5.1x10⁻⁶ hPa</u>
<u>ERA1</u>	<u>Chabrilat et al. (2018)</u>	<u>ERA-Interim (Dee et al., 2011)</u>	<u>none</u>	<u>2.5°x2°, L60</u>	<u>0.1 hPa</u>
<u>JRA55</u>	<u>Chabrilat et al. (2018)</u>	<u>JRA-55 (Kobayashi et al., 2015)</u>	<u>none</u>	<u>2.5°x2°, L60</u>	<u>0.1 hPa</u>
<u>MERRA</u>	<u>Chabrilat et al. (2018)</u>	<u>MERRA (Rienecker et al., 2011)</u>	<u>none</u>	<u>2.5°x2°, L72</u>	<u>0.01 hPa</u>
<u>MERRA2</u>	<u>Chabrilat et al. (2018)</u>	<u>MERRA2 (Gelaro et al., 2017)</u>	<u>none</u>	<u>2.5°x2°, L72</u>	<u>0.01 hPa</u>
<u>BRAM2</u>	<u>Errera et al. (2019)</u>	<u>ERA-Interim (Dee et al., 2011)</u>	<u>MLS (Livesey et al., 2015)</u>	<u>3.75°x2.5°, L37</u>	<u>0.1 hPa</u>

Table 1. Overview of the datasets used in this study.

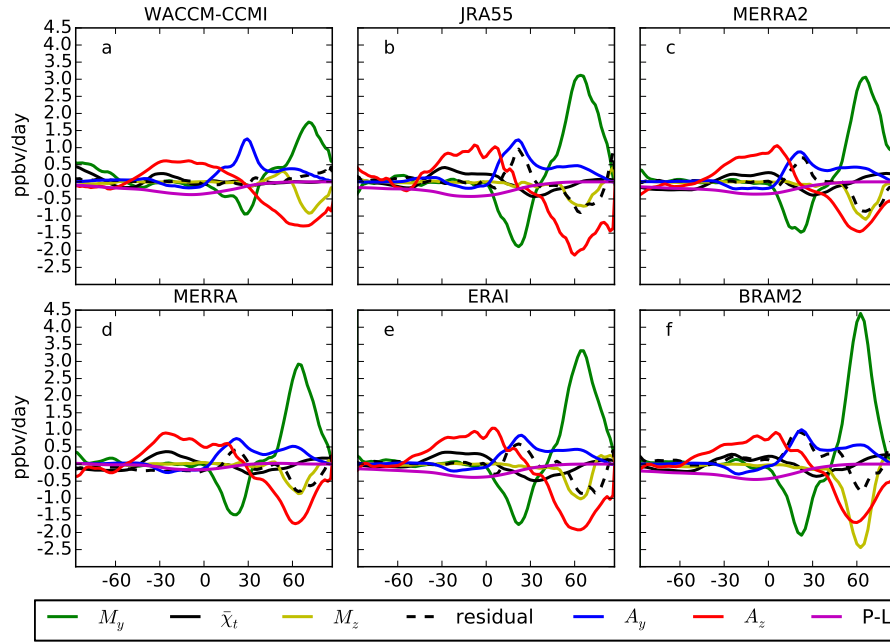


Figure 1. Latitudinal profiles of the N_2O TEM budget terms at 15 hPa averaged in DJF (2005-2014). Top row (left to right): WACCM-CCMI (a), JRA55 (b) and MERRA2 (c); bottom row (left to right): MERRA (d), ERAI (e) and BRAM2 (f). The color code is shown in the legend. Units are ppbv day^{-1}

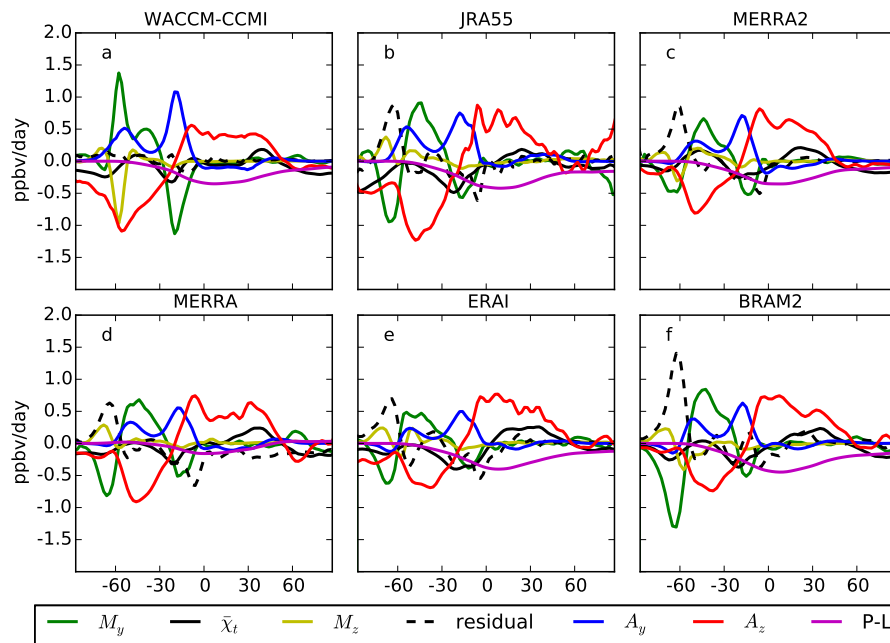


Figure 2. Same as previous figure but for JJA.

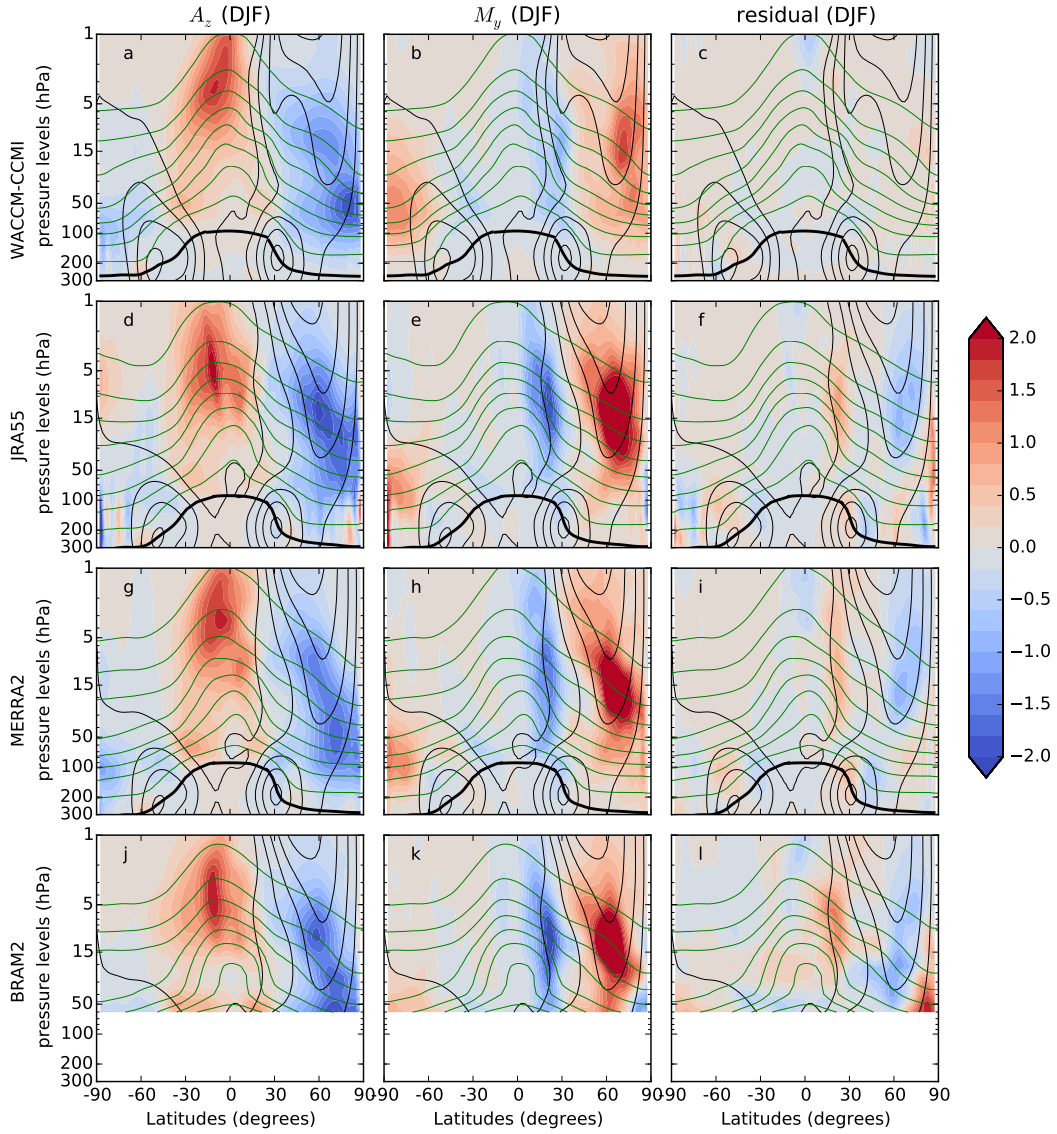


Figure 3. Climatological (2005-2014) latitude-pressure cross sections of three N₂O TEM budget terms averaged in DJF (ppbv day⁻¹): horizontal mixing term (left column), vertical residual advection term (central column) and residual term (right column). The datasets are, from top to bottom: WACCM-CCMI, JRA55, MERRA2, and BRAM2. The residual term for WACCM-CCMI is from a single realization of the model. The thin black lines show the zonal mean zonal wind (from 0 to 40 m/s every 10 m/s), the black thick line represents the dynamical tropopause for the considered season and the green thin lines show the climatological mixing ratio of N₂O (from 20 to 300 ppbv with 40 ppbv spacing).

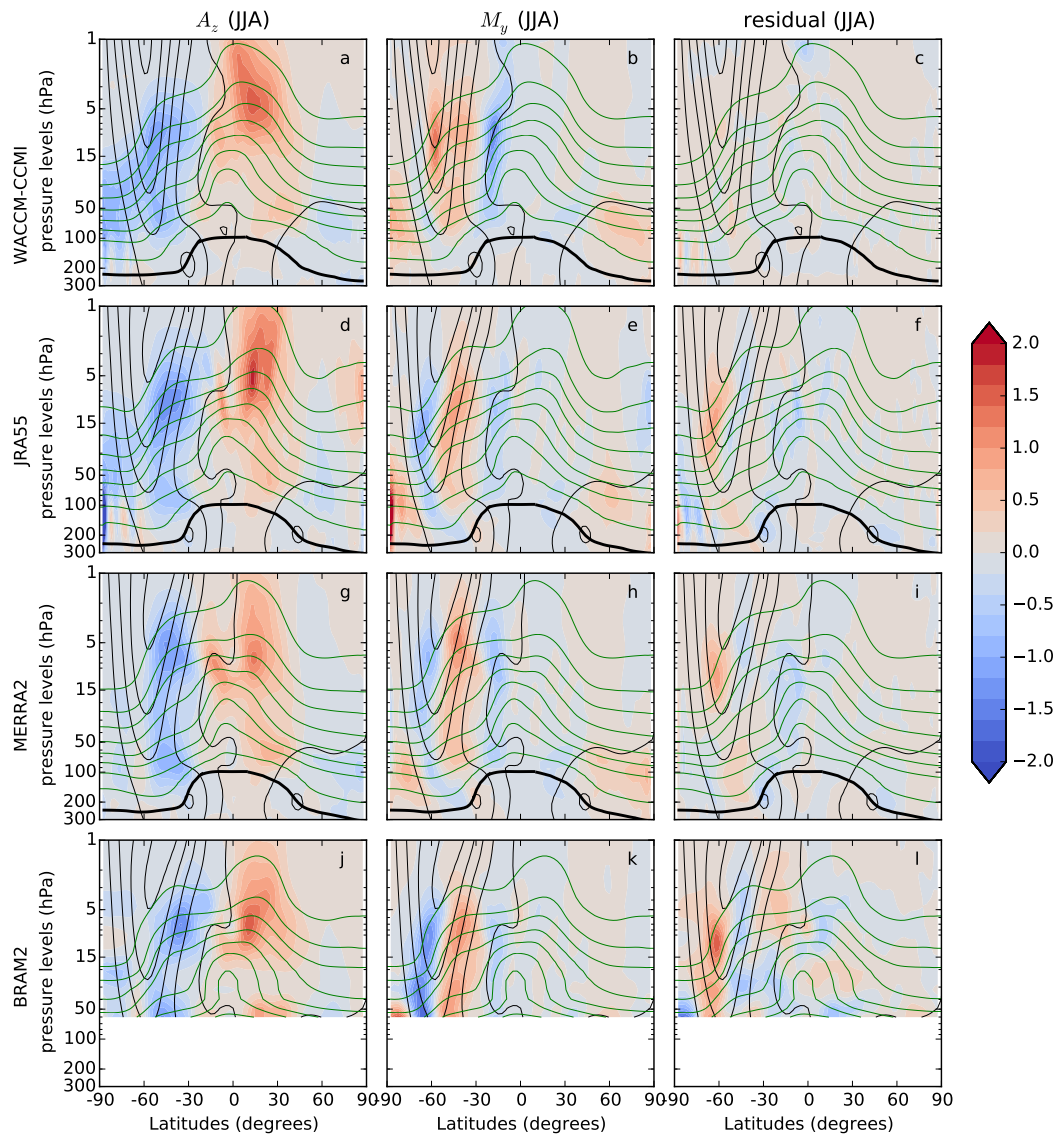


Figure 4. Same as previous figure but for JJA and with a different color scale. The thin black contours show the zonal mean zonal wind (from 0 to 100 m/s every 20 m/s).

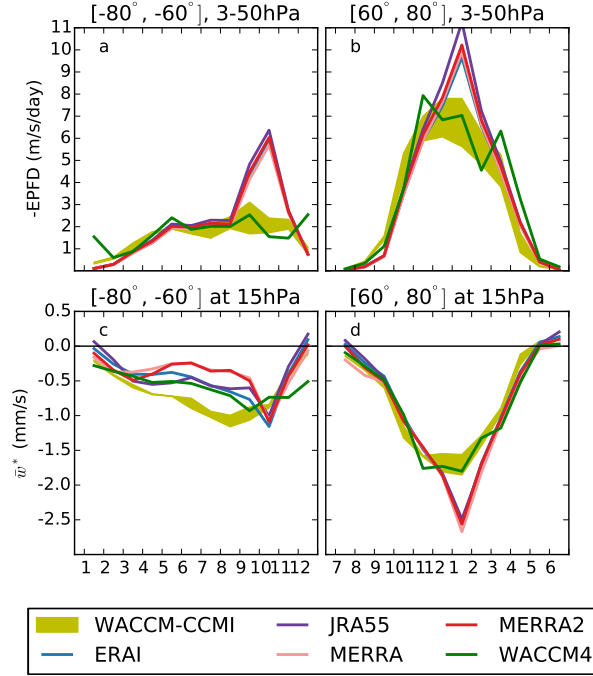
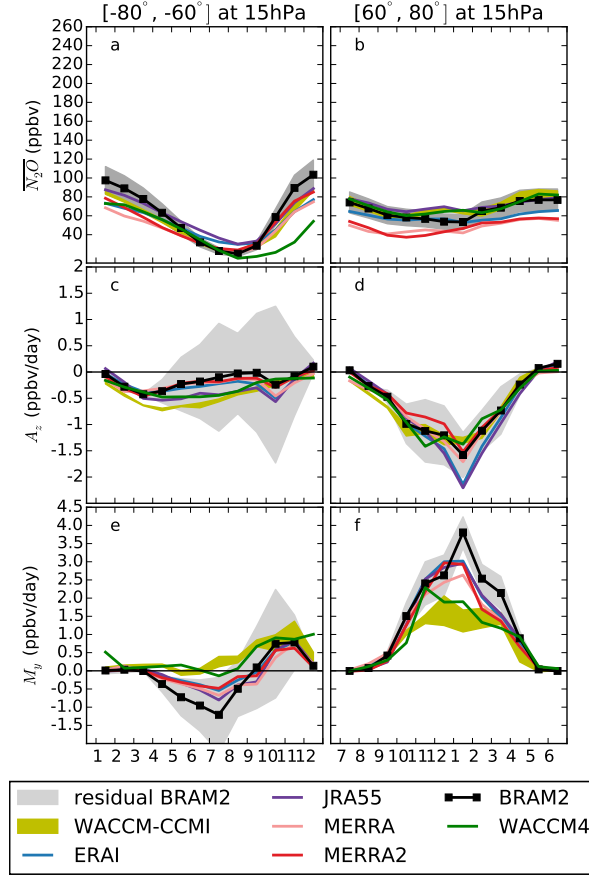


Figure 5. Monthly mean annual cycles at 15 hPa in the SH. First row: volume mixing ratio cycle of $-EPFD$ [$\text{m s}^{-1} \text{ day}^{-1}$]; second averaged between 3 and 50 hPa (upper row: horizontal mixing term); and \bar{w}^* [mm s^{-1}]; third at 15 hPa (bottom row: vertical residual advection term). Left column: polar-Antarctic region (60° - 80° S); middle. Right column: mid-latitudes-Arctic region (40° - 60° S); right column: Tropics (0 - 20° S). The color code is shown in the legend. The olive-yellow envelope shows the 3 realizations of the WACCM-CCMI simulation. The dark grey shading (top row) shows 15% uncertainty around BRAM2. The light grey shading (middle and bottom rows) shows BRAM2 plus and minus the residual term.



Monthly-standard-deviation Annual cycles over 2005-2014 at 15 hPa. First row: N_2O volume mixing ratio [ppbv], second row: horizontal-mixing-term M_y [ppbv day⁻¹]; third row: vertical-residual-advection-term A_z [ppbv day⁻¹]. From-left-to-right Left column: southern mid-latitudes Antarctic region (40°S-60°S), southern-tropics-, Right column: Arctic region (60°N-80°N), northern-tropics (0-20°N), northern mid-latitudes (40-60°N). The color-code is shown in the legend vertical scale differs for M_y and A_z . The yellow-olive envelope shows the 3 realizations of the WACCM-CCMI simulation. The color codes for the four CTM simulations follow the conventions of S-SRIP (Fujiwara et al., 2017). BRAM2 is depicted with a black line and symbols, as usually done for observations, because it is constrained by both dynamical and chemical observations. Since the N_2O mixing ratio in BRAM2 has been evaluated with a 15% uncertainty (1-sigma standard deviation) at 15 hPa (Errera et al., 2019), this is highlighted by a dark grey region in top rows. The light grey shading around the BRAM2 cycles represents the uncertainty arising from the residual term in the TEM budget, i.e. it is entirely interpreted first as an uncertainty on A_z and then as an uncertainty on M_y in order to remain cautious.

Monthly-standard-deviation Annual cycles over 2005-2014 at 15 hPa. First row: N_2O volume mixing ratio [ppbv], second row: horizontal-mixing-term M_y [ppbv day⁻¹]; third row: vertical-residual-advection-term A_z [ppbv day⁻¹]. From-left-to-right Left column: southern mid-latitudes Antarctic region (40°S-60°S), southern-tropics-, Right column: Arctic region (60°N-80°N), northern-tropics (0-20°N), northern mid-latitudes (40-60°N). The color-code is shown in the legend vertical scale differs for M_y and A_z . The yellow-olive envelope shows the 3 realizations of the WACCM-CCMI simulation. The color codes for the four CTM simulations follow the conventions of S-SRIP (Fujiwara et al., 2017). BRAM2 is depicted with a black line and symbols, as usually done for observations, because it is constrained by both dynamical and chemical observations. Since the N_2O mixing ratio in BRAM2 has been evaluated with a 15% uncertainty (1-sigma standard deviation) at 15 hPa (Errera et al., 2019), this is highlighted by a dark grey region in top rows. The light grey shading around the BRAM2 cycles represents the uncertainty arising from the residual term in the TEM budget, i.e. it is entirely interpreted first as an uncertainty on A_z and then as an uncertainty on M_y in order to remain cautious.

Figure 6. Same as previous figure but showing the NH.

Monthly-standard-deviation Annual cycles over 2005-2014 at 15 hPa. First row: N_2O volume mixing ratio [ppbv], second row: horizontal mixing-term M_y [ppbv day⁻¹]; third row: vertical-residual-advection-term A_z [ppbv day⁻¹]. From-left-to-right Left column: southern mid-latitudes Antarctic region (40°S-60°S), southern-tropics-, Right column: Arctic region (60°N-80°N), northern-tropics (0-20°N), northern mid-latitudes (40-60°N). The color-code is shown in the legend vertical scale differs for M_y and A_z . The yellow-olive envelope shows the 3 realizations of the WACCM-CCMI simulation. The color codes for the four CTM simulations follow the conventions of S-SRIP (Fujiwara et al., 2017). BRAM2 is depicted with a black line and symbols, as usually done for observations, because it is constrained by both dynamical and chemical observations. Since the N_2O mixing ratio in BRAM2 has been evaluated with a 15% uncertainty (1-sigma standard deviation) at 15 hPa (Errera et al., 2019), this is highlighted by a dark grey region in top rows. The light grey shading around the BRAM2 cycles represents the uncertainty arising from the residual term in the TEM budget, i.e. it is entirely interpreted first as an uncertainty on A_z and then as an uncertainty on M_y in order to remain cautious.

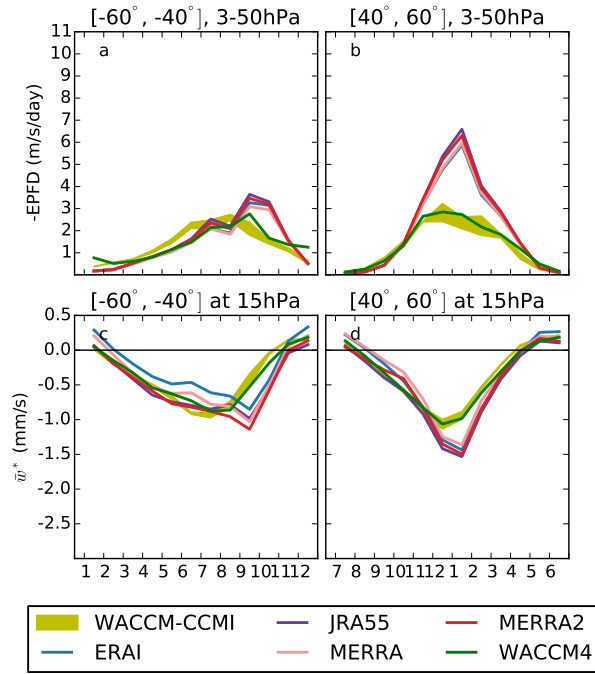


Figure 7. Same as previous figure As for Fig. 5 but for the polar regions (left middle latitudes. Left column: antarctic region, 80 southern mid-latitudes (40° - 60° S; right). Right column: arectic region, 60 northern mid-latitudes (40° - 80° - 60° N).

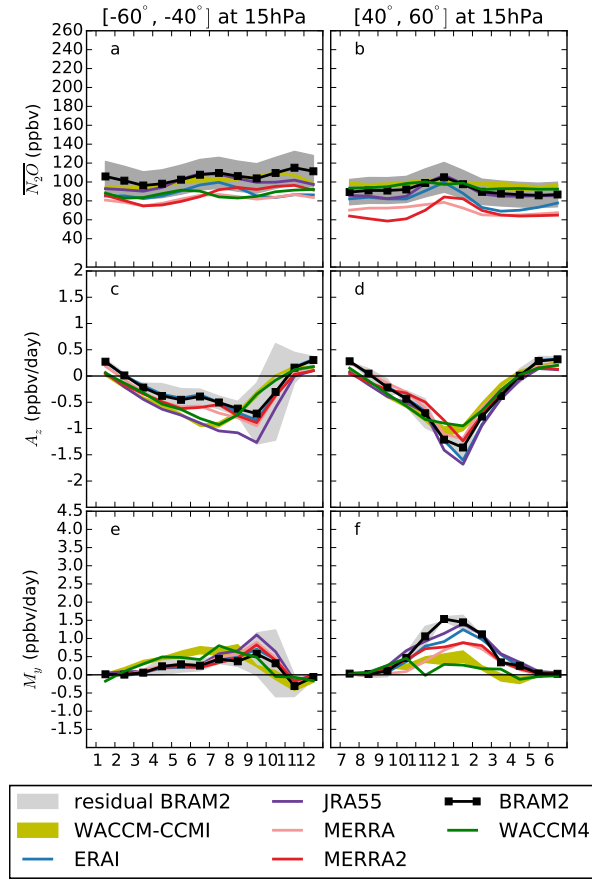


Figure 8. Monthly mean annual cycle of \bar{w}^* at 15 hPa in A_s for Fig. 6 but for the SH middle latitudes. From left to right Left column: southern mid-latitudes (40°-60° S). Right column: northern mid-latitudes (40°-60° N).

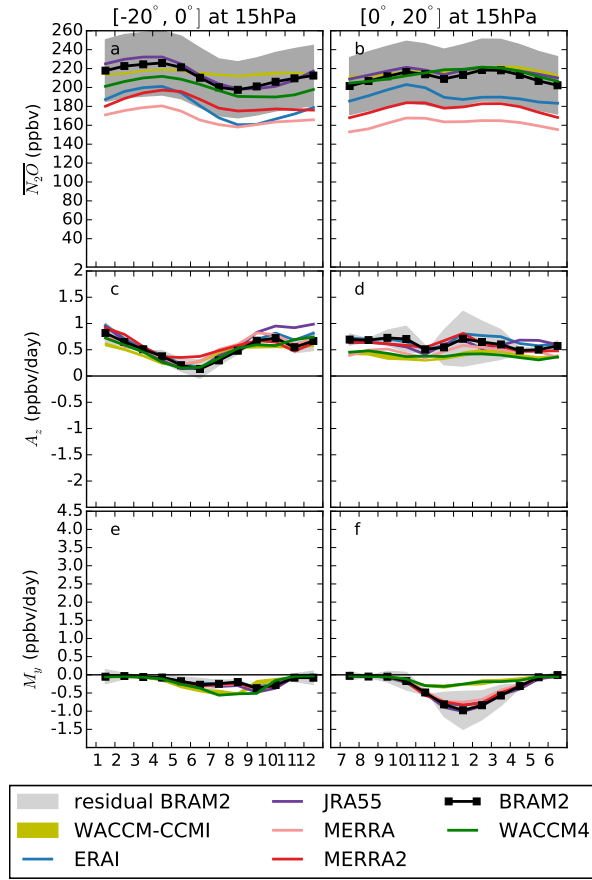


Figure 9. As for Fig. 6 but for the Tropics. Left column: southern tropics (0°-20° S). Right column: northern tropics (0°-20° N). northern mid-latitudes (40-60° N). The color code is shown in the legend. The yellow envelope shows the 3 realizations of the WACCM-CCMI simulation.

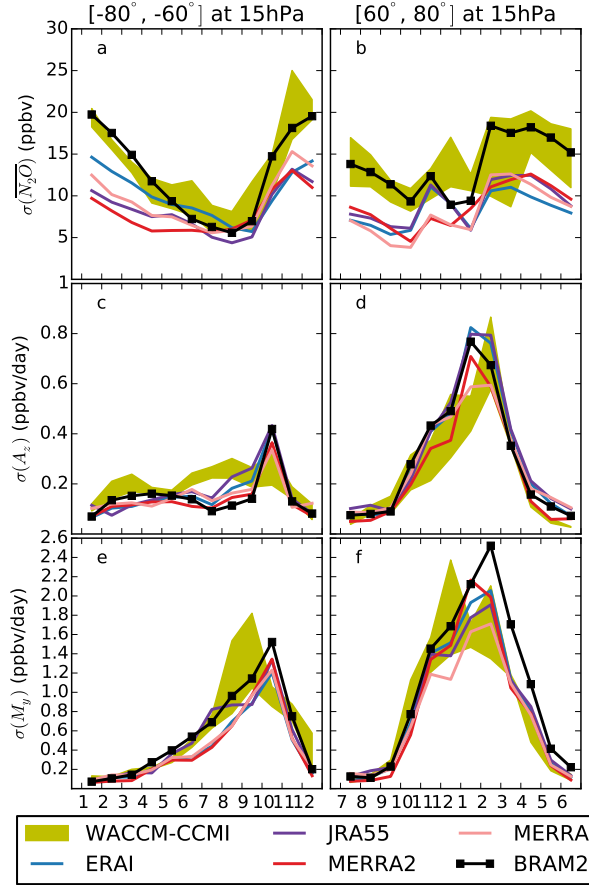


Figure 10. Monthly standard deviation over 2005-2014 at 15 hPa. First row: N₂O volume mixing ratio [ppb], second row: horizontal mixing term [ppbv day⁻¹]; third row: vertical residual advection term [ppbv day⁻¹]. Left column: Antarctic region (60°-80° S), right column: Arctic region (60°-80° N). The color code is shown in the legend. The yellow envelope shows the 3 realizations of the WACCM-CCMI simulation.

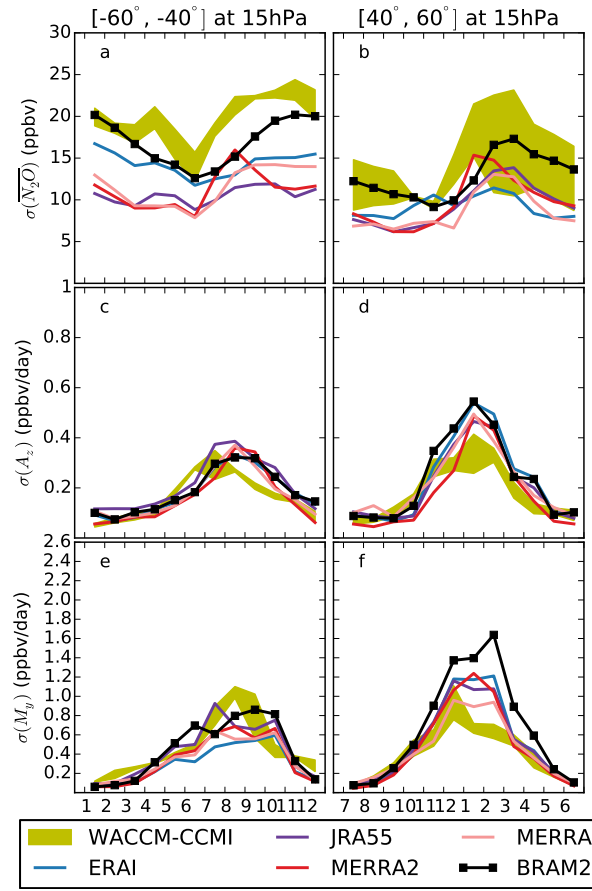


Figure 11. As Fig. 10 but the middle latitudes. Left column: southern mid-latitudes (40°-60° S). Right column: northern mid-latitudes (40°-60° N).

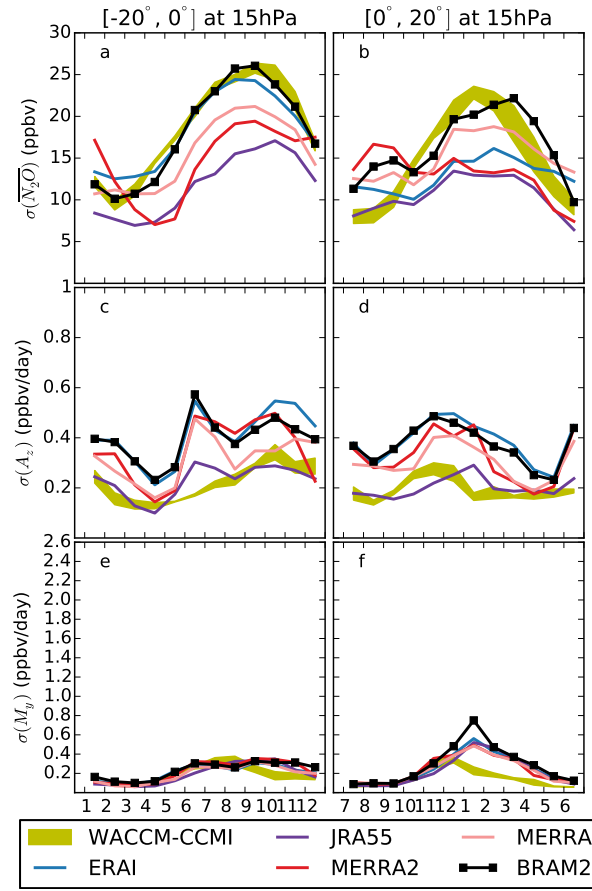


Figure 12. As Fig. 10 but the tropics. Left column: southern tropics (0°-20° S). Right column: northern tropics (0°-20° N).

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