Atmos. Chem. Phys. Discuss., 5, S5588–S5593, 2005
www.atmos-chem-phys.org/acpd/5/S5588/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

Interactive comment on "On the ability of chemical transport models to simulate the vertical structure of the N_2O, NO_2 and HNO_3 species in the mid-latitude stratosphere" *by* G. Berthet et al.

G. Berthet et al.

Received and published: 23 February 2006

Reviewer #2 comments were very helpful to clarify the paper. We address his suggestions in details below:

1) The reviewer suggests that we include satellite data to expand our analysis to a global scale. We think of course such a work would be very interesting and useful but we do not integrate satellite measurements in the presented study for several reasons: (i) Our aim is to make the scientific community be aware that using 6-hourly meteorological data appears to be responsible for a substantial part of the discrepancies when comparing CTMs and observations. This paper is a first step in the investiga-

tions that should also concern other families of stratospheric species. (ii) At this stage, our aim is not to analyse space-borne observations which is a different work implying different time and space scales. The originality of our paper is to present and analyse in situ simultaneous profiles of the N2O, NO2 and HNO3 nitrogen species which, to our knowledge, are available for the first time between 12 and 33 km thanks to the SPIRALE instrument described in the Moreau et al. (2005) publication. This is the first published modelling study of in situ observations of the reactive nitrogen species over such a wide vertical range. (iii) Unfortunately measurements from the instruments onboard ENVISAT and ODIN (and of other space-borne instruments) were not satisfyingly close in time and space to the SPIRALE observations for direct comparison. We now give this information in the text following reviewer #1's remark. Note that some modelling studies implying assimilation work and satellite and observations performed several days or weeks before SPIRALE measurements are currently under progress in our laboratories as part of ENVISAT validation papers. This assimilation work using a lagrangian model will give indirect comparisons of the different measurements and will allow us to test our understanding of the stratospheric chemistry by involving several sources of observations. For this last point a paper will also include GOMOS/ENVISAT, MIPAS/ENVISAT and ODIN observations compared to CTM calculations driven by 3-hourly and 1-hourly winds obtained by running directly the ECMWF model (note that 2 months of 1-hourly winds are already available).

2) Reviewer #2 points out that from the work of Stowasser et al. (2003) the X/NOy ratios should be independent of the absolute concentration of NOy and then of dynamical effects. Actually our conclusions are not totally comparable with the results of Stowasser et al. (2003). Indeed we can only show the NO2/HNO3 ratio since the SPIRALE instrument cannot observe all of the NOy species (note that NO can be usually measured by this instrument but this was unfortunately not possible for this flight) unlike MIPAS-B. The point is that this ratio gives only an estimation or a kind of approach of the X/NOy ratio but is not independent on the absolute concentration of NOy (and hence on the dynamical effects as rightly pointed out by the reviewer) since HNO3 is not a real

ACPD
5, S5588–S5593, 2005

tracer and cannot be attributed to total NOy. It is then clear that it would be more relevant to compare and analyse the X/NOy ratio (such as in the work of Stowasser et al. (2003)) rather than the X/HNO3 to drive definitive conclusions on our ability to simulate the NOy species partitioning (and to answer more precisely to the reviewer's point). Note that some trajectory work at Laboratoire de Météorologie Dynamique in Paris has shown that the trajectories ending at some altitudes of the measurements are sparse and originate from various regions (Ignacio Pisso, personal communication) due to the perturbed situation which could make these trajectories be more sensitive to the choice of meteorological data used to drive them. Different temperatures and Solar Zenith Angles on each backward trajectory depending on the chosen ECMWF data would affect the quantities of NO2 and of the other NOy species interacting with each other. Note that this is in particular the case for the other NOx species, NO, with computed mixing ratios that increase using 3-hourly data and that are comparable to the NO2 quantities at the time of the measurements. Other NOy species that contribute non-negligibly to the NOy partitioning (e.g. about 2 ppbv of N2O5 and 1 ppbv of ClONO2 simulated at 30 km) are more conserved between the various simulations. We have added the following comment in Chapter 5.2 Pg.12384 (rather than in the general conclusions): "Note that we clearly see in Fig. 9c that the NO2/HNO3 ratio depends on the meteorological data used to drive the model. In the perturbed dynamical situation studied here (as shown in Fig. 2) the geographical positions of the air-mass trajectories computed in Reprobus could depend on the chosen ECMWF data. Different temperatures and solar zenith angles along the computed trajectories between the 6-hourly and 3-hourly simulations would then affect the calculated quantities of the NOy species and especially of NO2 and NO."

Note also that following reviewer #1's comment we have replaced the sentence "We conclude from Fig. 5 that the NOy partitioning seems to be correctly reproduced by the CTM" by "We conclude from Fig. 5 that the NO2/HNO3 partitioning seems to be correctly reproduced by the CTM" since measurements of other NOy species are not available.

3) An important point mentioned by reviewer concerns the time-location of the model calculations for consistent comparisons with the observations. This point was also addressed by reviewer #1 (comment n°5) and we give here the same response following the reviewers' remarks: The REPROBUS model has a horizontal resolution of 2x2° and a vertical resolution of about 1.5 km in the mid-stratosphere (the vertical levels are those of the ECMWF model). The simulated vertical profiles shown in the paper correspond to the 44°N-0°E grid point and are located as close as possible to the SPIRALE measurements; indeed the balloon remained close to the same grid point during the flight since its displacement was only from 43.7°N-0.18°W to 43.60°N-0.16°E. It is then not necessary to interpolate the CTM results to the location of the SPIRALE measurements. Also this nicely ensures a very weak geographical variability in the observed profiles. But since we did not give enough information about the SPIRALE flight configuration, we have added the information about the displacement of the balloon in the revised version of the manuscript: Chapter 2 Pg.12376:"The measurement position remained rather constant during the ascent with a displacement of the balloon from 43.7°N-0.18°W to 43.60°N-0.16°E." However one could consider that the REPROBUS grid point at 42°N0°E should also be taken into account for the comparison with the SPIRALE measurements. We think it is more robust to take also into account the model profile at this location. Then we have done new plots shown in the revised version of the manuscript that correspond to the average of the profiles simulated at 42°N-0°E and 44°N-0°E that remained the closest grid points during all the balloon ascent (12-33 km). This average improves the comparisons between REPROBUS and SPIRALE but does not change our conclusions. The percentage difference values have been updated in the manuscript. Note that we have assessed that other grid points appear to be farther from the measurements and in different solar zenith angle conditions so that they can be also considered. We have added in Chapter 4.1.2 Pg. 12379: "The SPIRALE measurements were located between the 42°N-0°E and 44°N-0°E model grid points and remain close to these positions during the balloon ascent (from 43.7°N-0.18°W to 43.60°N-0.16°E). The profiles simulated at these two positions have been

ACPD
5, S5588–S5593, 2005

Interactive Comment

EGU

averaged for comparisons with the observations." The diurnal variation of NO_2 has also been included in the modelled profiles shown in the new figures following Reviewer #2's comment though this variation is only of 3-6% above 25 km. This information has been added in the text: Chapter 4.2 Pg. 12380: "The diurnal variation of the species has been taken into account in the simulated profiles described hereafter. The solar zenith angle varies from about 64° to 74° between 07:30 and 08:30 UT at the studied locations resulting in a variation of 3-6% for NO_2 above 25 km."

4) The reviewer asks whether using 6-hourly ECMWF forecasts instead of 6-hourly ECMWF operational analyses would improve the model-measurement comparisons. We did not use such meteorological data in our specific study (partly to avoid overloading the figures) but we quickly performed a preliminary sensitivity test on ozone simulations for a different period and without comparisons with measurements. There is less vertical diffusion using this kind of winds than using 6-hourly operational analysis but the best agreement is obtained using the 3-hourly forecasts. Interestingly, Legras et al. (2005) use this 6-hourly ECMWF short-time forecasts and obtain a much better agreement between reconstructed trajectories and the in situ observations of N_2O by the ER-2 than using the 6-hourly operational analyses. A better agreement is also obtained using 3-hourly winds from operational analyses interleaved with forecast as described in our manuscript (ope-for simulation). Unfortunately they could not perform their tests using 3-hourly forecasts since these winds were not archived at ECMWF before 2002. This test has been all the same performed by Ignacio Pisso from LMD, Paris, using the same approach as Legras et al. (2005) and he has obtained improved comparisons using 3-hourly forecasts rather than 6-hourly forecasts (personal communication) leading us not to perform a test using these former winds (which avoided us to archive 6 months of additional meteorological data set). One could use the 6-hourly ECMWF forecasts to drive a CTM if they are easier to compute than 3-hourly winds. Note that as stated by Legras et al. (2005), it remains still unclear whether 3 hours is a sufficiently short interval to achieve satisfying agreement with the observations. We and the LMD plan to test 1-hourly winds in addition to 6-hourly and 3-hourly winds us-

ing comparisons with balloon and satellite data as described above. Following reviewer #2's question it would be interesting to include simulations with 6-hourly forecasts.

Specific comments:

- Abstract: the correction has been done.

- Section 5.2, Pg.12383, l.8: the "new N2O profile" refers to both 3-hourly profiles of N2O. We have corrected the sentence in "Below 20 km the 3-hourly profiles of N2O are characterized..."

- Section 5.2, Pg.12383, l.22: the reviewer is right, we should have mentioned that the Michelsen et al. correlation may be only partly responsible for the discrepancy when the modelled NOy and the NOy* profiles are compared. It could also be due to a limitation of the CTM that does not produce enough NOy whatever the used meteorological data are. We have then modified the sentence by "This could be partly considered as a limitation..."

- Section 6, Pg.12385, l.26: We only mean that if we average the NO2 mixing ratios of the two 1-D simulations above 29 km (figure 10), midlatitude and midlat+trop simulation, we obtain values perfectly in agreement with the measurements. We have suppressed this sentence which is useless.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 12373, 2005.