

The authors thank the referees for their very constructive comments, which certainly will help to improve the manuscript.

In the following we give a point-to-point reply (blue) to the referee comments (black).

Responses to the comments of referee#2

General comments:

1. It uses NO₂ measurements by balloon-borne DOAS to constrain uncertainties in the photolysis of N₂O₅. This is however not clear from the abstract where only MIPAS-B is mentioned explicitly.

→ We accordingly changed the text to: “The method relies on the observation of the diurnal variation of NO₂ by limb scanning DOAS measurements, supported by detailed photochemical modelling of NO_y (...) and a non-linear least square fitting of the model result to the NO₂ observations.”

2. A section on the comparison of simulated N₂O₅ with MIPAS-B (or ENVISAT) observations using the original and the modified photolysis rates is missing (the discussion in section 5 is too short).

→ A comparison of simulated N₂O₅ with MIPAS-B (or ENVISAT) observations using the original and the modified photolysis rates would require a dedicated study, which was not possible within this work.

We accordingly added the following text: “(e.g., in yet unpublished comparison studies of SLIM CAT simulations with EMVISAT-MIPAS N₂O₅ measurements).”

3. Also a discussion of the effects of the different product channels of N₂O₅ photolysis is missing. Does the study always assume a yield of 1 for NO₃+NO₂?

→ We changed the text accordingly and added: “ Since all product channels finally lead to the production of NO_x through fast reactions (within several 10th of seconds), and the equilibrium between NO and NO₂ is rapidly established in the sunlit stratosphere, the N₂O₅ photolysis is the time limiting factor for the NO₂ production.” See also our reaction to comment 4 under ‘specific comments.’

Specific comments:

1. The abstract needs clarifications and has to be extended with respect to the instruments.

→ See our reaction to the general comment 1.

2. The uncertainty by a factor of 2 is in the cited table but from the spectra of absorption cross sections in the literature it appears to be exaggerated. Is it due to uncertainties in product channels?

→ In the table 4-2 of the JPL-2011 compilation, the combined uncertainty in the N₂O₅ cross sections and quantum yield is assessed to a factor 2, which we used in our study.

3. In the first paragraph of the introduction also the production of ozone via NO+HO₂ or RO₂ below about 30km should be mentioned (with reference). The text has to be improved there.

→ We accordingly change the first sentence of the introduction to: “The NO_x ozone loss cycle dominates catalytic ozone loss 35 in the mid-stratosphere (25 - 45 km), and the ozone production via coupling with the HO_x cycle below about 20 km (e.g., Brasseur and Solomon, 2005).”

4. The quantum yield is mentioned on page 4692 but details are given only 4 pages later. This should be rearranged or on page 4692 at least a sentence should be included. This holds also for the other product channels in R13.

→ We accordingly changed the text and included the reactions (16 to 18) into the various channels.

5. I'm surprised that the simple correction using the ozone ratio in Eqn. 2 is valid without any additional factor at all altitudes. From the chemistry discussed earlier it should be different above and below 30km. Also the mean should be independent of time (index j).

→ The re-normalisation correction corrects for pointing issues in the NO₂ retrieval and it needs the assumption of constant ozone for the observing period. The mean is indeed independent of time, so we deleted index j for the mean.

6. The Schumann-Runge bands are between about 180 and 200 nm, a region which is not addressed. Please correct line 10 on page 4696. The contribution of that region to N₂O₅ photolysis in the upper stratosphere is non negligible because of large cross sections.

→ We accordingly corrected the text to: "Figure 3 displays the actinic spectrum relevant for the photolysis of N₂O₅ at stratospheric conditions. It shows that two distinct wavelength regions are important for the photolysis of N₂O₅, the region w1 (200 - 258 nm) and the region w2 (260 - 420 nm). While N₂O₅ (;T) decreases over the whole wavelength region, the actinic flux modulates the actinic spectrum, mainly due to the absorption by O₂ and O₃. The minimum between the two maxima is caused by the combined effects of the O₃ absorption in the Hartley band, and the O₂ absorption in the Herzberg band. Thus, overhead O₂ and O₃ are crucial for the photolysis of N₂O₅ in the UV-C."

7. What is the a priori on page 4699, line 26? Please expand slightly. On page 4702, line 12, appears to be a contradiction to the abstract (typo?).

→ As a priori, s₁ and s₂ is set to unity, in agreement with the a prior information taken from the JPL-2011 compilations.

The sentence on page 4702, line 12 addresses the NO₂ absorption cross section, which is used in the photochemical modelling. We accordingly added to the text on page 4699 : "Consequently the a priori for s₁ and s₂ is assumed to (1,1) with a covariance are (2,2), in agreement with the values in Table 4-2 in JPL-2011"

8. Scattering from below, for example by clouds, is relevant for UV-A and visible. The sentences on top of page 4703 should be improved concerning this. The solar cycle effects are small against other uncertainties if only radiation with wavelength larger than 200nm is considered. Some sentences are misleading here.

→ We partly concur, but our limb intensity retrieval performed prior to the NO₂ profile retrieval (Kritten et al., 2010) did not show an radiation enhancement for wavelength larger than 320 nm. For quality how well limb radiances (and actinic fluxes) can be modelled see Figures 5 and 6 in Deutschmann et al., (2011) and Figure 2 in Kreytcy et al., (2013) (both cited in the text). Additionally we added to the text: "Scattering from below by clouds is not considered, because that could be excluded through a comparison of modelled and measured limb radiances. "

9. How is the total uncertainty in Table 1 calculated? From the numbers it is not the root of the sum of the squares.

→ Combined uncertainties are calculated by Gaussian quadrature. Unfortunately, a small error was found in the calculation, which we accordingly corrected for in the text.

Technical comments:

1. Typos: page 4690, line 14; page 4704, line 13.

→ We accordingly corrected the text.

2. In the sentence on page 4695, line 16, something must be wrong or missing.

→ We accordingly corrected the text.

3. Give reference for Facsimile (page 4696)

→ We included a reference to the FACSIMILE software (MCPA Software Ltd., Oxfordshire, UK)

4. The notation for a scaling factor in Eqn. 3 is odd.

→ We accordingly changed the notation.

5. Give reference for EMAC (page 4705).

→ We included a reference to atmospheric chemistry general circulation model EMAC (ECHAM5/MESSy2.4) (e.g. Jöckel, P., Sander, R., Kerkweg, A., Tost, H., and Lelieveld, J.: Technical Note: The Modular Earth Submodel System (MESSy) – a new approach towards Earth System Modeling, Atmos. Chem. Phys., 5, 433–444, doi:10.5194/acp-5-433-2005, 2005)

6. Use 'scaled' instead of 'updated' on page 4706, line 8.

→ We accordingly changed the text.

7. Use steps corresponding to the boxes in the color bars of Figs. 4 and 6.

→ The steps in the color bars of figure 4 and 6 are corresponding to the boxes in the figures with high resolution.

Responses to the comments of referee #1

1. The developed method is applied to only one balloon flight (Teresina, Brazil, 30 June 2005). However, since the launch of ENVISAT in 2002, several balloon flights have been conducted by the Heidelberg Group in the tropics and at mid- and high-latitudes. So, why the authors have not applied their method to these flights using SCIAMACHY limb (O₃) and MIPAS (NO_y) vertical profiles to constrain their photochemical model in the case of balloon measurements of these species were not available? This would certainly improve the statistics of the study. Moreover these profiles could also be used to validate the photochemical model output obtained using JPL-recommended and scaled N₂O₅ XS, which is actually missing in the manuscript.

→ The uniqueness of our approach comes with the selection of a photochemical regime where the NO_y photochemistry is largely dominated by the night-time formation of N₂O₅, and its photolysis into NO_x at daytime (see Figure 1 of the manuscript). In fact such a photochemical regime can only be found in the daytime tropical middle stratosphere, where we conducted only one successful mini-DOAS flight of reasonably long duration from where N₂O₅ measurements from MIPAS are available.

2. As Referee #2, I would recommend to discuss into more details the impact of the different pathways of the N₂O₅ photolysis on your results

→ See our response to comment 3 (general comments) of referee #2.

Typos:

Page 4705, line 5: 2 and 5 of N₂O₅ should be in subscript.

→ We accordingly changed the text.