Atmos. Chem. Phys. Discuss., 4, S3694–S3697, 2004
www.atmos-chem-phys.org/acpd/4/S3694/
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

Interactive comment on "4-D comparison method to study the NO_y partitioning in summer polar stratosphere – Influence of aerosol burden" *by* G. Dufour et al.

G. Dufour et al.

Received and published: 22 February 2005

Response to M. Van Roozendael (Referee)

The authors thank the referee for his interest to the article, his careful reading and his improvement suggestions. The authors apologize for some mistakes and confusing sentences that were left in the manuscript The comments made are addressed below.

General comments:

- Part (1): Concerning the following comments "E.g. it is mentioned in the first paragraph of section 3 that the 3D simulations were initialized on 15 Oct. 2000 and driven

by ECMWF analysis until June 2001, while the comparison is made on 21-22 Augsut. Is the dynamics of June also for August?" - Response: The authors apologize for this confusing mistake. The 3D simulations are of course driven by ECMWF analysis until the 21 and 22 August 2001. This has been modified.

Response concerning the remark on equation (2): The high latitudes in summer are particularly well adapted to study the gas-phase reactions involved in the NOy partitioning because of the few heterogeneous reactions and the low transport rate. On the simulation time (maximum 4 hours), the transport can be neglected and that's why the authors have used a box model for their study. Consequently, the equation (2) was not used to correct an eventual transport effect between the initialization time and the actual measurement time. It was just used to temporally "translate" the measured profiles to the initialization time (no measurements are available for the location sounded during the sunset and the sunrise at the initialization time). The Reprobus CTM can actually be used to scale the LPMA profiles as we have shown in the first simulation that the model (1D and then 3D) reproduces well the relative values, the problem comes just from the initialization.

On another hand, the authors showed with Figure 2 that the spatial variability of species like ozone for example can be relevant (around 1 ppmv) in the area sounded by LPMA and DOAS. To avoid errors during model/measurement comparison, this possible variability should be accounted for. The model outputs have then to be compared to the measurements at the actual measurement locations (different for each tangent point) and at the actual measurement time (4D comparison method). For this, the simulation outputs are interpolated at the measurement locations and the chemical time step of the box model is chosen small enough (15s) to match the measurement time.

To clarify these points the authors have added some precisions in the paper.

- Part (2): The authors have added a figure comparing the aerosol surface area profile deduced from the Reprobus CTM seasonal calculations and the same profile deduced

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from the balloon-borne aerosol concentration measurements performed by Deshler in 2002. Although the accuracy of surface area derived from measurements is quite low (40%), the difference between the modeled surface area and the measured ones is larger than the errors below 20 km. The surface area profile deduced from measurements performed in 2002 (used for this study) has been compared with the same profiles derived from 2001 and 2003 measurements. The 2001 profile was measured down to around 16 km. The agreement between the 2001 and 2002 profiles is very good down to 18 km and stays within the error bars between 18 and 16 km. The smallest altitude of the 2003 profile is around 15 km. A good agreement is observed for all the altitudes except the smallest one for which the 2003 value is unrealistically large. As the 2002 profile reaches 10 km (the lowest tangent point probed by LPMA is around 13 km) and doesn't show any unrealistic values, the authors have used it for this study.

- Part (3): Concerning the remark in the referee specific comments about the missing discussion on the possible reasons of the NOy underestimation by the model, this modeling problem is a general problem, not specific to Reprobus, but for now, any references aren't focused on this subject. The reasons of this misbehavior are not clearly identified and understood (the transport description is likely responsible of the NOy underestimation). This problem is under investigation and asked to be treated deeply in a separate paper. The authors prefer then not to include a discussion about this problem, which would be incomplete.

Specific comments:

- Page 8173, line25: replace "within" by "in" - Response: this has been replaced

- Page 8174, line 2: correct "Using this updated rate..." by "Using these updated rate..." - Response: this has been corrected

- Page 8174, line8: the reference to Sander et al., 2003 is not given at the end of the paper (only one to Sander et al., 2002) - Response: This has been checked and corrected.

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- Page 8174, same line: as written, the sentence beginning with “A robust” is not correct. I’d suggest ... simulated profiles...” - Response: This has been corrected.

- Page 8174, line 10: remove “Here” in the sentence beginning after “sunset” - Response: this has been changed.

- Page 8174, line 23: remove “The ozone profile was also used in this study” - Response: This has been removed.

- Page 8174, line 26: change “the consistency of ...” by “the consistency of the retrieved O3 and NO2 vertical profiles” - Response: this has been changed.

- Page 8176, line 15: change “... between 89° and 91° solar zenith angle” by “... between 89° and 91° of solar zenith angle” - Response: this has been changed.

-Page 8177, line 17: replace “22 August 2002” by “22 August 2001” - Response: this has been replaced.

- Page 8177, line 21: remove the word “retrieved” after the list of molecules - Response: this has been removed.

- Page 8177, line 23: I think that sentence beginning ... in Figure 4. - Response: Figure 3 presents the results obtained for individual profiles of NO, NO2 and HNO3 while Figure 4 presents the results obtained for NOx/NOy ratio. The authors prefer not moving this sentence as it concerns an “individual” molecule.

- Page 8180, line 1: replace “... this discrepancy can be explained best by ...” by “... this discrepancy can be explained to a large extent by ...” - Response: this has been replaced.

- Page 8180, line 8: correct “assigns” by “assign” - Response: “the instruments assigns” has been changed by “the instrument assigns”.

Interactive comment on Atmos. Chem. Phys. Discuss., 4, 8171, 2004.