PRELIMINARY VALIDATION OF SCIAMACHY NADIR OCLO SCDS

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ABSTRACT

Measurements of OClO total columns can serve as an indicator of stratospheric chlorine activation. It has been shown in several studies that OClO observations from GOME are well suited to characterise the spatial and temporal evolution of the chlorine activation in both hemispheres. Here we present OClO observations from SCIAMACHY in nadir mode. Since to date no operational OClO products are available we investigated scientific products processed by the Universities of Heidelberg and Bremen. We compared the SCIAMACHY OClO SCDs to simultaneous observations from GOME and also to OClO observations made by the AMAYX-DOAS instrument on the DLR-Falcon. In both cases a good agreement was found. However, compared to GOME data the SCIAMACHY OClO SCDs still show a relatively large offset and scatter.

1. INTRODUCTION

Solomon et al. [1] first proposed that activation of stratospheric inorganic chlorine, namely the conversion of the ozone-inert reservoir species HCl and $ClONO_2$ into ozone-destroying chlorine oxides (ClO, Cl_2O_2), is responsible for the dramatic ozone destruction observed since the beginning of the 1980s over Antarctica during late winter and spring [2]. In the cold stratosphere of the polar night inorganic reservoir species are converted into activated chlorine species by heterogeneous reactions, which occur on polar stratospheric clouds (PSCs) formed at the low stratospheric temperatures prevailing during polar night [1,3].
One commonly used remote sensing technique for the monitoring of the stratospheric chlorine activation is the spectroscopic measurement of OClO [4,5,6,7,8,9,10]. The most important stratospheric source of OClO is the reaction of BrO and ClO. In the sunlit stratosphere inorganic bromine primarily resides in BrO. BrO concentrations vary only slightly, and the availability of ClO often limits the formation of OClO. It has been shown by [11] that OClO can (at SZA < 92°) serve as an indicator for the amount of stratospheric ClO, i.e. the chlorine activation [see also 4].
In this study we present OClO data derived from spectra measured by GOME and SCIAMACHY. These data allow to monitor the stratospheric chlorine activation on a daily basis.

2. DATA ANALYSIS AND RESULTS

The SCIAMACHY DOAS OClO analysis was performed in the UV spectral range (for details see [7,8]. In contrast to the GOME analysis for the SCIAMACHY in addition to the trace gas cross sections further spectra were included in the DOAS fitting process (see Table 1).

Table 1 Reference spectra used in the DOAS analysis

Spectrum	Remark
OClO	213 K, Krominga et al., 2003 [12]
O_3	223 K, Bogumil et al., 2003 [13]
O_4	298 K Greenblatt et al., 1990 [14]
NO_2	220 K, Vandaele et al., 1997 [15]
Ring	Calculated, Bussemer, 1993 [16]
Offset	$1/I_0$ included in Fit
Polarisation 1	Eta from key data
Polarisation 2	Zeta from key data
Fraunhofer 1	Atmospheric spectrum, SZA 70°
Fraunhofer 2	Direct solar spectrum, ESM
Fraunhofer 3	Direct solar spectrum, ASM

These spectra characterise the polsarisation sensitivity of SCIAMACHY and were taken from the key data. Moreover, also different Fraunhofer reference spectra were used: Besides atmospheric measurements at low SZA (see [7]) also direct solar spectra via the ESM or ASM diffuser were selected. In Figure 1 and 2 the influence of the different choices is presented. Without the use of the polarisation corrections spectra the derived OClO SCDs show two major shortcomings: First the OClO SCDs outside the polar vortex and at small SZA can show values significantly different from zero. In addition, also a large scatter of the data is found. Including the polarisation spectra in the fitting procedure reduces both problems and leads to much more convincing results. The Bremen SCIAMACHY OClO analysis includes additional spectra in the DOAS

Proceedings of the Second Workshop on the Atmospheric Chemistry Validation of ENVISAT (ACVE-2)
ESA-ESRIN, Frascati, Italy, 3-7 May 2004 (ESA SP-562, August 2004) ESC02TW

fitting process; they are partly derived empirically. Accordingly the OClO SCDs from this analysis show a further reduced scatter (Fig. 3). Nevertheless, still an offset is found, which leads to apparent negative values outside polar regions.

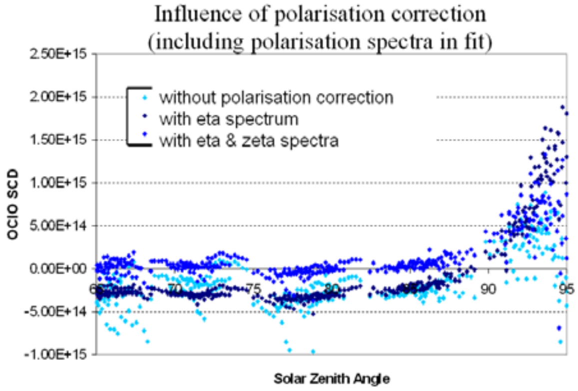

Fig. 1 OClO SCD derived from SCIAMACHY (scientific algorithm from the University of Heidelberg, SCIA-orbit 2510). Including polarization spectra from key-data in the DOAS fit improves significantly the consistency of the results.

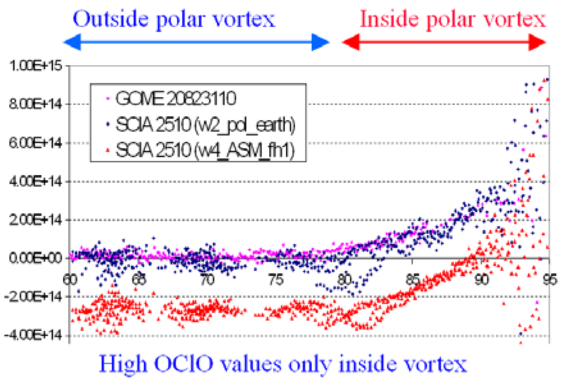

Fig. 2 Influence of different Fraunhofer spectra on the OClO SCDs derived from SCIAMACHY observations (scientific algorithm from the University of Heidelberg, SCIA-orbit 2510). Results from simultaneous GOME observations are also included in the diagram.

In Fig. 4 maps of the OClO SCDs from GOME and SCIAMACHY are shown. In both data sets a good agreement is found: High values towards the south pole as can be expected from meteorology and viewing geometry. It is important to note here, that there is a small temporal difference between both overpasses, which can explain at least part of the remaining differences.

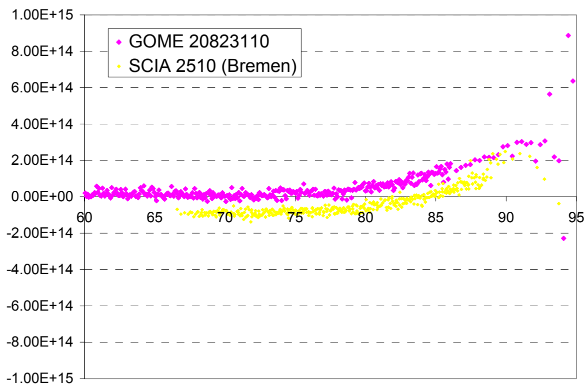

Fig. 3 SCIAMACHY OClO (orbit 2510) results from the scientific algorithm of the University of Bremen.

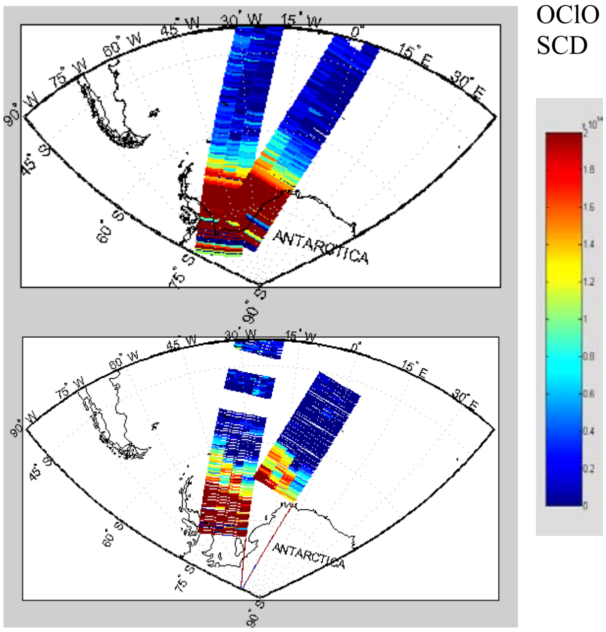

Fig. 4 Comparison of maps of OClO SCDs derived from GOME and SCIAMACHY (scientific algorithms from the University of Heidelberg).

We also compared the SCIAMACHY OClO SCDs to values derived from the AMAX-DOAS instrument flown on the DLR-Falcon [17,18]. On January 26[th], the DLR-Falcon flew from Kiruna (67.82°N, 20.34°E) to the north east, turned at 80°N, 30°E and came back on almost the same route. During the flight OClO slant columns were measured by the AMAXDOAS instrument in the 180°, 92°, and 88° viewing directions. The OClO slant columns for 92° are shown in Fig. 5.

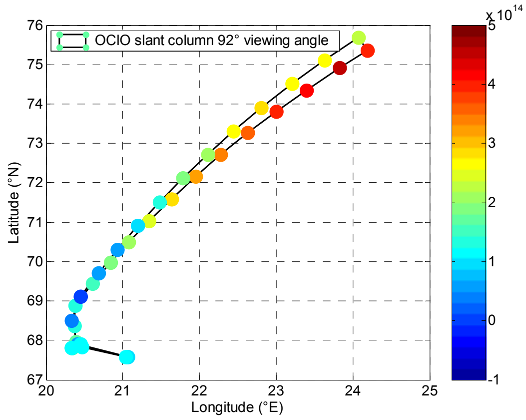

Fig. 5. AMAX-DOAS OClO slant columns measured in January 26 in the 92° viewing direction as a function of latitude and longitude.

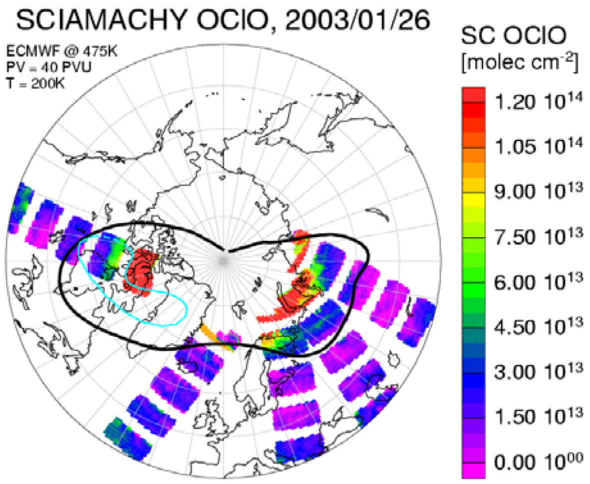

Fig. 6: OClO SCDS from SCIAMACHY (scientific algorithm, Uni-Bremen). The black line is the vortex boundary at 40 PVU, the cyan line is the 200K temperature isoline, both at 475K potential temperature. Only 5 orbits of SCIAMACHY data are available up to date.

In the northeast OClO concentrations are high which partly is a result of large SZA but also of larger local OClO concentrations. This is supported by the GOME and SCIAMACHY measurements and also the ECMWF temperature and PV analysis data. The vortex position and SCIAMACHY OClO SCDs are plotted in Fig. 6. From the satellite maps it is clear, that OClO was present within the vortex, which was close to Kiruna on the day of measurements, leading to a large gradient along the flight track. During the flight, OClO SCDs ranged between 0.7-3.5*10^{14} Molec/cm^2 as a function of latitude and SZA, which again is in excellent agreement with the variation observed in the SCIAMACHY data north of Kiruna. While this is a very encouraging result, a point by point validation of the satellite data will only be possible if the full radiative transfer and horizontal change in OClO is taken into account.

3. CONCLUSIONS

We presented first OClO SCDs derived from SCIAMACHY using two scientific algorithms (developed at the Universities of Heidelberg and Bremen). Reasonable OClO results can so far only be achieved if additional spectra are included in the fitting procedure. The main improvement is found if polarisation spectra from the SCIAMACHY key-data are used indicating that still problems in the absolute calibration of SCIAMACHY spectra exist. The comparison of SCIAMACHY OClO SCDs with GOME and aircraft observations shows very good overall agreement. Remaining limitations of the derived SCIAMACHY OClO SCDs are a large scatter of the SCIAMACHY data and an offset which can lead to (unphysical) values ≠ 0 outside polar regions. It is interesting to note that already a simple normalization procedure might yield a pragmatic solution for the offset problem: the OClO results of a whole orbit must be shifted in order to lead to values close to zero for low latitudes.

The derived SCIAMACHY OClO SCDs are already a very promising data set. They allow to continue the OClO time series from GOME and thus to monitor the evolution of stratospheric chlorine activation during the period of probable Cl_y decrease in the stratosphere. It is recommended that a specific OClO analysis following the scientific retrievals should soon be implemented as an operational data product.

4. ACKNOWLEGEMENTS

The authors wish to thank the DLR Falcon team and Flugbetrieb for their great support in both setting the AMAXDOAS instrument up in the Falcon as well as performing the large number of flights under difficult conditions. Parts of this work have been funded by the University of Bremen and the Bundesrepublik Deutschland through projects 50EE0023 and 50EE0024. Lv1 and lv0 data from GOME and SCIAMACHY have been provided by ESA and DLR/DFD Oberpfaffenhofen. ECMWF meteorological analysis data have kindly been provided by ECMWF.

5. REFERENCES

1. Solomon, S., R.R. Garcia, F.S. Rowland, and D.J. Wuebbles, On the depletion of Antarctic ozone, *Nature, 321,* 755-758, 1986.

2. Farman, J.C., B.G. Gardiner, and J.D. Shanklin, Large losses of total ozone in Antarctic reveal seasonal ClO_x/NO_x interaction, *Nature, 315,* 207-210, 1985.
3. Toon, O.B., P. Hamill, R.P. Turco, and J. Pinto, Condensation of HNO_3 and HCl in the winter polar stratosphere, *Geophys. Res. Lett., 13,* 1284-1287, 1986.
4. Solomon, S., A. L. Schmeltekopf, and R. W. Sanders, On the interpretation of zenith sky absorption measurements, *J. Geophys. Res., 92,* 8311-8319, 1987a.
5. Wittrock, F., A. Richter, and J. P. Burrows, Validation of GOME and OClO observations in the north hemisphere, in *Proceedings of the European Symposium on Atmospheric Measurements from Space (ESAMA 99), Jan. 18-22, ESTEC, Noordwijk, Netherlands*, WPP-161, pp. 735-738, Eur. Space Agency, Noordwijk, Netherlands, 1999.
6. Otten, C., F. Ferlemann, U. Platt, T. Wagner, and K. Pfeilsticker, Ground-based DOAS UV/visible measurements at Kiruna (Sweden) during the SESAME winters 1993/94 and 1994/95, *J. Atmos. Chem., 30,* 141-162*,* 1998.
7. Wagner, T., C. Leue, K. Pfeilsticker, and U. Platt, Monitoring of the stratospheric chlorine activation by Global Ozone Monitoring Experiment (GOME) OClO measurements in the austral and boreal winters 1995 through 1999, J. Geophys. Res., 106, 4971-4986, 2001.
8. Wagner, T., F. Wittrock, A.Richter, M. Wenig, J. P. Burrows, and U. Platt, Continuous monitoring of the high and persistent chlorine activation during the Arctic winter 1999/2000 by the GOME instrument on ERS-2, *J. Geophys. Res.*, 107(D20), 8267, 2002.
9. Kühl S., Wilms-Grabe W., Beirle S., Frankenberg C., Grzegorski M., Hollwedel J., Khokhar F., Kraus S., Platt U., Shangavi S., von Friedeburg C. and Wagner T., Stratospheric Chlorine Activation in the Arctic winters 1995/96 to 2001/02 derived from GOME OClO Measurements, Adv. Space Res., in press, 2004.
10. Richter, A., F. Wittrock, M. Weber, S. Beirle, S. Kühl, U. Platt, T. Wagner, W. Wilms-Grabe, and J. P. Burrows, GOME observations of stratospheric trace gas distributions during the splitting vortex event in the Antarctic winter 2002 Part I: Measurements, *accepted for publication in J. Atmos. Sci.*, 2004.
11. Schiller, C., and A. Wahner, Comment on „Stratospheric OClO measurements as a poor quantitative indicator of chlorine activation“ by J. Sessler, M.P. Chipperfield, J.A. Pyle, and R. Tuomi, *Geophys. Res. Lett., 23,* 1053-1054*,* 1996.
12. Kromminga, H., J. Orphal, P. Spietz, S. Voigt and J.P. Burrows, "New measurements of OClO absorption cross sections in the 325-435 nm and their temperature dependence between 213-293 K, *J. Photochem. Photobiol. A.*: Chemistry, 157, 149-160, 2003.
13. Bogumil, K., J. Orphal, T. Homann, S. Voigt, P.Spietz, O.C. Fleischmann, A. Vogel, M. Hartmann, H. Bovensmann, J. Frerik and J.P. Burrows, Measurements of Molecular Absorption Spectra with the SCIAMACHY Pre-Flight Model: Instrument Characterization and Reference Data for Atmospheric Remote-Sensing in the 230-2380 nm Region, J. Photochem. Photobiol. A., 157, 167-184, 2003.
14. Greenblatt, G. D., J. J. Orlando, J. B. Burkholder, and A. R. Ravishankara, Absorption measurements of oxygen between 330 and 1140nm, *J. Geophys. Res.*, 95: 18577-18582, 1990.
15. Vandaele, A. C., C. Hermans, P. C. Simon, M. Carleer, R. Colin, S. Fally, M.-F. Mérienne, A. Jenouvrier, and B. Coquart, Measurements of the NO_2 Absorption Cross-section from 42000 cm-1 to 10000 cm-1 (238-1000 nm) at 220 K and 294 K, J. Quant. Spectrosc. Radiat. Transfer, 59, 171-184, 1997.
16. Bussemer, M., Der Ring-Effekt: Ursachen und Einfluß auf die Messung stratospärischer Spurenstoffe, *Diploma Thesis, University of Heidelberg,* 1993.
17. Wagner T., Bruns M., Burrows J.P., Fietkau S., Finocchi F., Heue K.-P., Hönninger G., Platt U., Pundt I., Richter A., Rollenbeck R., von Friedeburg C., Wittrock F., Xie P., The AMAX instrument and its application for SCIAMACHY validation, *Proceedings of the 15th ESA symposium on rocket and balloon program and related research,* Biarritz France, 28-31 May 2001, ESA SP-471, August 2001.
18. Wang, P. A. Richter, M. Bruns, J.P. Burrows, K.-P. Heue, I. Pundt, T. Wagner, U. Platt, AMAXDOAS measurements and first results for the EUPLEX campaign, *Proceedings of the 16th ESA symposium on rocket and balloon program and related research,* St Gallen France, 2 – 5 June 2003.