Initial intercomparison of ozone and nitrogen dioxide number density profiles retrieved by the ACE-FTS and GOMOS occultation experiments

D. Fussen, F. Vanhellemont, J. Dodion, and C. Bingen
Institut d'Aéronomie Spatiale de Belgique, Brussels, Belgium

K. A. Walker, C. D. Boone, S. D. McLeod, and P. F. Bernath
Department of Chemistry, University of Waterloo, Waterloo, ON, Canada

Received 18 January 2005; revised 30 March 2005; accepted 2 May 2005; published 22 June 2005.

[1] The ozone and nitrogen dioxide vertical number density profiles measured by the solar occultation Fourier transform spectrometer ACE-FTS and the UV-Vis stellar occultation instrument GOMOS are intercompared for 370 quasi-coincident observations. A good agreement is found for ozone, mostly better than 10% between 15 and 45 km. Also, there is no evidence for a systematic altitude registration error in the ACE-FTS profiles. A considerable ACE-FTS negative bias (50–100%) is found however for the nitrogen dioxide data that cannot be explained by the use of a local photochemical model. **Citation:** Fussen, D., F. Vanhellemont, J. Dodion, C. Bingen, K. A. Walker, C. D. Boone, S. D. McLeod, and P. F. Bernath (2005), Initial intercomparison of ozone and nitrogen dioxide number density profiles retrieved by the ACE-FTS and GOMOS occultation experiments, *Geophys. Res. Lett.*, *32*, L16S02, doi:10.1029/2005GL022468.

1. Introduction

[2] Limb occultation measurements from spaceborne instruments possess an unique advantage for the remote sounding of the Earth's atmosphere: they give access to the absolute value of the slant path optical thickness from the relative measurement of the atmospheric transmittance [*Kirchengast et al.*, 2004]. This property is also particularly important for long-term monitoring as the instrumental optics and the detector devices possibly deteriorate in the space environment. The advantage of self-calibration by an exo-atmospheric measurement has a price: the necessity of a directional source of light, compatible with the orbital ephemerides of the carrier satellite. The consequent reduction in the geographical sampling rate is also to be evaluated with respect to the usually better vertical resolution. The Sun, the Moon, stars or planets may be used with quite different experimental possibilities depending on the signal-to-noise ratio (SNR), the apparent vertical resolution, the source irradiance uniformity and the frequency of available occultations.

[3] Recently, two major space occultation instruments have been put into orbit. The GOMOS spectrometer onboard the European platform ENVISAT is functioning since March 2002. It is a UV-Vis-NIR spectrometer aimed at the observation of stellar occultations from an heliosynchronous circular orbit at an altitude of 800 km. The geophysical objectives associated with the UV-Vis part of the instrument are the O_3, NO_2, NO_3, OClO, BrO, air number density and aerosol extinction vertical profiles [*Kyrölä et al.*, 2004]. So far, GOMOS has observed several hundreds of thousands of occultations and its ozone product (version 6.0a) has been validated with respect to ground-based measurements and other satellite data [*Meijer et al.*, 2004].

[4] The ACE-FTS solar occultation instrument is described in this dedicated section of GRL [*Bernath et al.*, 2005]. Briefly, it is an infrared Fourier transform spectrometer (FTS) operating in the 2 to 13 micron domain that was launched on August 12, 2003 with the companion experiment MAESTRO. Amongst the large number of targeted trace gases absorbing in the infrared, the spectrometer also measures O_3, NO_2 and atmospheric extinction profiles. The satellite operates from a circular orbit at an altitude of 650 km, and the orbital plane inclination of 74 degrees allows for a global coverage with some predominance of the polar regions.

[5] This paper will present the results of an intercomparison exercise between the O_3 and NO_2 number density profiles retrieved by both instruments during the first part of 2004.

2. Intercomparison Protocol

[6] Although both experiments are using the limb occultation technique, they considerably differ in many aspects: most of the GOMOS information comes from the UV-Vis spectrum wheras ACE-FTS is using the infrared; the stars are faint light sources of variable magnitude and temperature that hardly compete with the very large constant SNR delivered by the Sun; the nominal vertical resolution of GOMOS is 1–1.7 km to compare to 3–4 km for ACE-FTS; the stellar occultations can be observed at a higher rate (20–40 per orbit) instead of a unique sunrise/sunset pair per orbit. Finally, the GOMOS transmittance spectra are known to contain some residual star scintillation for oblique occultations that necessitated a Tikhonov regularization in the vertical inversion algorithm. A target ozone profile resolution of 2 km up to 40 km and 3 km above 40 km was selected in agreement with the climatological ozone profile smoothness [*Sofieva et al.*, 2004]. The NO_2 resolution is about 4 km at all altitudes. Apart from the comparison of the absolute number density profiles, this study has addressed the validation of the ACE-FTS tangent altitude assignment as derived by the pressure retrieval.

0094-8276/05/2005GL022468

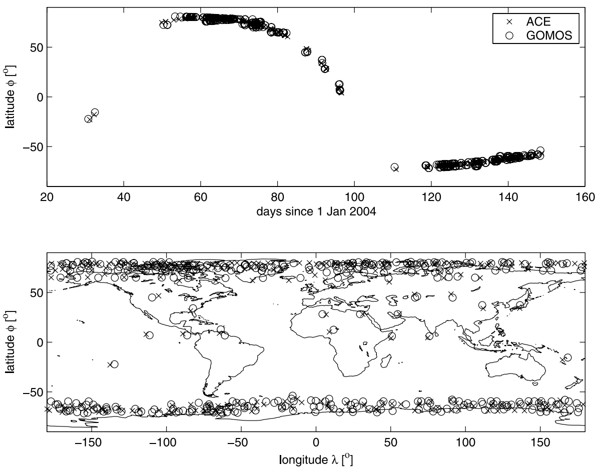

Figure 1. (top) Latitude/time plot of the ACE (crosses) and GOMOS (circles) common geolocation sites. (bottom) Corresponding geographical distribution.

[7] In preamble to any interpretation of the comparison between the retrieved profiles, it is worth stating that most of the measured extinction occurs near the tangent point along an effective optical path length L. If the extinction cross section equals σ, an exponentially decreasing number density profile (like air) of the form $n_a(z) = N_0 \exp(-z/H_a)$ leads to the following approximate value for the slant path optical thickness at the tangent altitude h:

$$\tau_a(h) \simeq \sigma n_a(h) L_a = \sigma n_a(h) \sqrt{2\pi H_a R} \quad (1)$$

where R stands for the Earth's radius. When the number density profile has a maximum around z_0 and may be locally approximated by a Gaussian shape $n_b(z) = N_0 \exp(-((z - z_0)/H_b)^2)$, a shifted maximum is also observed in the optical thickness at tangent altitude $h_* = z_0 - 0.541 H_b$ and

$$\tau_b(h_*) \simeq \sigma N_0 L_b = \sigma N_0 \sqrt{9.26\, H_b R} \quad (2)$$

[8] Using $H_a \simeq 7$ km for the air scale height and $H_b \simeq 16$ km for a typical ozone profile, one finds $L_a \simeq 530$ km and $L_b \simeq 750$ km. Clearly, a limb occultation experiment considers a spherically homogeneous atmosphere in the corresponding geographical domain and this assumption has always to be kept in mind in special circumstances (e.g. near the polar vortex). Hence, the possible coincidences between both instruments were constrained not to differ by more than 0.5 day and 500 km at maximum.

[9] In Figure 1, we have plotted the respective geolocations associated with the 370 available data sets. Notice the identical phenomenon for the Sun or for a particular star: once observed during a given orbit, both are still observable at the next one, at about the same latitude and for a longitude displaced westward by a value equivalent to the Earth's rotation during the orbital period. This is the reason for the "tracks" in the latitude/time subplot of Figure 1. In Figure 2, we have plotted four ozone profiles, quite representative of the whole data set although outliers exist in some GOMOS data probably contaminated by residual scintillation. The agreement is fair above 15 km. In particular, there is no clear evidence of a significant ($\leq$1 km) altitude bias in the ACE-FTS data. Also, the ACE-FTS ozone maximum seems to be slightly lower (by 5–10%) than the GOMOS value and this is probably caused by the lower vertical resolution of the solar occultation technique. One (upper left) of the measured profiles exhibits a sharp drop below 20 km. As this is observed by two independent instruments and techniques, we suspect that a thin cirrus or a strong convective cloud might have produced such a sharp increase in atmospheric extinction.

[10] In Figure 3, we have plotted the distribution of the relative differences $\Delta(\%)$ of the ozone profiles for the entire data set. In view of the presence of outlying GOMOS profiles (from 25% in dark limb to 75% in the difficult bright limb cases), we have preferred the use of more robust

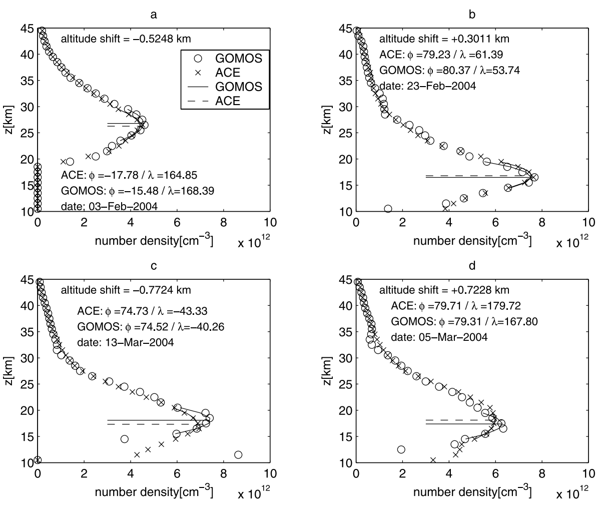

Figure 2. Four typical O_3 comparisons between ACE-FTS (crosses) and GOMOS (circles). The peak centers (short straight lines) have been computed by using a local quadratic fit around the ozone maximum. Notice the upper left case where both profiles are truncated below 20 km by a probable cirrus cloud.

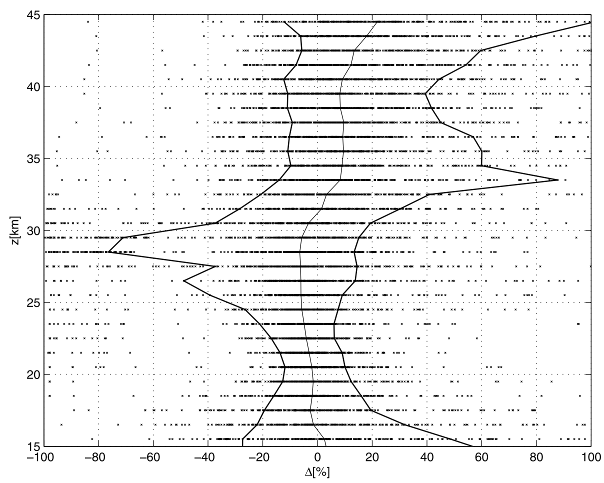

Figure 3. Relative differences (100*(ACE-GOMOS)/GOMOS) of all O_3 profiles. The thin line is the median value and the thick lines refer to the 0.16 and 0.84 percentiles of the distribution.

estimators like the median and the difference $D(\Delta)$ between the 0.16 and 0.84 percentiles. Although it would be preferable to avoid the use of bright limb occultations (we refer the reader to *Meijer et al.* [2004] for a general discussion of the GOMOS validation), the use of bright limb cases was justified for the sake of statistical significance. We conclude that there is a slightly negative [3–7%] ACE-FTS bias with respect to GOMOS in the [15–30 km] altitude range and a positive [10–15%] bias between 30 and 45 km.

[11] In Figure 4, we have plotted the evolution of $D(\Delta)$ for all altitudes between 15 and 45 km when the GOMOS-ACE time difference and the tangent point interdistance are increased. The very weak increase of the dispersion of the comparison geolocations legitimizes the selected time and interdistance ranges.

[12] The comparison of NO_2 profiles turned out to be less successful and the results presented here are only preliminary. In Figure 5, a clear ACE-FTS negative bias of 50–100% shows up between 15 and 45 km. However, it is quite difficult to conclude that the retrieved values are incorrect. Indeed, the official GOMOS "twilight" event classification corresponds to solar zenith angles (SZA) ranging from 95 to 120 degrees. Photochemical simulations [*Lambert et al.*, 1999] show that the day-night transition appears to be very sharp (a few SZA degrees) but also depends on latitude and season. The photochemical correction should be applied independently for all GOMOS-ACE cross-comparisons but this was out of the scope of this initial exercice. Instead, we have used the model developed by *Hendrick et al.* [2004] applied to the median GOMOS occultation (absolute value of latitude: 68 degrees, julian day = 95 and median solar zenith angle of 106 degrees). The correction factor is shown in Figure 5 but it is unable to explain the systematic difference below 30 km.

3. Conclusions

[13] We have presented the results of the intercomparison of about 370 occultations between the stellar GOMOS and solar ACE-FTS occultation instruments. Keeping in mind the finite spatio-temporal resolution associated with the limb occultation geometry, the preliminary analysis shows a good agreement between the retrieved ozone profiles, mostly better than 10% in the 15–45 km altitude range. In particular, no significant altitude shift has been observed by comparing the positions of the maxima of ozone number density. The comparison between the nitrogen dioxide profiles is less satisfactory but should require more statistics in order to restrict the observation geometry to identical twilight conditions.

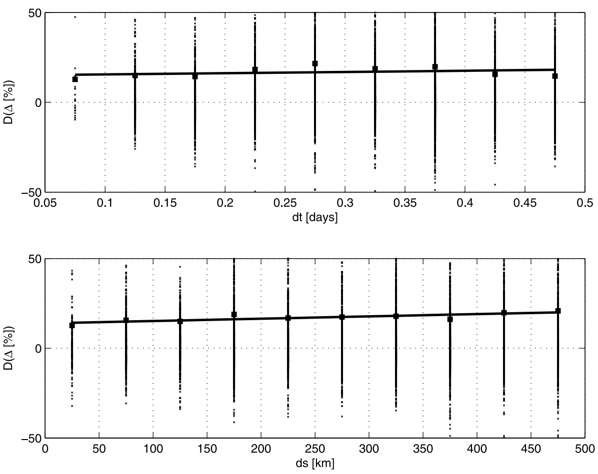

Figure 4. Evolution of the distribution width of the relative error distribution when the allowed (top) time difference and (bottom) interdistance (between the ACE/GOMOS observations is increased.

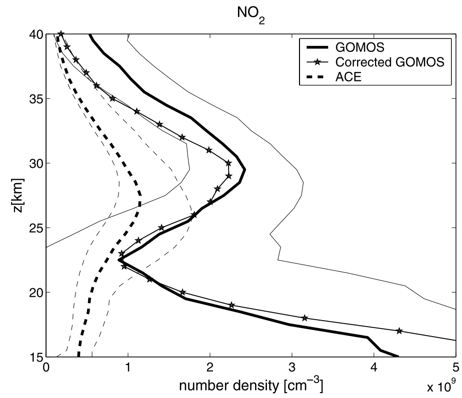

Figure 5. The median NO_2 profiles (with respective 0.16 and 0.84 percentile limits) measured by ACE-FTS (dashed lines) and GOMOS (full lines). The corrected GOMOS (full line with stars) represent the photochemical extrapolation of the median GOMOS profile to the ACE twilight conditions.

[14] **Acknowledgments.** The present study was funded by two PRODEX 7 contracts (SADE and PROACE) under the authority of the Belgian Space Science Office (BELSPO). The ACE mission is funded by the Canadian Space Agency and the Natural Sciences and Engineering Research Council of Canada (NSERC). Funding at Waterloo is also provided by the NSERC-Bomem-CSA-MSC Industrial Research Chair in Fourier Transform Spectroscopy. The authors thank F. Hendrick for the computation of the photochemical correction factor.

References

Bernath, P. F., et al. (2005), Atmospheric Chemistry Experiment (ACE): Mission overview, *Geophys. Res. Lett.*, *32*, L15S01, doi:10.1029/2005GL022386.

Hendrick, F., et al. (2004), Retrieval of nitrogen dioxide stratospheric profiles from ground-based zenith-sky UV-visible observations: Validation of the technique through correlative comparisons, *Atmos. Chem. Phys. Discuss.*, *4*, 2867–2904.

Kirchengast, G., U. Foelsche, and A. K. Steiner (2004), *Occultations for Probing Atmosphere and Climate*, Springer, New York.

Kyrölä, E., et al. (2004), GOMOS on Envisat: An overview, *Adv. Space Res.*, *33*, 1020–1028.

Lambert, J.-C., J. Granville, M. Roozendael, J.-F. Müller, J.-P. Pommereau, F. Goutail, and A. Sarkissian (1999), A pseudo-global correlative study of ERS-2 GOME NO_2 data with ground-, balloon-, and space-based observations, in *Proceedings of the European Symposium on Atmospheric Measurements from Space (ESAMS)*, *ESA WPP-161*, vol. 1, pp. 217–224, Eur. Space. Res. and Technol. Cent., Noordwijk, Netherlands.

Meijer, Y. J., et al. (2004), Pole-to-pole validation of Envisat GOMOS ozone profiles using data from ground-based and balloon sonde measurements, *J. Geophys. Res.*, *109*, D23305, doi:10.1029/2004JD004834.

Sofieva, V. F., J. Tamminen, H. Haario, E. Kyrölä, and M. Lehtinen (2004), Ozone profile smoothness as a priori information of limb measurements, *Ann. Geophys.*, *22*, 3411–3420.

P. F. Bernath, C. D. Boone, S. D. McLeod, and K. A. Walker, Department of Chemistry, University of Waterloo, 200 University Ave. W, Waterloo, ON, Canada N2L 3G1.

C. Bingen, J. Dodion, D. Fussen, and F. Vanhellemont, Institut d'Aéronomie Spatiale de Belgique, 3, avenue Circulaire, B-1180, Bruxelles, Belgium. (didier.fussen@oma.be)