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Atmospheric
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Direct comparisons of GOMOS and SAGE III NO_3 vertical profiles

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Abstract

In this paper we present first global comparisons between the two unique satellite-borne data sets of NO_3 vertical profiles retrieved from the GOMOS (Global Ozone Monitoring by the Occultation of Stars) stellar occultations and the SAGE III (Stratospheric Aerosols and Gas Experiment) lunar occultations. The comparison results indicate that between the altitudes 25 km and 45 km the median difference between these two data sets is within ±25 %. The study of zonal median profiles shows agreement between these data sets. The agreement is at its best in tropics and slightly deviating towards the poles.

1 Introduction

The radical nitrate NO_3 is important in the stratospheric nighttime photochemistry. It controls the level of nitrogen oxides ($NO_x = NO + NO_2$), whose reactions in the middle atmosphere, form the primely catalytic ozone destruction cycle (Marchand et al., 2004). NO_3 has strong diurnal variation, and during the sunrise and sunset photolysis extremely quickly destroys NO_3 in the presence of sunlight. During the nighttime, in the absence of heterogeneous processes, the NO_3 chemistry scheme is believed to be relatively simple with three reactions:

$$NO_2 + O_3 \rightarrow NO_3 + O_2 \tag{R1}$$

$$NO_3 + NO_2 + M \rightarrow N_2O_5 + M \tag{R2}$$

$$N_2O_5 + M \rightarrow NO_3 + NO_2 + M \tag{R3}$$

NO_3 is mainly produced by Reaction (R1) of NO_2 and O_3. The sink of NO_3 is the Reaction (R2) with NO_2 which produces N_2O_5, which during the polar winter and spring, reacts on the surface of stratospheric sulphate aerosol heterogeneously to form HNO_3

and polar stratospheric clouds (Amekudzi et al., 2005). The reverse Reaction (R3) is an additional source of NO_3.

Historically NO_3 has been observed by ground-based lunar measurements, and the first measurements of NO_3 were published by Noxon et al. (1978). In addition to ground-based measurements, balloon-borne measurements to observe the vertical structures, have been made using stellar and lunar occultations (Naudet et al., 1981; Renard et al., 1996). Recently, NO_3 slant-column densities have been observed through sunrise and sunset using limb-scattered solar light measured by OSIRIS (McLinden and Haley, 2008).

Due to the strong diurnal variation of NO_3, in practice existing only nighttime and being undetectable during daytime, there are only a few available data sets of satellite-borne NO_3 profiles. The GOMOS instrument provides a long data set of simultaneous NO_2 and NO_3 observations since August 2002 (Hauchecorne et al., 2005; Kyrölä et al., 2010a). While GOMOS uses stellar light as a light source to measure the vertical structures of the atmosphere, SAGE III and SCIAMACHY employ the lunar occultation technique to observe NO_3.

The strong diurnal variation makes the validation challenging and many previous validations of the satellite-borne NO_3 profiles include models and/or chemical data assimilation schemes (e.g. Marchand et al., 2004; Amekudzi et al., 2007). In the study of Marchand et al. (2004) self-consistency of GOMOS NO_3, NO_2 and O_3 was verified. Their results also indicate that there is no substantial bias in GOMOS NO_3 data. In the study of Renard et al. (2008), four GOMOS NO_3 profiles were compared against balloon-borne observations as an "one-shot" validation exercise.

In our understanding this is the first paper, where the satellite-borne NO_3 profiles are directly compared with each other.

2 Materials and methods

2.1 GOMOS data

The GOMOS (Global Ozone Monitoring by the Occultation of Stars) instrument was launched on 1 March 2002 by European Space Agency on board the ENVISAT platform (Bertaux et al., 2010). Since August 2002 GOMOS has provided more than 850 000 individual vertical profiles of ozone, NO_2, NO_3 and other species. About half of the occultations are made during nighttime. In the beginning of the mission the instrument made about 400–500 occultations in a day, but due to the instrumental problems leading to the reduced viewing angle, the number of occultations has been somewhat smaller since January 2005.

The GOMOS inversion has been split in two parts: the spectral inversion and the vertical inversion (Kyrölä et al., 2010b). In the first part, horizontally integrated line densities of O_3, NO_2, NO_3 and aerosols are retrieved simultaneously using a combination of absolute and differential cross sections. In the second part, NO_3 profiles are retrieved from these horizontally integrated line densities at different tangent altitudes. In the latter part Tikhonov regularization is applied to compensate low signal to noise ratio. The vertical resolution of the NO_3, with the current regularization parameter, is 4 km while sampling resolution is smaller (0.5–1.7 km). Beside smoothing requirement used in Tikhonov regularization, the GOMOS inversion does not use any a priori information of NO_3 profiles.

As GOMOS uses stellar light as a light source, the quality of the measurements and the observations varies from star to star. In the study of Tamminen et al. (2010) GOMOS data were fully characterized. They concluded that NO_3 can be observed with 25–45 km altitude range with the precision of 20–40 % with the bright and medium bright stars, and noted also that the cool stars are slightly more favorable for the NO_3 retrieval. In this work we study only the NO_3 profiles that have been retrieved using stars brighter than 1.9 magnitude and cooler than 15 000 K. We have also screened the

GOMOS data so that the solar zenith angle is higher than 107°. We use the GOMOS data version IPF 5.0.

2.2 SAGE III data

SAGE III continued the heritage of SAGE I (1979–1981) and SAGE II (1984–2005) by measuring ozone, nitrogen dioxide, water vapor, and aerosol extinctions by solar occultation in addition to performing new measurements using lunar occultation and limb scattering techniques (McCormick et al., 1989). The instrument was launched 10 December 2001 on board a Russian Meteor-3M spacecraft, and the instrument recorded data between 7 May 2002–26 October 2005 in lunar occultation mode.

The current SAGE III lunar data set version 3.0 includes 583, 717, 959 and 302 vertical profiles for the years 2002, 2003, 2004 and 2005, respectively. The v3.0 data set contains nocturnal vertical profiles of ozone, NO_2, and NO_3 with near-global coverage. Approximately 32 % of SAGE III lunar measurements occurred between 23:00 and 24:00 local standard time at the tangent point, and approximately 49 % of the measurements occurred poleward of 60 degrees latitude. Measurements were attempted when the lunar phase was 40 % or greater and the solar zenith angle greater than 95 degrees. Data reduction was accomplished using techniques similar to the SAGE II (Chu et al., 1989) and SAGE III solar processing (Chu and Veiga, 1998). The gas species retrieval algorithms were developed prior to launch using a complex forward simulation model. The simulation incorporated a solar spectrum over the SAGE III wavelengths reflected by a modeled lunar disk with variable albedo, ray tracing through the atmosphere, and the effects of Rayleigh scattering and absorption by molecular gases and aerosols. Briefly, the retrieval procedure used a differential optical absorption spectroscopy algorithm (Platt et al., 1979) to compute gas species line of sight concentrations and then performed an inversion using an onion peel algorithm to compute number density concentrations.

The vertical resolution of the SAGE III NO_3 profiles is 1 km and the data are given on a 0.5 km fixed grid between 20–60 km. For a detailed description of the lunar

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processing algorithms, instrument, cross section data, and measurement accuracies see the SAGE III ATBD Team (2002). In this study SAGE III data has been screened, so that the solar zenith angle is higher than 100°.

2.3 Collocations and comparisons

These two data sets provide unique information of the nighttime NO_3 profiles measured from the Earth's limb. Temporal overlap of the data sets starts in August 2002 and continues until the end of 2005. Still, finding suitable collocations between GOMOS and SAGE III is quite difficult, because the two data sets are not homogeneously distributed (see Fig. 1). So in practice, we need to find a compromise between spatio-temporal limits and statistical representativeness. In order to find matches between GOMOS and SAGE III, we set the maximum latitudinal and longitudinal difference to be 2 and 5 degrees, respectively. For temporal difference we allowed the measurements to be 24 h away from each other, and at same time set the local hour difference to be less than 2 h. With this criteria we found 5, 28, 9 collocated pairs for the years 2002, 2003 and 2004, respectively. For the year 2005 we did not find any matches, mainly due to the fact that SAGE III measured most of its data in time when GOMOS was suffering from a technical anomaly. If we allow the temporal difference to be one week, still demanding the local hour difference to be less than 2 h, we can find 148 matches. In case multiple GOMOS matches were found to an individual SAGE III profile we selected the one that had smallest time difference. The spatial distribution of these 148 matches is shown in Fig. 2.

For statistical analysis we use symmetrically normalized GOMOS to SAGE III difference defined as $200 \times (\text{GOMOS} - \text{SAGE III})/(\text{GOMOS} + \text{SAGE III})$ [%]. Because GOMOS and SAGE III profiles share different vertical resolution, GOMOS averaging kernels are applied to SAGE III profiles.

3 Results

Herein, we seek to verify the NO_3 measurement accuracies by direct comparison with GOMOS and SAGE III NO_3 observations.

In Fig. 3 a good individual match, illustrating a two-peaked NO_3 profile before and after the application of the GOMOS averaging kernel, is shown. In Fig. 4, for the same example, we show the GOMOS spectral fit at 40 km and the mean GOMOS spectral fit calculated from the tangent heights between 30 and 45 km. In Fig. 4, the NO_3 absorption features located at 623 nm and 662 nm are clearly visible. The GOMOS star used in this occultation is star number 2, which is optimal for measuring NO_3. It is bright and cool star and thus the signal to noise ratio around NO_3 absorption bands is high.

In Fig. 5 we show the statistics calculated from 42 collocated pairs found from the years 2002–2004. Black solid line is the median of the individual differences and the dashed lines correspond to median ± interquartile deviation. Green solid line is the mean and green horizontal lines represents its 95 % confidence limits (±standard error of the mean × 1.96). For calculating the mean, we neglect the differences where the distance between the value and the median value is higher than 3 × 1.4826 × median absolute deviation in order to exclude clear outliers. From Fig. 5, we observe quite large deviations and median values oscillating between ±25 %, but we cannot note any negative or positive bias pattern between the altitudes 25 km and 45 km. Above 50 km the median of GOMOS to SAGE III difference increases up to 100 %. These findings are also valid for the years 2003 and 2004 separately. For the year 2002 the structure of the differences is very noisy mainly due to the fact, that we have only 5 collocated pairs (not shown in figures).

The statistics from 148 collocate pairs, where the temporal difference is allowed to be one week, are shown in Fig. 6. We can observe small positive bias of some 10 % below 40 km. Since this bias does not appear in Fig. 5, it may not be real, but due to the effects caused by too large temporal limits. From interquartile deviation we can clearly see that the spread between the observations starts growing below 30 km and above 45 km. One can observe, that the general structures of the medians in Figs. 5 and 6 are similar.

In addition to differences of collocated pairs we compared also the zonal medians in different latitude bands in 2004. In order to make these zonal medians more comparable we concentrated on three month periods where the distributions inside the latitude band are closest. Based on Fig. 1 for latitude band 90° S–60° S we selected months 8–10, for 60° S–30° S months 3–5 and for other latitude bands we selected months 10–12.

The results from these six different latitude bands are showed in Fig. 7. We can see similarities with these profiles: in the tropics being almost identical and slightly deviating towards the poles, still representing the same features. We must note that the number of profiles (see the caption of Fig. 7), that is used to calculate the median, is far from equal. Also temporal and spatial sampling of the observations vary, although we have concentrated on areas where the distributions are closest. Still these comparisons confirm that GOMOS and SAGE III nighttime NO_3 climatologies agree well with each other and we do not observe any clear systematic differences between them.

4 Conclusions and remarks

In this work we compared GOMOS and SAGE III NO_3 vertical profile data sets, retrieved using stellar and lunar occultation technique, respectively. Statistical analysis of limited amount of collocated pairs indicates a good overall agreement between GOMOS and SAGE III. Between the altitudes 25 km and 45 km the median difference between these two data sets is within ±25 %. From the zonal median profiles we can see reasonable agreement, showing that the climatological median profiles are comparable. The agreement is at its best in the tropics and slightly worse in other latitude bands.

It is worth noting that, despite the noise and other limitations, these two data sets are the only publicly available data sets of NO_3 vertical profiles, leaving GOMOS the sole since the termination of the SAGE III mission in 2006. Vertical profiles of NO_3 are also retrieved from ENVISAT/SCIAMACHY lunar occultations (Amekudzi et al., 2005), but unfortunately the limited scientific data set did not provide useful information (no collocations found) for comparison with the GOMOS NO_3 vertical profiles. A copy of the SAGE III instrument will be deported on International Space Station (ISS) in 2014. It will continue to record data in lunar occultation mode and hence it will provide NO_3 vertical profiles.

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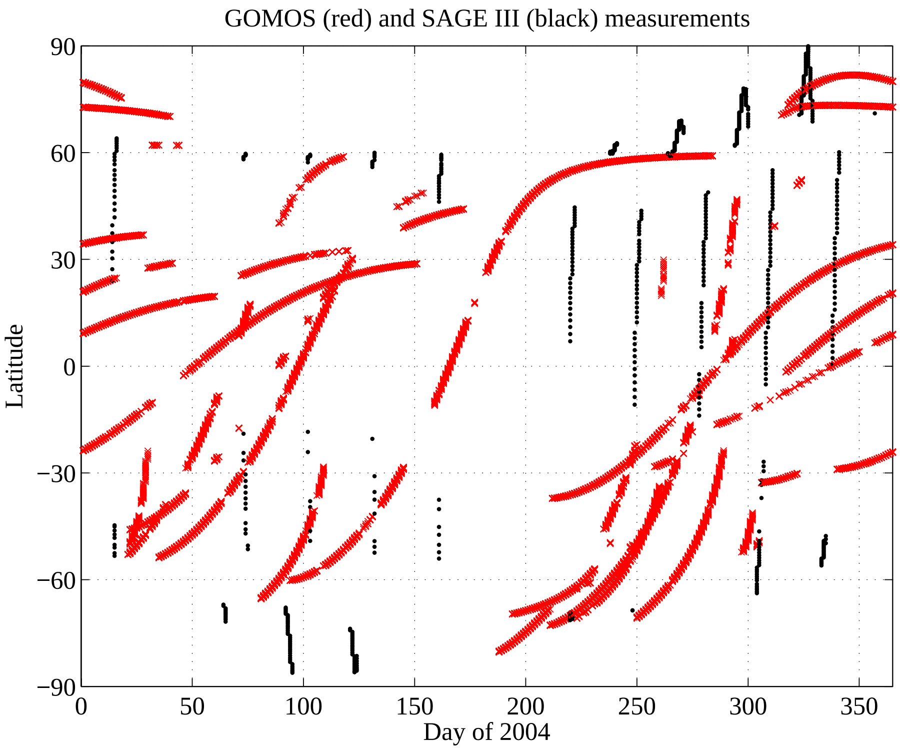

Fig. 1. GOMOS (red) and SAGE III (black) measurements in the year 2004. The GOMOS data has been screened, so that the solar zenith angle is higher than 107° and the stars used are brighter than 1.9 magnitude and cooler than 15 000 K. The SAGE III data has been screened, so that the solar zenith angle is higher than 100°.

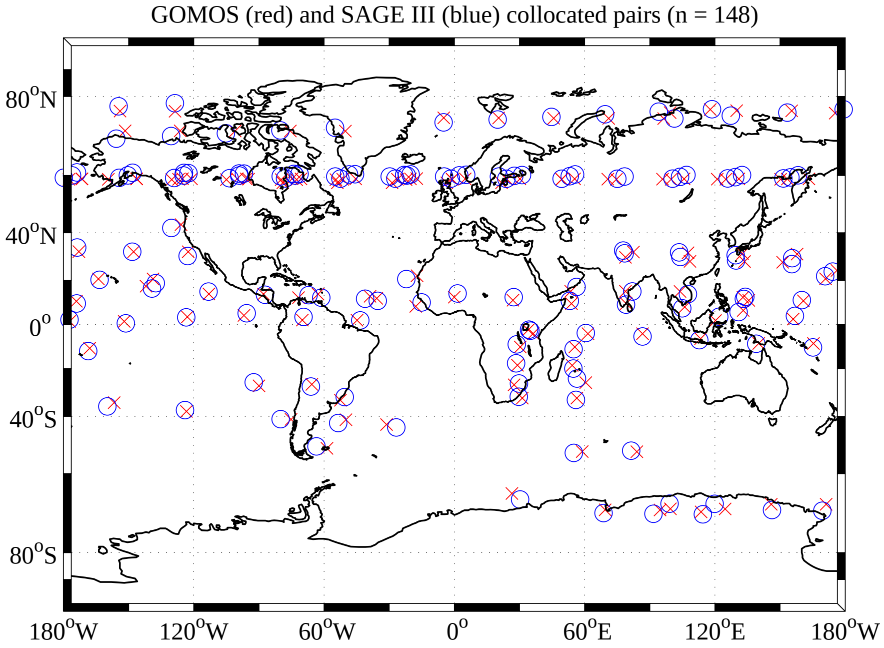

Fig. 2. Spatial distribution of 148 collocated GOMOS-SAGE III pairs (red cross and blue ring) from the years 2002–2004.

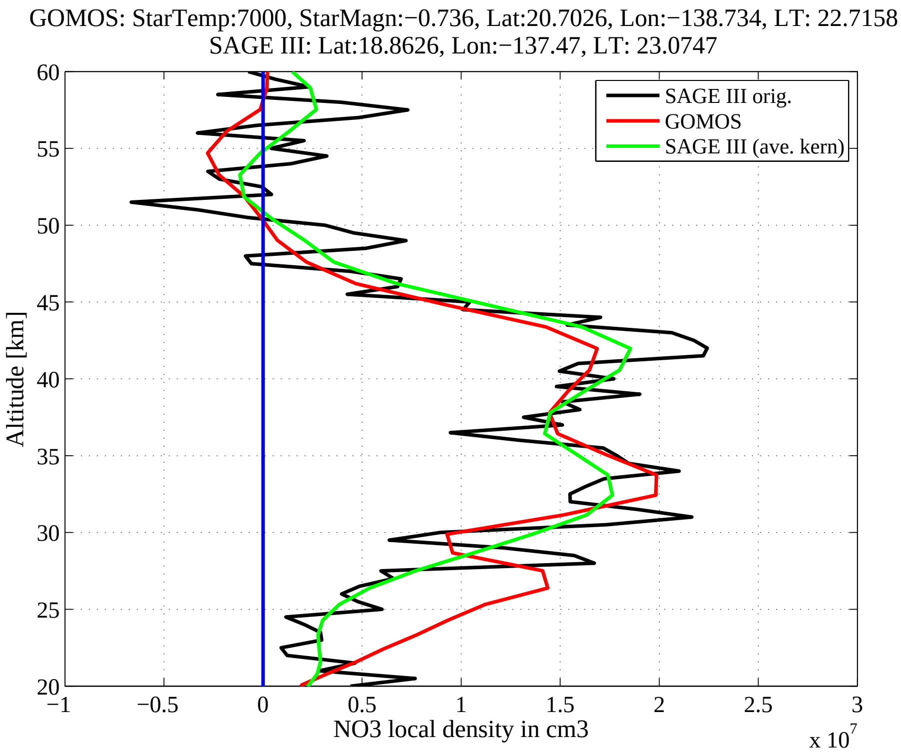

Fig. 3. A very good individual match between GOMOS and SAGE III. SAGE III profile is plotted before and after the GOMOS averaging kernel was applied. Figure illustrates two-peaked NO_3 profile.

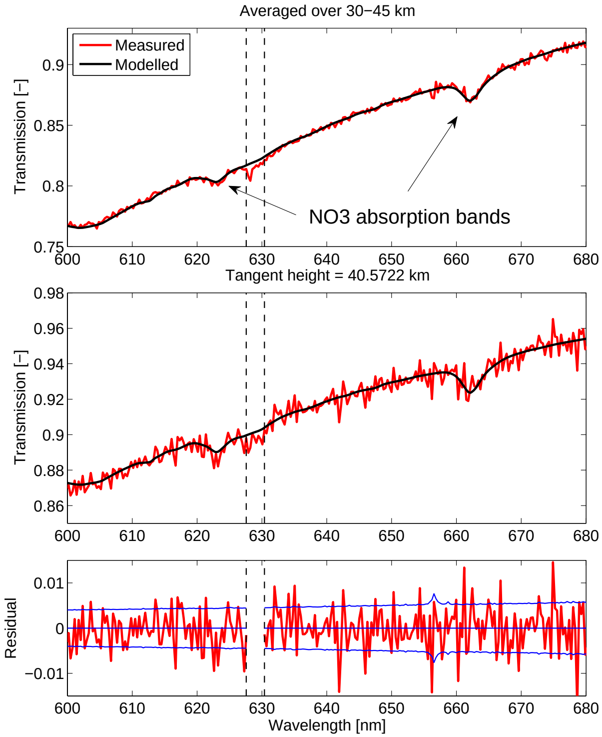

Fig. 4. Uppermost panel shows GOMOS mean spectral fit calculated from the tangent heights between 30 and 45 km. Middle panel shows spectral fit at 40 km and the lowermost panel is the residual at 40 km. The example is the same as in Fig. 3. NO_3 absorption features are clearly visible. Note that in the GOMOS Level 2 processing the spectral range 627.9–630.0 nm covering the oxygen line has been flagged and this area is not used in the retrieval (Kyrölä et al., 2010b).

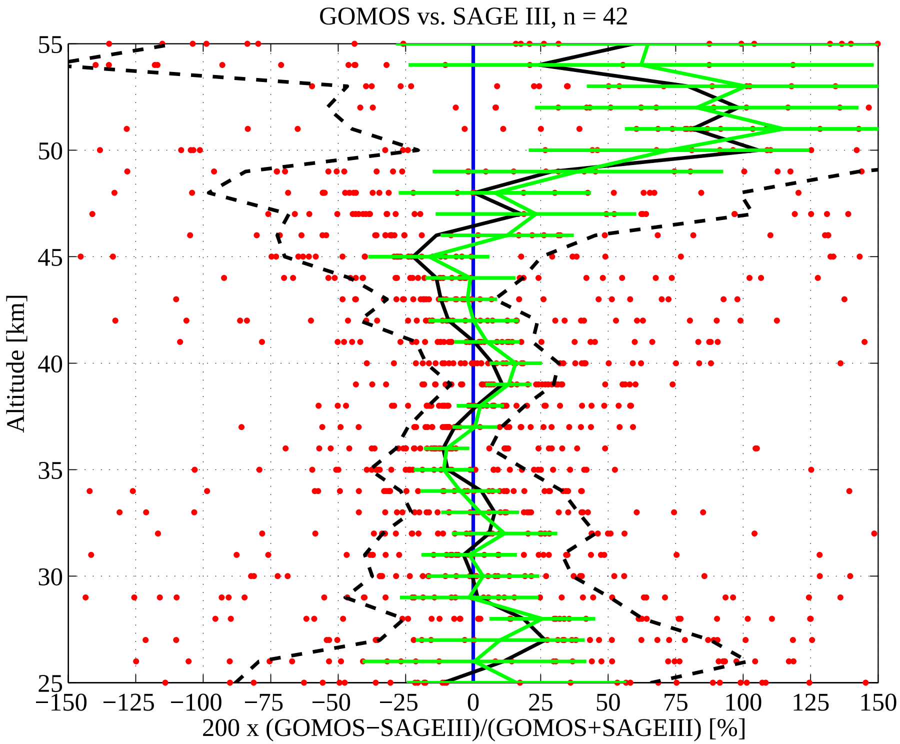

Fig. 5. The statistics of the collocated GOMOS-SAGE III pairs from the years 2002–2004. Black solid line is the median of the individual differences and the dashed lines correspond to median ± interquartile deviation. Green solid line is the median filtered mean and green horizontal lines represents its 95 % confidence limits.

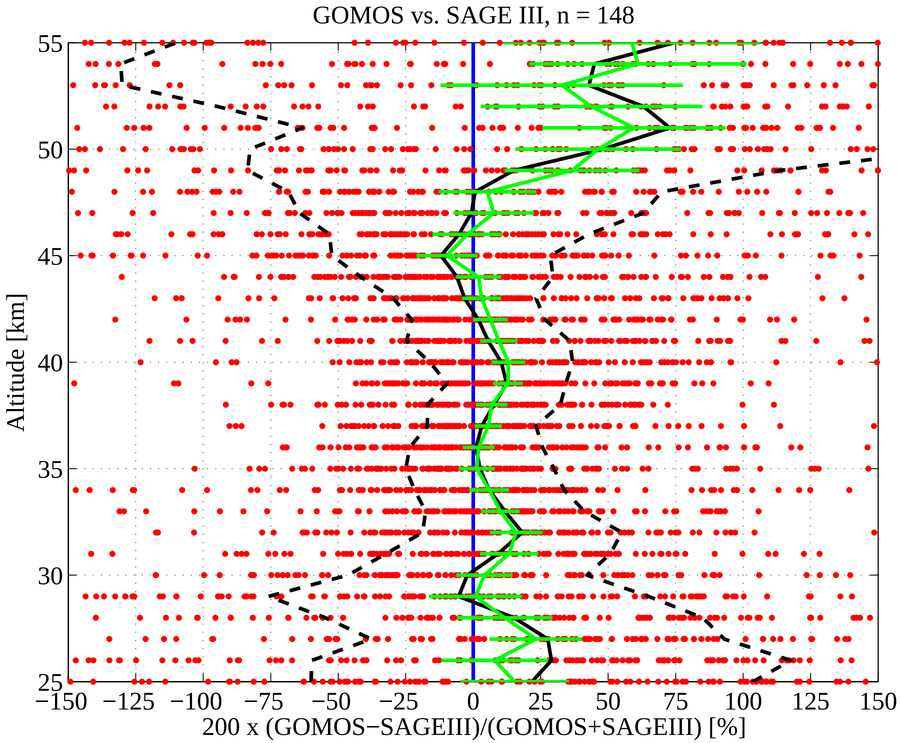

Fig. 6. Same as Fig. 5., except the temporal difference between the measurements is allowed to be as much as one week.

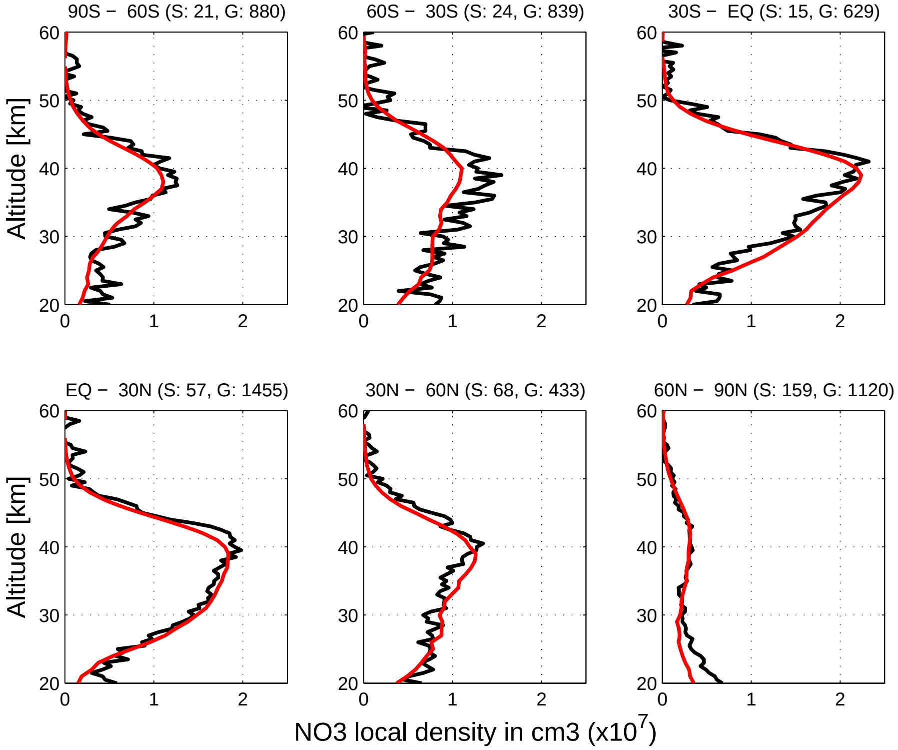

Fig. 7. GOMOS (red) and SAGE III (black) zonal medians in six different latitude bands in 2004. The zonal medians are calculated from three month periods where the spatial and temporal distributions of the measurements inside the latitude band is closest. The numbers after S and G in titles indicate from how many profiles the median was calculated for SAGE III and GOMOS, respectively.