

# Tropospheric ozone columns and ozone profiles for Kiev in 2007

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## Abstract

We report on ground-based FTIR observations being performed within the framework of the ESA-NIVR-KNMI project 2907 entitled “OMI validation by ground based remote sensing: ozone columns and atmospheric profiles” for the purpose of OMI data validation. FTIR observations were performed during the time frames August-October 2005, June-October 2006 and March-October 2007, mostly under cloud free and clear sky conditions and in some days from early morning to sunset covering the full range of solar zenith angles possible. Ozone column and ozone profile data were obtained for the year 2005 using spectral modeling of the ozone spectral band profile near 9.6 microns with the MODTRAN3 band model based on the HITRAN-96 molecular absorption database. The total ozone column values retrieved from FTIR observations are biased low with respect to OMI-DOAS data by 8-10 DU on average, where they have a relatively small standard error of about 2%. FTIR observations for the year 2006 were simulated by MODTRAN4 modeling. For the retrieval of ozone column estimates and particularly ozone profiles from our FTIR observations, we used the following data sources to as input files to construct the (a priori ) information for the model: satellite Aqua-AIRS water vapor and temperature profiles; Aura-MLS stratospheric ozone profiles (version 1.5), TEMIS [4] climatological ozone profiles and the simultaneously performed surface ozone measurements. Ozone total columns obtained from our FTIR observations for year 2006 with MODTRAN4 modeling are matching rather well with OMI-TOMS and OMI-DOAS data where standard errors are 0.68% and 1.11%, respectively. AURA-MLS data of version 2.2 which have become available in 2007 allow us to retrieve tropospheric ozone profiles. For some days Aura-TES tropospheric profiles were also available and were compared with our retrieved profiles for validation. A preliminary analysis of troposphere ozone variability was performed. Observation during the time frame March-October demonstrate daily photochemical variability of tropospheric ozone and reveal mixing processes during the night.

## 1 Introduction

It is common knowledge that the stratospheric ozone layer is very important for sustaining life on Earth – the ozone layer protects life on Earth from the harmful and damaging ultraviolet

solar radiation. Ozone in the lower atmosphere, or troposphere, acts as a pollutant but is also an important greenhouse gas. Ozone is not emitted directly by any natural source. However, tropospheric ozone is formed under high ultraviolet radiation flux conditions from natural and anthropogenic emissions of nitrogen oxides ( $\text{NO}_x$ ) and volatile organic compounds (VOCs). Satellite remote sensing is used to understand and quantify key processes in the global ozone budgets. Nowadays satellite observations are readily available for total ozone column and atmospheric ozone profiles. Nevertheless, ground based monitoring is important to validate and to complement space-based measurements and to clarify local/regional specific sources and sinks of this gas. Such ground based data can assist to derive the dynamical behavior of air pollution from space and ground-based observations and to check compliance to the pollutants transport models. They will also aid to the development of an environmental policy, in particular policies on greenhouse gases, on a local and regional scale.

Our first attempts to obtain total ozone columns from FTIR direct sun observations in the Main Astronomical Observatory were successful [11] and allows to us to submit the proposal on OMI validation, which was accepted.

## 2 OMI satellite observations

The Dutch-Finnish Ozone Monitoring Instrument (OMI) [5, 6] aboard the NASA Earth Observing System (EOS) Aura satellite [10] is a compact nadir viewing, wide swath, ultraviolet-visible (270–500 nm) hyperspectral imaging spectrometer that provides daily global coverage with high spatial and spectral resolution. The Aura orbit is sun-synchronous at 705 km altitude with a 98 degrees inclination and ascending node equator-crossing time roughly at 13:45. OMI measures backscattered solar radiance in the dayside portion of each orbit and solar irradiance near the northern hemisphere terminator once per day. The OMI satellite data products are derived from the ratio of Earth radiance and solar irradiance. At the time of writing the OMI TOMS [2, 5, 7] and OMI DOAS [2, 7, 13] total ozone column estimates are publicly available from the NASA DISC systems. The OMI-TOMS algorithm is based on the TOMS V8 algorithm that has been used to process data from a series of four TOMS instruments flown since November 1978. This algorithm uses measurements at 4 discrete 1 nm wide wavelength bands centered at 313, 318, 331 and 360 nm.

The OMI-DOAS algorithm [13] takes advantage of the hyper-spectral feature of OMI. It is based on the principle of Differential Optical Absorption Spectroscopy (DOAS) [8]. The algorithm uses  $\approx 25$  OMI measurements in the wavelength range 331.1 nm to 336.6 nm, as described in [13]. The key difference between the two algorithms is that the DOAS algorithm removes the effects of aerosols, clouds, volcanic sulfur dioxide, and surface effects by spectral fitting while the TOMS algorithm applies an empirical correction to remove these effects. In addition, the TOMS algorithm uses a cloud height climatology that was derived using infrared satellite data, while the DOAS algorithm uses cloud information derived from OMI measurements in the 470 nm  $\text{O}_2\text{-O}_2$  absorption band. The two algorithms also respond to instrumental errors very differently. Validation is key to quantify and understand these differences as a function of measurement geometry, season and geolocation.

## 3 Ground based ftir observations

Ground based FTIR observations are performed with a Fourier Transform Infra-Red (FTIR) spectrometer, model “Infracum FT 801”, which was modernized for the task of monitoring the

atmosphere by direct sun observations [3]. The main advantage of this device is its small size and small sensitivity of the optical arrangement to vibrations. The working spectral range of the FTIR spectrometer is 2–12 microns ( $800\text{--}5000\text{ cm}^{-1}$ ) with the highest possible spectral resolution of about  $1.00\text{ cm}^{-1}$ . Following the modernization of our spectrometer and updating the software for the initial treatment of the registered spectra in 2006, the system now allows us to average 2–99 individual spectra during the observation period. We averaged 4 single spectra as was recommended by the developers of the spectrometer device to avoid a degradation of the averaged spectrum due to the recording of atmospheric instabilities at longer exposure times. Our averaged spectra have signal-to-noise ratios S/N of 150–200. We registered 3–4 averaged spectra during 2–3 minutes of recording time.

Prior to further treatment of the observed spectra we checked the repeatability of these 3–4 spectra and choose the spectrum with the best signal-to-noise ratio S/N to be fitted with the model spectra.

## 4 Modtran spectra modeling and analysis

The column amounts of ozone ( $\text{O}_3$ ) molecules are recovered by using the radiation transfer codes MODTRAN3 and MODTRAN4, a moderate resolution model of transmission [1]. These codes are widely applied to the interpretation of ground based, airborne and spaceborne (satellite) observations of spectra of the Earth's atmosphere. The codes calculate atmospheric transmission and reflection of electromagnetic radiation with frequencies from 0 up to  $50000\text{ cm}^{-1}$ . The model uses a spherical source function for the light originating from the Sun and scattered from the Moon, and standard model atmospheres and user specified atmospheric profiles of gases, aerosols, clouds, fogs and even rain. It uses a two-parameter (temperature and pressure) model of molecular absorption bands, which is calculated on the basis of a large array of previously accumulated data of spectral lines stored in the HITRAN database. Here we use absorption cross-section data for 12 light molecules ( $\text{H}_2\text{O}$ ,  $\text{CO}_2$ ,  $\text{O}_3$ ,  $\text{CO}$ ,  $\text{CH}_4$ ,  $\text{O}_2$ ,  $\text{NO}$ ,  $\text{SO}_2$ ,  $\text{NO}_2$ ,  $\text{N}_2\text{O}$ ,  $\text{NH}_4$  and  $\text{HNO}_3$ ), for heavy molecules – CFC (9 molecules) and for  $\text{ClONO}_2$ ,  $\text{HNO}_4$ ,  $\text{CCl}_4$  and  $\text{N}_2\text{O}_5$ . The calculations are carried out only in an local thermal equilibrium (LTE) approximation for the moderate spectral resolution ( $2\text{ cm}^{-1}$ ) which just corresponds to our observed Fourier spectra. The Band Model parameters were re-calculated by us on the base of HITRAN-2004 according to the paper of [1]. Measurements of surface ozone concentrations by the collocated ozonometer together with satellite remote sensing data from the Atmospheric Infrared Sounder Instrument (AIRS - <http://disc.gsfc.nasa.gov/AIRS/>) aboard the NASA EOS-Aqua platform and the Microwave Limb Sounder ((MLS, <http://avdc.gsfc.nasa.gov/Data/Aura/>) aboard the NASA EOS-Aura platform were used for the construction of atmospheric ozone, temperature and water vapor input profiles for the MODTRAN4.3 code. For the analysis of the 2006 FTIR observations we used MLS version 1.5 data, which then had a preliminary character. We modified the shape of the MLS stratospheric ozone profile to obtain a better fit to the MODTRAN4.3 model output and to our FTIR spectra of the ozone band around 9.6 microns [12].

Fortunately, in 2007 the all new and more precise version v2.2 of MLS data became available, that allows us to develop a new approach to the analysis: we now modified the input tropospheric ozone profile and we only scaled the stratospheric ozone profiles of Aura-MLS v2.2 data within 2–5% (declared precision of these data) without any modification to its shape. The tropospheric part of the input (a priori) ozone profile was constructed from surface ozone measurement and the TEMIS climatological (monthly averaged) ozone atmospheric profiles [4], which were downloaded from the TEMIS-KNMI website. In this way we tried to obtain the best possible fit of the model computed spectra to the FTIR observed spectral band on 9.6 mi-

crons. Aura-TES data available from the AVDC website were also used if they were available for observational days.

To modify the tropospheric ozone profiles we used a smooth function determined between the  $J1$  and  $J2$  points of altitude in the model atmosphere. For any  $J$  point of the model we then adopt:

$$x(B) = (J - J1)/(J2 - J1),$$

then

$$P_J = P0_J(1 + B(\sin x))^a$$

determines the shape of the correction function,  $a$  and  $B$  determine the amplitude of changes of input tropospheric ozone profile, where  $B > -1$  and  $a > 0$ . Using the MODTRAN4 code we compute a grid of the theoretical spectra. To determine the best fit parameters, we compare the observed and computed spectra following a two-step optimization procedure.

Firstly, we determined the best fit to observed water vapor lines in the spectral region 800–1240  $\text{cm}^{-1}$ , i.e., here we exclude the ozone band from the analysis. Secondly, we fit the observed spectrum around the 9.6 micron ozone band with the grid of calculated ozone bands including the previously determined best atmospheric water profile. Hence we determine tropospheric ozone profiles, total and tropospheric ozone column from the best fit of the modeled and observed ozone band spectra, where we included the unaltered Aura-MLS stratospheric profiles.

Figure 1 (a,b) presents the comparison of our FTIR total ozone column values with the OMI-DOAS and OMI-TOMS satellite total ozone column data for the year 2006 and 2007. On average the difference of satellite and ground based observations in 2006 amounts to 0.37 DU and -0.25 DU for OMI-DOAS and OMI-TOMS respectively, with a 8.77 DU and 5.37 DU standard deviation (1.11 DU and 0.68 DU standard errors). For 2007 average difference of satellite minus ground based amounts to -0.24 DU and -4.17 DU for OMI-DOAS and OMI-TOMS respectively, with 10.50 DU and 10.73 DU standard deviations (1.31 DU and 1.35 DU standard errors).

In Figure 2 we show in the left figure all the observed spectra for the day 29th of September 2007 (29.09.07). We also demonstrate the best fit of the modeled spectra to the FTIR spectrum observed at 13 h 01 min local time in Figure 2 (right).

The Figures 3-6 present our retrieved ozone profiles for four representative cases: two spring enhanced ozone episodes, summer ozone photochemistry episode and autumn low stratospheric ozone episode. The latter case was observed during 3 days: 29th of September 2007 (29.09.07), 1st of October 2007 (01.10.07) and the 2nd of October 2007 (2.10.07). The figures demonstrate the specifics of each episode and the daily dynamics of tropospheric ozone variability due to the underlying photochemical processes.

Figure 3 shows the retrieved ozone atmospheric profiles for the 28th of March 2007 (28.03.07), recorded at 08 h 54 min and 10 h 47 min (upper figures) local time, and 13 h 12 min and 18 h 21 min (lower figures) local time. From these figures one observes the low ozone concentrations in the boundary layer for the morning observation at 08 h 54 min LT. Here most probably ozone titration by the nitrogen oxides ( $\text{NO}_x$ ) as emitted from cars during the morning traffic is taking place. From the 10 h 47 min LT observation we see the abatement of tropospheric ozone, most clearly over the vertical range 2–11 km. The enhancement of ozone due to the photochemical processes in the atmosphere are seen in the lower two figures. Our simultaneously performed surface ozone measurements reflect this dynamics also with the supportive values 27.3 ppb, 40.2 ppb, 48.8 ppb, and 57.3 ppb recorded for exactly these moments in time. For the comparison, we also show the Aura-TES ozone vertical profile for the 28-th of March 2007 (28.03.07), which can be considered as the valid satellite profile in the troposphere only.

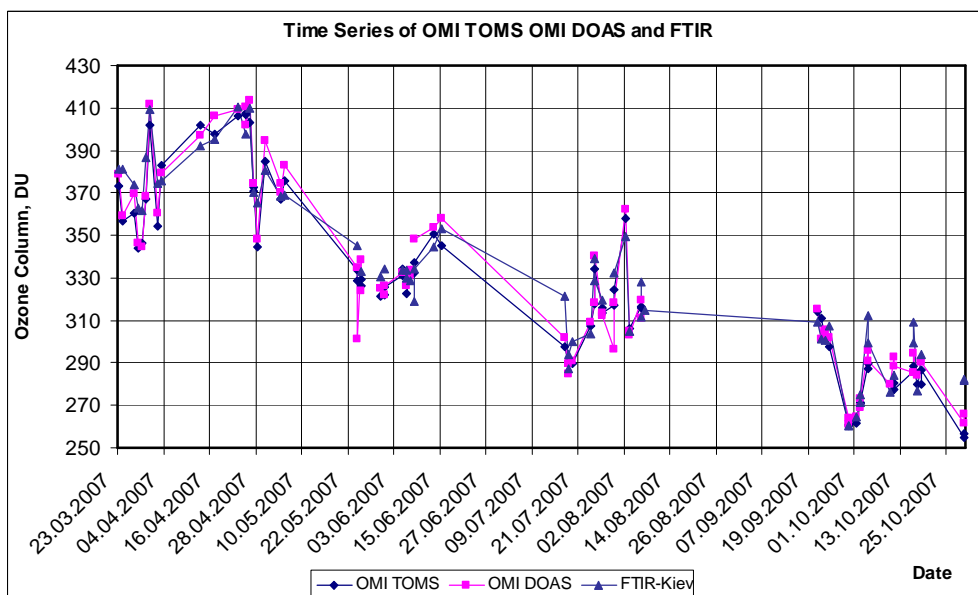
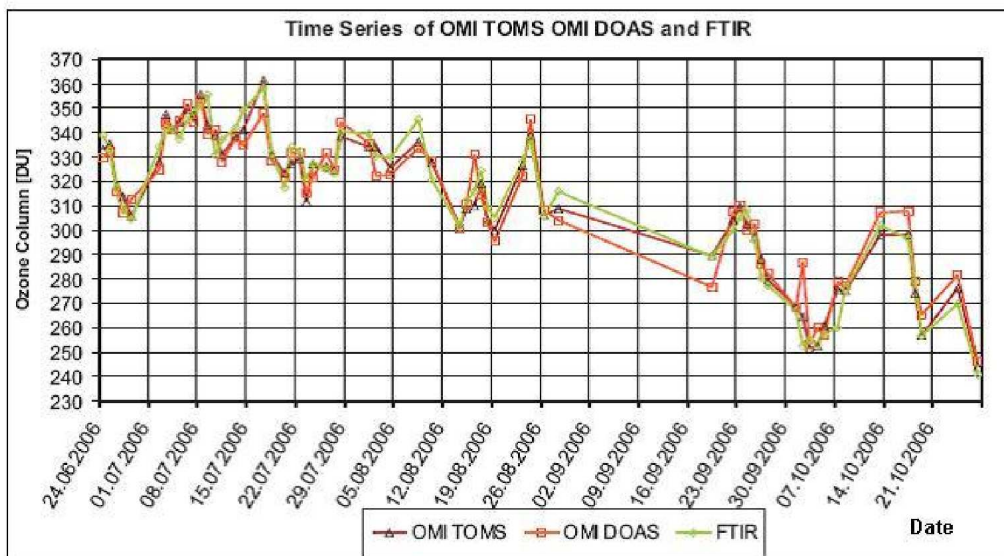


Figure 1: a) Time series of the OMI total ozone column and the ground based FTIR total ozone data of 2006 for the ground site of Kiev (MAO). Average difference of satellite minus ground based amounts to 0.37 DU and -0.25 DU for OMI-DOAS and OMI-TOMS respectively, with a 8.77 DU and 5.37 DU standard deviation (1.11 DU and 0.68 DU standard errors). b) Time series of the OMI total ozone and the ground based FTIR total ozone data of 2007 for Kiev (MAO). Average difference of satellite minus ground based amounts to -0.24 DU and -4.17 DU for OMI-DOAS and OMI-TOMS respectively, with 10.50 DU and 10.73 DU standard deviations (1.31 DU and 1.35 DU standard errors).

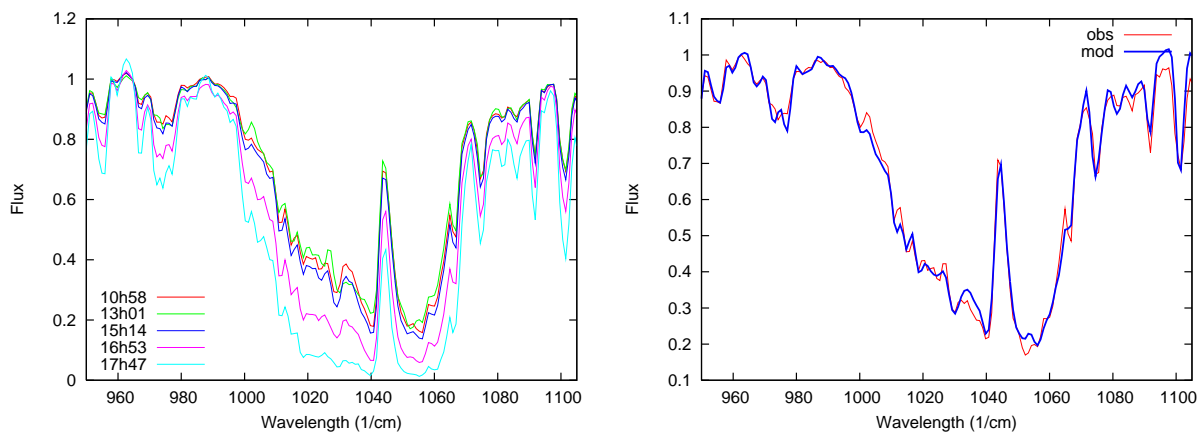


Figure 2: The observed FTIR spectra of the 9.6 micron ozone band for the 29th of September 2007 (29.09.07) (left) and the comparison of the observed FTIR spectra and modeled MODTRAN 4 spectra following the procedure for best fitting for the observation at 13 h 01 min local time on this day.

Figure 4 shows the retrieved ozone atmospheric profiles for the 23rd of April 2007 (23.04.07) recorded at 09 h 22 min and 11 h 15 min (upper figures) local time, and recorded at 14 h 35 min and 15 h 40 min (lower figures) local time. On this day the values of both total ozone columns (411.0 DU by FTIR) and tropospheric ozone columns are very high. Possibly we are here observing a stratospheric intrusion event as the highest OMI value of total ozone column in 2007 was 448 DU for the 22nd of April 2007 (22.04.07).

Figure 5 shows the retrieved ozone atmospheric profiles for the 18-th of July 2007 (18.07.07) recorded at 13 h 35 min and 16 h 10 min (upper figures) local time, and recorded at 17 h 20 min and 19 h 27 min (lower figures) local time. The very high tropospheric ozone columns and surface ozone concentrations (see, for the exact numbers) and their daily dynamics are characteristic for episodes of strongly enhanced surface and tropospheric ozone due to tropospheric photochemistry.

Please note that on this day the total ozone column is actually rather low (291.5 DU). Finally, in Figure 6 we show the retrieved atmospheric ozone profiles for the 1st of October 2007 (01.10.07) recorded at 8 h 8 min and 9 h 49 min (upper figures) local time and recorded at 16 h 21 min and 17 h 41 min (lower figures) local time. Please note that on this day the FTIR total ozone column is rather low: only 262 DU. Nevertheless, we can see the daily dynamics of tropospheric ozone: in the morning ozone titration by  $\text{NO}_x$  is taking place leading to rather high ozone concentrations later in the afternoon. Unfortunately, for this day Aura-TES data are absent and hence the tropospheric part of the input ozone profile for the MODTRAN modeling process was constructed on the basis of the TEMIS monthly averaged climatological data.

## 5 Conclusion

We have obtained a long series of total ozone column estimates on the base of ground based FTIR observations and MODTRAN modelling for the years 2005–2007. Our values of the total ozone columns agree well with OMI satellite remote sensing data. Differences are in the percentile range. We note some significant differences under insufficiently clear sky conditions, which are indicative of the influence of clouds on FTIR observations.

The work presented here is the first step towards ozone profile retrievals on a regular basis. For this we need to further develop our retrieval procedures and we need to perform testing of our model calculations through “line-by-line” radiative transfer model calculations alike FAS-

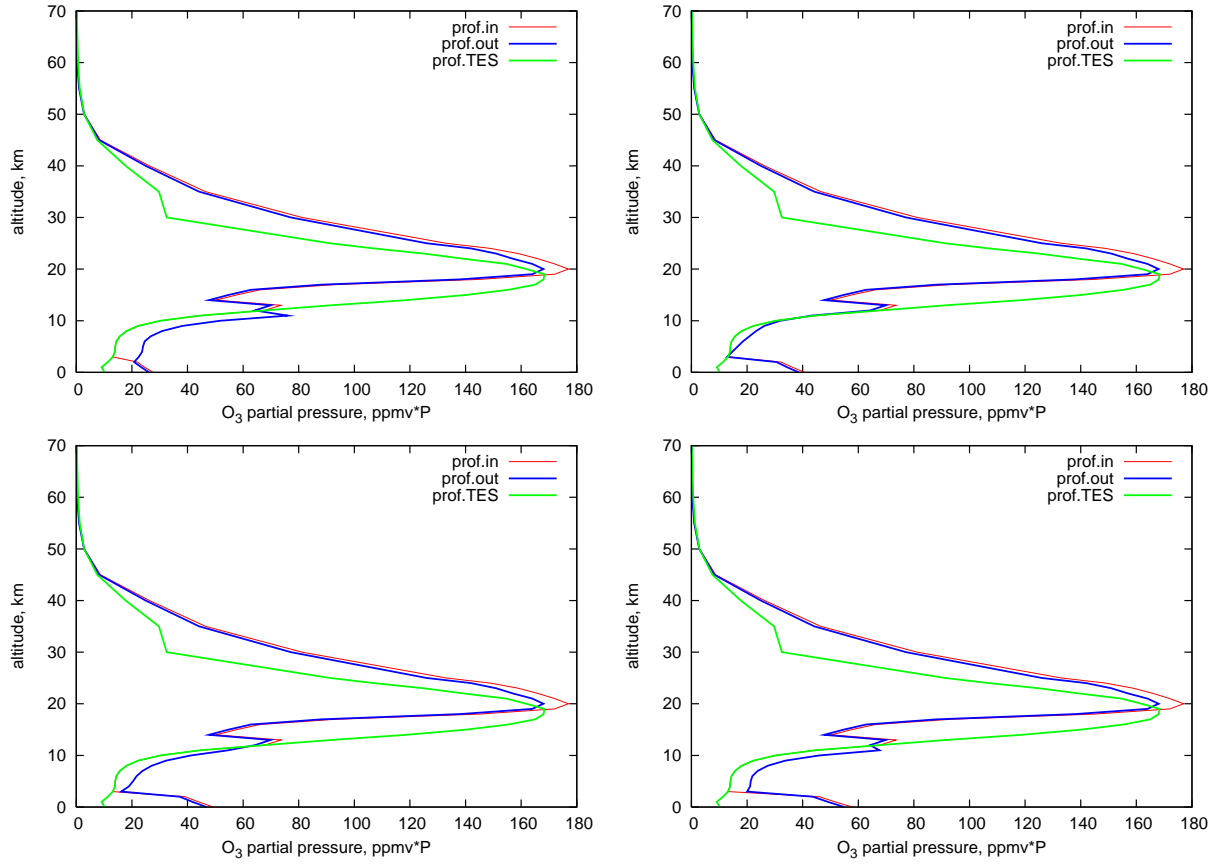


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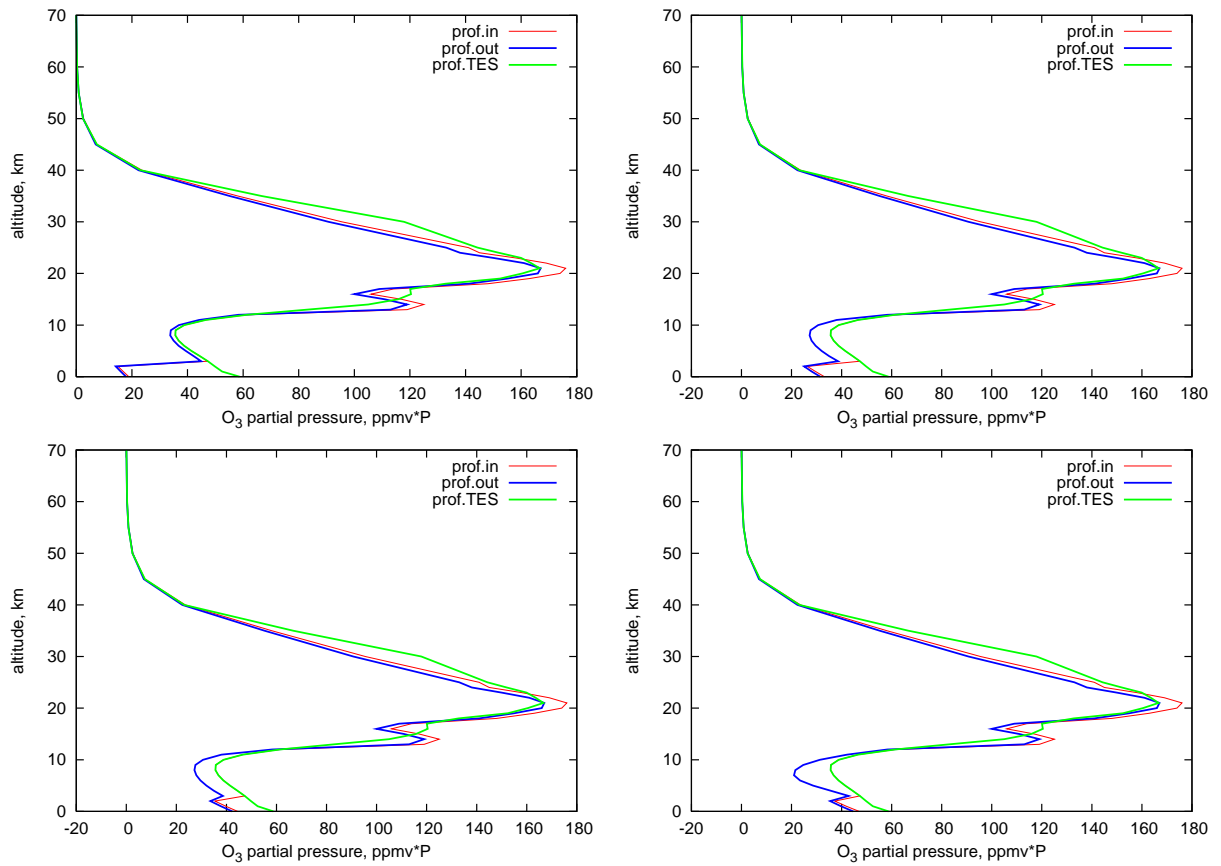


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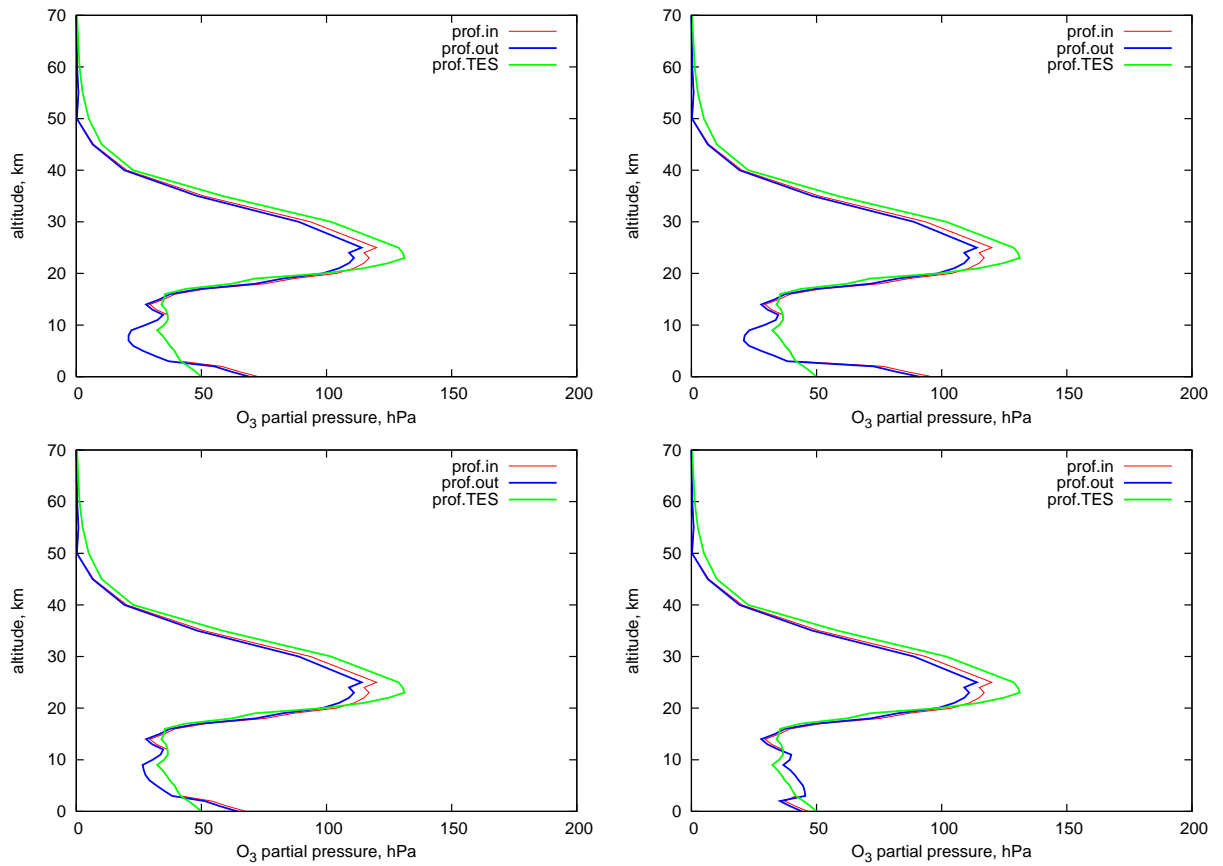


Figure 5: The retrieved ozone atmospheric profiles for the 18th of July 2007 (18.07.07) recorded at 13 h 35 min and 16 h 10 min (upper figures) local time, and recorded at 17 h 20 min and 19 h 27 min (lower figures) local time. The very high tropospheric ozone columns and surface ozone concentrations (see Table 1 for the exact numbers) and their daily dynamics are characteristic for episodes of strongly enhanced surface and tropospheric ozone due to tropospheric photochemistry. Please note that on this day the total ozone column is actually rather low (291.5 DU).

Table 1: Total ozone columns and tropospheric ozone columns for some days of 2007.

Date	Time, h, min	ZSA, grad	TOC, DU	OMI-TOMS, DU	OMI-DOAS, DU	Tr.OC, DU, our, TES	Surface O <sub>3</sub> , ppb	Htrop, km
28.03.07	8 54	70.434	364.24			47.15	27.3	12.0
	10 47	58.459	363.57			36.06	40.2	
	13 12	47.469	361.39	344.2	356.0	44.13	48.8	
	14 46	52.131	363.94	353.2	363.2	46.72	65.9	
	16 51	67.169	363.54			46.33	64.0	
	17 51	76.192	359.91			43.54	56.5	
	18 21	80.375	366.27			44.93	57.3	
23.04.07	9 22	57.622	411.01			48.06	18.7	12.5
	11 15	43.200	410.30			47.34	32.8	
	14 35	42.879	410.27	412.0 <sup>a</sup>	414.5	47.30	44.1	
	15 40	50.375	409.54	414.7	417.6	46.57	46.5	
9.06.07	6 39	75.28	348.37			38.40	20	12.0
	8 44	55.66	341.53			31.70	22	
	11 56	29.93	346.05	347.6	349.0	35.47	42.8	
	16 08	45.92	352.76			36.38	51	
	17 53	62.42	349.56			39.56	57	
14.06.07	6 52	73.20	355.54			42.9	14	12.0
	7 05	71.21	351.04			44.75	13	
	9 05	52.25	352.81			39.81	15	
	12 06	28.96	348.72	347.6	349.6	42.86	46	
	17 45	60.75	357.36			44.76	50	
18.07.07	13 35	29.93	287.12	291.5	289.6	44.32	72	12.6
	14 52	36.16	294.07			51.27	85	
	16 10	46.62	290.91			49.37	95	
	17 20	58.23	294.39			51.60	67	
	18 15	66.19	292.85			50.09	58	
	19 27	77.39	296.60			53.80	46	
						53.55 <sup>b</sup>		
29.09.07	10 35	57.722	269.21			29.96	13.0	13.0
	13 01	52.756	260.34	261.2	263.9	32.64	29.0	
	15 14	61.211	260.38			32.31	39.0	
	16 37	71.676	261.44	260.2	260.9	33.73	40.0	
	17 47	82.059	266.62			38.92	35.0	
1.10.07	8 08	79.704	271.75			30.31	8.0	12.5
	9 49	65.672	261.95			28.09	18.0	
	13 22	53.971	264.68	261.7	264.9	30.69	40.0	
	16 21	70.201	271.41			37.42	45.0	
	17 41	81.844	277.23			43.24	39.0	
2.10.07	8 31	76.545	279.16			40.43	8.0	12.5
	9 43	66.709	276.51			37.78	12.0	
	12 58	53.897	271.42	270.9	269.1	34.93	43.0	
	15 20	63.019	274.80			36.08	47.0	
					39.19 <sup>b</sup>			

<sup>a</sup>22.04.07 OMI total column value = 448 DU

<sup>b</sup>TESL3 tropospheric ozone column

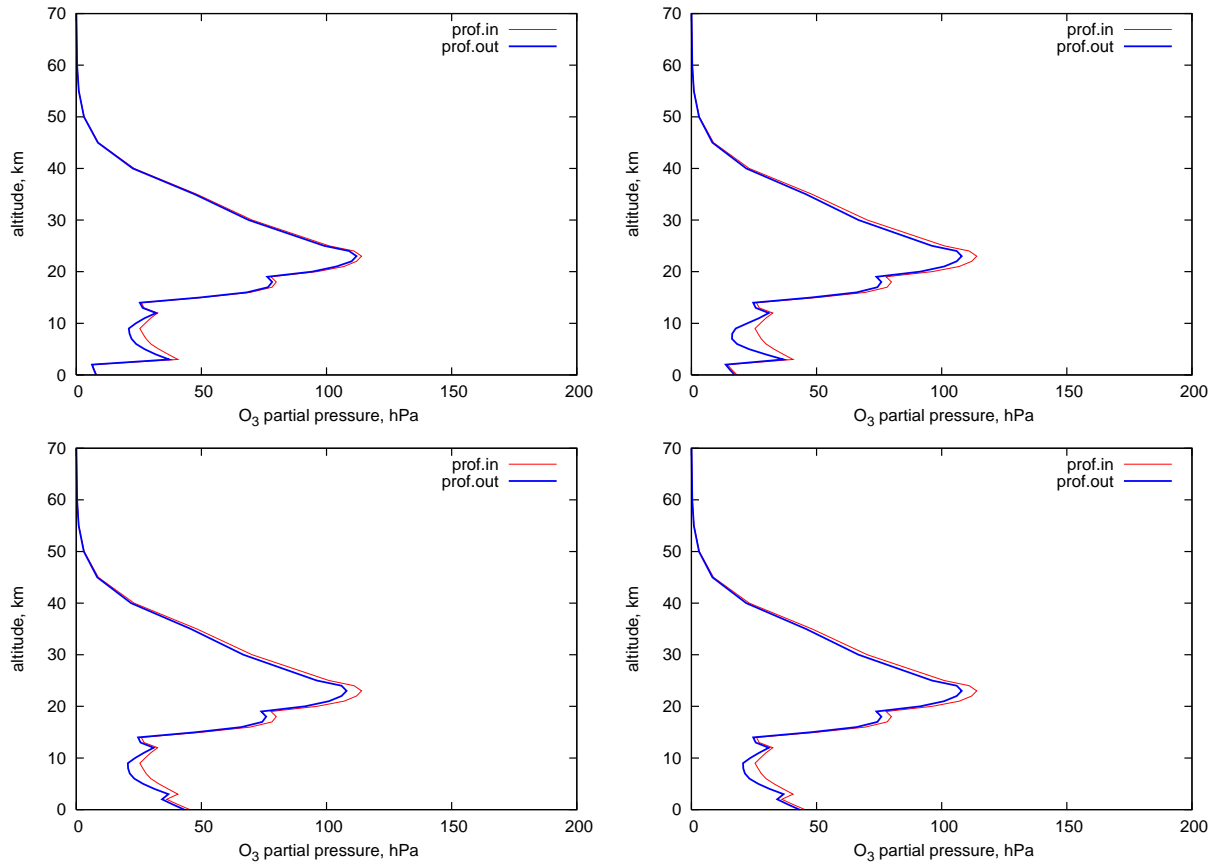


Figure 6: The retrieved atmospheric ozone profiles for the 1st of October 2007 (01.10.07) recorded at 08 h 08 min and 9 h 49 min (upper figures) local time and recorded at 16 h 21 min and 17 h 41 min (lower figures) local time. Please note that on this day the FTIR total ozone column is rather low: only 262 DU. Nevertheless, we can see the daily dynamics of tropospheric ozone: in the morning ozone titration by  $\text{NO}_x$  is taking place leading to rather high ozone concentrations later in the afternoon. Unfortunately, for this day Aura-TES data are absent and hence the tropospheric part of the input ozone profile for the MODTRAN modeling process was constructed on the basis of the TEMIS monthly averaged climatological data.

CODE. Since we do not have this code available we need to develop such coding in the near future ourselves. The procedure of quantitative comparison of our retrieved profile and other available data must be developed.

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