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Sensitivity of the OMI ozone profile retrieval (OMO3PR) to a priori assumptions

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Received: 31 January 2014 - Accepted: 14 February 2014 - Published: 25 February 2014

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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We have assessed the sensitivity of the operational OMI ozone profile retrieval (OMO3PR) algorithm to a number of a priori assumptions. We studied the effect of stray light correction, surface albedo assumptions and a priori ozone profiles on the retrieved ozone profile. Then, we studied how to modify the algorithm to improve the retrieval of tropospheric ozone. We found that stray light corrections have a significant effect on the retrieved ozone profile but mainly at high altitudes. Surface albedo assumptions, on the other hand, have the largest impact at the lowest layers. Selection of an ozone profile climatology which is used as a priori information has small effects on the retrievals at all altitudes. However, the usage of climatological a priori covariance matrix has a significant effect. Based on these sensitivity tests, we made several modifications to the OMO3PR algorithm: the a priori ozone climatology was replaced with a new climatology (TpO3), the a priori covariance matrix was calculated from the climatological ozone variance values, and the surface albedo was assumed to be linearly dependent on wavelength in the UV2 channel. We found that the a priori covariance matrix basically defines the vertical distribution of degrees of freedom for a retrieval. Moreover, all the studied versions of the OMO3PR algorithm were equally effective in reducing uncertainty in the retrieved ozone profile. This implies that the posterior error values depend mostly on the assumed a priori errors. Our case study over Europe showed that the new version produced over 10 % smaller ozone abundances which reduced the systematic overestimation of ozone in the OMO3PR algorithm and improved correspondence with IASI retrievals.

Introduction

Atmospheric ozone is distributed both in the stratosphere and the troposphere. In the stratosphere, ozone acts as a shield that protects the surface from energetic ultraviolet radiation. Tropospheric ozone, on the other hand, is a greenhouse gas that warms

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the atmosphere, and also a pollutant that causes respiratory problems in humans and damages crops. It is a short-lived species when compared with transport times, and therefore, inhomogeneously mixed. A large fraction of ozone precursors are emitted from anthropogenic sources. (Shindell et al., 2007) In order to understand the ozone related physical and chemical processes in the atmosphere, global measurements of vertical ozone profiles are essential. Consequently, the total ozone column and ozone profiles have been monitored with spaceborne instruments since the late 1970s. Over the years, several methods have been developed for ozone monitoring: instruments use both limb and nadir viewing geometries and spectral regions range from ultraviolet to microwave. Limb (and occultation) measurements have good vertical resolution but their horizontal resolutions are limited and they are not able to detect ozone in the lower troposphere. Microwave measurements are not affected by clouds and they can be done during night and day (like infrared measurements) whereas ultraviolet measurements are limited to daytime. However, nadir UV measurements have much better horizontal resolution than the other methods. As part of this line of instruments the Ozone Monitoring Instrument (OMI; Levelt et al., 2006a, b) on-board Earth Observing System (EOS) Aura (Schoeberl et al., 2006) satellite was launched in 2004. Currently, two algorithms are used to retrieve ozone profiles from the OMI measurements: operational OMO3PR retrieval developed at KNMI (Kroon et al., 2011) and a scientific algorithm developed at NASA (Liu et al., 2010). Both of the algorithms are based on the optimal estimation retrieval technique (Rodgers, 2000) but they differ in the implementation. For example, the algorithms use different radiometric calibration (the NASA algorithm uses vicarious calibration), radiative transfer models, vertical grids and a priori covariance matrices. The OMO3PR retrieval provides global coverage on daily basis with a vertical resolution of 6-7 km. Kroon et al. (2011) validated the retrieved ozone profiles with several satellite products and balloon-borne ozone sondes. As the summary in Table 1 shows, OMO3PR retrievals were in agreement with the Microwave Limb Sounder (MLS; Waters et al., 2006) retrievals within ±10% except for the Polar regions during the ozone hole seasons where differences up to ±30 % were found. The

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comparisons with MLS and other correlative data sets show that the biases vary with altitude. In addition, they found that OMI overestimates tropospheric ozone abundances from 0 to 30 %, when compared with MLS, the Tropospheric Emission Spectrometer (TES; Beer et al., 2001) and ozone sondes.

In this work, we studied how different assumptions in the OMO3PR retrieval affect the results and searched for ways to improve the accuracy of the retrieval. We concentrated on the a priori information: ozone climatology and the corresponding error covariance matrix, wavelength dependency of surface albedo, and stray light correction. In addition to a priori assumptions the retrieval is also affected by a number of other things, e.g. correction for rotational Raman scattering in the L1B reflectance data and cloud properties obtained from longer wavelengths. However, we limited this work to parameters that could be modify within the OMO3PR algorithm. First, we tested how sensitive the algorithm is to these a priori assumptions. Then, we implemented those a priori assumptions which appeared to improve the algorithm's performance in the troposphere. Finally, we compared the operational and the modified retrievals of tropospheric ozone to a case study over Europe presented in the paper by Eremenko et al. (2008).

This paper is organized as follows: first the operational OMI ozone profile retrieval algorithm is introduced in Sect. 2. In Sect. 3 we show how changes in the a priori assumptions affect the retrieved ozone profiles and select the most appropriate assumptions for the new version of the algorithm. In Sect. 4 we compare the results from the operational and modified algorithms with Infrared Atmospheric Sounding Interferometer (IASI; Clerbaux et al., 2009) retrievals over Europe. Section 5 summarizes our findings.

OMO3PR algorithm

Ozone Monitoring Instrument (OMI) is on-board the NASA Earth Observing System (EOS) Aura satellite. It is a nadir viewing, ultraviolet-visible (270-500 nm) imaging

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spectrometer, which provides daily global coverage with high spatial and spectral resolution (Levelt et al., 2006a, b). It has been measuring since 2004.

A detailed description of the OMI ozone profile algorithm (OMO3PR) is given by Kroon et al. (2011). Briefly, the retrieval is based on the strong decrease in the ozone ₅ absorption cross section between wavelengths of 270 nm and 330 nm. The radiation at the longer wavelengths goes through the whole atmosphere while the shortest wavelengths are only affected by the highest layers of the atmosphere. Therefore, spectral information of UV radiation can provide information on the vertical distribution of ozone. The measurements are taken from the UV1 channel (270.0–308.5 nm) and the first part of the UV2 channel (311.5-330.0 nm). The retrieval algorithm uses optimal estimation (Rodgers, 2000; termed maximum a posteriori method in the book), where the difference between the measured and modeled sun-normalized radiance is minimized by adjusting the amount of ozone in 18 atmospheric layers. This method requires a priori information on ozone profiles and other parameters like the surface albedo in order to constrain the retrievals. The operational ozone profile retrieval uses the LLM ozone climatology (McPeters et al., 2007), which varies with month and latitude. The a priori ozone profiles are given constant relative variability of 20 % except for ozone hole conditions. Ozone hole conditions are assumed to occur between August and December at latitudes south of 50°S. There, the variability is 60% for altitudes between 21 km and 50 km, and 30 % for the other altitudes. The vertical correlation length of ozone is an a priori constraint and it is set to 6 km. To ensure that the retrieved ozone volume mixing ratios are positive, the algorithm operates with logarithm of the volume mixing ratio for each layer.

Surface albedo is also fitted in the retrieval and the OMI surface albedo climatology (Kleipool et al., 2008) is used as an initial value for the surface underneath the atmosphere. The wavelength dependence of the albedo in both UV1 and UV2 channels is described with a second order polynomial. Surface albedo is fitted for all wavelengths (although the shortest ones do not "see" the surface) to partly account for the presence of aerosols and clouds which are not known or modeled in the retrieval.

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Stray light refers to light of other wavelengths that is scattered by the imperfect OMI optics onto the detector of a specific wavelength. This effect is more pronounced in the UV1 channel than in the UV2 channel because the detected radiances at the shorter wavelengths are smaller and, therefore, the measured signal is affected more by radiation from other wavelengths. The shortest wavelengths are reflected only from the highest altitude layers in the atmosphere, thus, stray light has the largest effect on the retrieval of ozone at these altitudes. Dobber et al. (2006) have provided a detailed description of stray light features in the OMI instrument. Regarding the ozone profile retrieval, correction for stray light is done in two steps. The first correction is done during the production of the L1B spectra (OML1BRUG; van der Oord et al., 2006). In this correction, specific wavelength ranges are used to define so-called source and target regions. For the source regions averaged signals are calculated using the information over the whole swath. These signals are multiplied by a polynomial that distributes the stray light over the target regions and then they are subtracted from all pixels in the corresponding target areas. The second stray light correction is done during the processing of the Level 2 ozone profile retrieval by fitting second order polynomials to both UV1 and UV2 channels separately.

Table 2 summarizes the above mentioned information. It presents the content of the optimal estimation state vector used in the OMO3PR retrieval. The state vector contains the ozone profile with 18 layers and six values for surface albedo and for stray light parameters. If the cloud fraction is lower than 0.2, surface albedos are fitted. Otherwise, cloud albedo values are fitted. In the operational retrieval, the effect of NO₂, SO₂ and aerosols are not considered. In addition to the actual ozone product, the OMO3PR algorithm produces metrics, like posterior error and averaging kernel, that can be used to evaluate the retrieval.

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versa.

Turning off the Level 1 (L1) stray light correction while doing the Level 2 (L2) correction caused only minor changes in the ozone profiles, as can be seen from Fig. 1c. As the plot shows, the systematic changes are small at higher altitudes and almost nonexistent in the troposphere. Moreover, the usage of the Level 2 stray light correction reduces variability at all altitudes as can be seen by comparing plot 1c with 1a and 1b. The large variations at the lowest altitudes are caused by cloud-free retrievals (cloud

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fraction < 0.1) where the retrievals without complete stray light correction produce over 10 times larger ozone amounts than the operational retrieval.

Turning off the Level 2 stray light correction affected the ozone profiles mainly in the stratosphere, as Fig. 1b shows. For some layers, the changes can be up to 20 % but on 5 average they stay below 10%. In the troposphere, the average difference is well below 10%. When comparing these values with the validation results shown in Table 1, it is evident that the effect of stray light correction is too small to explain completely the systematic differences between the OMI and other ozone profile retrievals.

Surface albedo parametrization

As mentioned in Sect. 2, the operational version of the retrieval algorithm uses second order polynomials to describe the dependency of surface albedo on wavelength at both UV1 and UV2 channels separately. To study the sensitivity of the retrieval to this assumption we varied the wavelength dependencies of the albedos. We tested constant, first order (linear) and second order polynomials for both channels separately. We processed all the pixels from one orbit (06704, 1496*30 pixels) and studied how the ozone profiles change when compared to the operational product on average and in six different latitude bands. In the comparison, we concentrated on tropospheric features. To our knowledge, there is no applicable surface albedo database for these wavelengths which could be used to select the most physical representation for the wavelength dependency. Therefore, we decided to concentrate on the configuration that provided the largest decrease in the tropospheric ozone. Kroon et al. (2011) had shown that OMI ozone values were consistently larger than TES or ozonesonde values in the troposphere, and the amount of overestimation varied from a few percents to at least 30% depending on the latitude band.

Figure 2 presents comparisons of two modified albedo parameterizations against the operational retrieval for the whole orbit. Other parametrizations were also tested but we show here only the most suitable ones. The largest decrease in tropospheric ozone was found by using a second order polynomial at UV1 channel and a constant

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value at UV2 channel (Fig. 2b). The largest decrease in ozone levels was found close to the poles but near the equator the changes were small. At high latitudes the changes were in the range of 20 %. If a first order polynomial was used for the surface albedo at both channels, the amount of tropospheric ozone increased up to 10% on average, as Fig. 2a shows.

We also studied how the posterior errors were affected when the albedo assumptions were modified. Figure 3 shows the averaged posterior errors (solid lines) and a priori errors (dashed lines) for the studied orbit (06704). The peaks in the a priori error profiles at 3 hPa are caused by the assumption of larger a priori errors for the ozone hole conditions. We found that all three albedo assumptions produced identical posterior errors for pressure levels smaller than 30 hPa. Between 100-30 hPa pressure levels the assumption of linear albedos for both UV bands produced the largest posterior errors while the usage of a second order polynomial at UV1 band and a constant value at UV2 band produced the smallest errors. For pressure levels over 100 hPa the situation is the opposite and the assumption of linear albedos produces the smallest errors and the constant albedo at the UV2 channel the largest. This means that the usage of a constant albedo at the UV2 band decreases the amount of ozone in the troposphere (Fig. 2) but it increases the posterior errors. The situation is the opposite when using linear albedos at both bands.

3.3 Ozone climatologies as a priori information

To constrain the retrievals, the OMO3PR algorithm uses climatological ozone profiles as a priori. In addition to the profiles themselves, a priori information on the variability of ozone at each layer is used. Recently, two new ozone climatologies became available: McPeters and Labow (2012) (henceforth ML), and Sofieva et al. (2014) (henceforth TpO3).

The ML climatology is formed by combining ozone soundings and MLS data. The climatology consists of monthly average ozone profiles and standard deviation for ten

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degree latitude zones with altitudes from 0 to 66 km. A more detailed description of the climatology is given by McPeters and Labouw (2012).

The TpO3 climatology is based on a combination of ozone soundings and SAGE II (McCormick et al., 1989) satellite data. Mean ozone profiles and standard deviations 5 are given for ten degree latitude zones and for each month. The ozone mixing ratio profiles are presented on a 1 km vertical grid. In addition, the profiles are grouped for tropopause heights in 1 km intervals. This is an important addition because variation in the tropopause height is the main driver for variability in climatological ozone values in the upper troposphere and lower stratosphere (Sofieva et al., 2014). This variability increases the a priori errors in this altitude range. The tropopause heights have values between 6 and 17 km but the number and altitude of the tropopause heights varies for different latitude bands and months. In order to have constant dimensions for the a priori ozone look-up table in the OMO3PR algorithm, all latitude and month cases were assigned with 12 tropopause heights. Tropopause height was calculated in the retrieval algorithm using temperature profiles from ECMWF data following the guidelines given by Sofieva et al. (2014). If a calculated tropopause height was outside the range of the climatological tropopause heights, the nearest climatological value was assigned for it or an average of the two closest ones was used.

In order to see how the usage of these climatologies affect the ozone retrievals, we processed two orbits (18 October 2005, orbits 06702 and 06704) using all three climatologies. For the evaluation, every 10th measurement and 10 pixels for each measurement from the middle of the swath were taken into account. In the comparisons we used the new climatologies with the operational a priori covariance matrix and with error covariance matrices calculated from the variance values given in the climatologies. A correlation length of 6 km was used to calculate the non-diagonal elements of the a priori covariance matrix.

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In the first comparison, we only changed the average ozone profiles and used the operational a priori covariance matrix. This combination produced the largest differences at the highest altitudes when compared with the operational retrieval (Fig. 4a). This was expected because the largest changes to the climatology when compared with the LLM climatology are at the top of the profiles. In addition, the amount of ozone increases around 200 hPa.

In the second comparison, we replaced the operational a priori covariance matrix with a climatological one that was calculated from the variance values given in the climatology. For this case, as Fig. 4c shows, the highest altitudes do not change much. At altitudes below 10 hPa the difference with the operational retrieval shows an oscillating behavior that increases towards lower altitudes. This is caused by the larger a priori variance of ozone in the troposphere as can be seen from Fig. 5, which presents the a priori and posterior errors for the retrievals with different ozone climatologies. In the stratosphere the ML a priori errors (Fig. 5, green dashed line) are much smaller than the operational values but for pressure levels over 50 hPa the situation is the opposite. For the posterior errors (solid green line) the situation is similar. This can also be seen from Table 3 which presents the mean a priori and posterior errors for the different retrieval versions. The unweighted mean errors are calculated for the whole profile and for the lower part of the atmosphere in order to highlight the change at the lowest altitudes. Regarding the values for the whole profile, the ML errors are close to the operational ones, however, at the lowest altitudes ML values are significantly larger (a priori $\sim 60\%$ larger, posterior $\sim 35\%$ larger).

3.3.2 TpO3 climatology

For the first comparison, we used operational a priori covariance matrix with the TpO3 climatology. As Fig. 4b shows, the largest differences are seen around 200 hPa, TropO3 giving smaller ozone values than the operational retrieval.

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When the operational a priori covariance matrix was replaced with a climatological version, the differences in the retrieved ozone profiles grew larger, as Fig. 4d shows. The differences around 50 hPa are larger and the profile oscillates more.

When comparing the results from the new climatologies with the operational one, it is important to notice that the new climatologies increase the amount of ozone in the troposphere, except when TpO3 is used with a climatological a priori covariance matrix.

Regarding the errors in the stratosphere, the TpO3 a priori errors (Fig. 5, red dashed line) are much smaller than the operational values but for altitudes below 60 hPa the situation is the opposite. For posterior errors (solid red line) the situation is similar. As Table 3 shows, the errors of TpO3 for the whole column are slightly smaller than the operational ones, however, at the lowest altitudes TpO3 values are significantly larger (a priori ~50 % larger, posterior ~25 % larger). Both climatologies (TpO3 and ML) cause similar peaks in the difference plots between 10–100 hPa when climatological a priori covariances are used (Fig. 4c and d). This is caused by the similar uncertainty values in both climatologies for these altitudes. When compared with the ML errors, TpO3 errors are always smaller, except near the surface.

Visual comparison of difference profiles at different latitude bands with the OMI-MLS validation results presented by Kroon et al. (2011) indicate that the oscillating effects caused by the use of a climatological error covariance matrix might improve the agreement between MLS and OMI ozone profiles.

Based on these results TpO3 appears as the most promising climatology for our purposes. As a next step, we tested how the the albedo parametrizations combined with the TpO3 climatology (with operational a priori error covariance) affected the retrieved profiles. Figure 6 shows three comparisons with the operational retrieval: modified version has linear albedos at both channels (6a), constant albedo at UV2 channel (6b), or linear albedo at UV2 (6c). When compared with the results presented in Fig. 2, the situation changes slightly. With the operational climatology the largest decrease in tropospheric ozone was achieved by using a constant albedo in the UV2 channel,

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whereas, with the TpO3 climatology the largest decrease was achieved with a linear albedo in the UV2 albedo, as can be seen from Fig. 6.

3.4 Effect of modifications on averaging kernels and errors

In addition to the retrieved ozone profiles, OMO3PR algorithm produces several metrics that can be used to assess the retrievals. Averaging kernel is one of them. We compared averaging kernels from different versions of the algorithm to study how the information content was distributed in the retrieved profiles. The degrees of freedom for a retrieval can be calculated as a sum of the diagonal elements of the averaging kernel. For this comparison, we calculated mean averaging kernels from 34 pixels from a region with high tropospheric ozone values (45-55° N, 20-30° E) on 17 July 2007, in order to have sufficient signal also from the lowest altitudes. Figure 7a shows the mean averaging kernel for the operational retrieval. From this plot it is evident that the operational retrieval has very little information from the troposphere. When the a priori ozone profiles are taken from the TpO3 climatology, the averaging kernel is almost identical with the operational one, as Fig. 7b shows. Moreover, the usage of linear albedo in the UV2 channel (with TpO3 climatology) does not have an effect on the averaging kernels (Fig. 7c). When the operational a priori covariance matrix is replaced by a climatological version (based on the variance values from the TpO3 climatology) the averaging kernels change significantly (Fig. 7d). The information content increases in the troposphere and decreases at the highest levels. This is caused by the larger a priori variance values in the troposphere when compared with the operational retrieval which assumes 20% variance there (see Fig. 5). The opposite holds true for the stratosphere. When a minimum value of 20% is set to the variances in the climatological error covariance matrix, the averaging kernels change slightly (Fig. 7e). The degrees of freedom in the troposphere decrease while the degrees of freedom in the stratosphere increase. Figure 7f is the same as 7e but with a minimum variance of 10 %. By comparing these two plots, it is clear that smaller a priori variance values in the stratosphere produce larger degrees of freedom in the troposphere and smaller in

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the a priori covariance matrix. In addition to degrees of freedom, it is important to consider how these different a 5 priori assumptions affect the a priori and posterior errors. Figure 8 presents the errors for the discussed versions of the retrieval (dashed lines for a priori and solid lines for posterior). Figure 8a shows the errors for the operational retrieval and for two versions with TpO3 climatology. The first version (TpO3) has the same albedo assumptions as the operational while for the second one (TpO3 alb) a linear albedo is assumed in the UV2 channel. As the dashed lines show, all the retrievals have the same a priori errors. The usage of the TpO3 climatology produces slightly smaller posterior errors than the operational version at some altitudes and the modified albedo only has a minor effect at the lowest altitude. Figure 8b shows the errors for the retrieval versions with the TpO3 climatology, modified albedo and climatological a priori covariance matrix. For the first version (TpO3 alb covar), the a priori errors are taken directly from the climatological variance values, thus the a priori errors in the stratosphere are small. For the two other versions, minimum a priori errors were assumed to be either 10% (TpO3_alb_covar_10) or 20 % (TpO3_alb_covar_20). As the plot shows, posterior errors are significantly different for these versions. In the troposphere, the posterior errors are larger than for the retrievals with the operational a priori errors. In the stratosphere, smaller a priori errors produce smaller posterior errors which is to be expected. Although the errors look significantly different for the retrievals with operational and climatological a priori error covariance matrices, the relative change between the a priori and posterior errors stay in the same range for all the versions, especially in the troposphere. In the stratosphere, the operational a priori covariance matrix produces slightly larger reduction in the uncertainty. Based on these results it seems that all the retrieval versions are as effective in reducing the uncertainty and that the posterior error values depend mostly on the assumed a priori errors.

the stratosphere. As a whole, Fig. 7 shows shows that the distribution of degrees of freedom for a retrieval depend mainly on the selection of the variance values used in

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Based on the sensitivity tests discussed in this section and our aim to improve the accuracy of the OMO3PR retrievals in the troposphere, we decided to make the following modifications to the algorithm: (1) we replaced the LLM ozone climatology with the TpO3 climatology, (2) we replaced the operational a priori covariance matrix with a climatological version that is based on the variance from the TpO3 climatology (with min. 10% standard deviation, separated tropo- and stratosphere), and (3) we replaced the second order polynomial for the albedo in the UV2 channel with a first order polynomial. The tropo- and stratosphere were separated in the climatological error covariance matrix, although, our sensitivity studies (not shown) did not show visible effects in ozone abundances caused by this modification. In addition, we updated the retrieval's vertical pressure grid to $P_i = 2^{-i \cdot 1.37/2} \times 1000$ for i = 0.18 which follows the principles presented by Liu et al. (2010) without changing the number of layers. We also tested how sensitive the retrieval is to the selection of the correlation length. The operational retrieval uses correlation length of 6 km, thus, we tested how the retrieved ozone abundances change if correlation lengths of 1 km, 3 km or 12 km are used instead. Our comparisons showed that only the selection of 1 km as correlation length had a significant effect on the retrieved ozone profiles. This selection increased oscillations in the profiles. Based on these results we decided to use the operational correlation length.

Evaluation of the modified algorithm

In order to see if the modifications to the algorithm improved the correspondence of the tropospheric ozone profile retrievals with other methods, we compared our results with the results published by Eremenko et al. (2008). The modified version of the algorithm uses a different pressure grid than the operational one, thus we had to interpolate the profiles in order to compare values for the exactly same altitude range. This was done by calculating a cumulative ozone profile in Dobson units from the top of the atmosphere down to the bottom of each layer and interpolating it to keep the total ozone amount in the column constant. Then, the tropospheric ozone abundance was taken

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from the 400 hPa level which corresponds to the altitude of 6 km used in the analysis by Eremenko et al. (2008). For the comparison, we calculated the ozone profiles first in Dobson units from the cumulative profile and then transferred the layer values into mixing ratios with the following equation: $vmr = dobs/(pres_{bottom} - pres_{top}) \cdot 1.26720 \times 10^{-6}$, where dobs is the ozone abundance of a layer in dobson units, press_{bottom} is the pressure at the bottom of the layer, and pres_{top} is the pressure at the top of the layer. The constant is required to transfer the dobson units into volume mixing ratios.

Eremenko et al. (2008) compared tropospheric ozone column retrievals from IASI retrievals with predicted values from the CHIMERE model for three days in July 2007. The IASI ozone profile retrievals are done with an analytical altitude-dependent regularization method (Eremenko et al., 2008). We processed the same dates with different versions of the OMO3PR algorithm (using every second measurement and retrievals with cloud fraction < 0.3) to study how modifications in the algorithm affected the retrieved tropospheric ozone abundances. Here, we concentrate on results from a single day, 17 July 2007, because during this day high tropospheric ozone values occurred in Eastern Europe. Before comparing the modified OMO3PR retrievals with the IASI results, we studied how the modifications of the algorithm affected the spatial distribution of tropospheric ozone for this day. Figure 9 shows the difference between the operational retrieval and the modified versions in percents. The averaged values for the studied region are given in Table 4. The usage of the TpO3 climatology (Fig. 9a) decreases the amount of tropospheric ozone at Bay of Biscay and increases it in Eastern Europe. The usage of a linear albedo at the UV2 channel does not have as large effect as the climatology (Fig. 9b). However, decrease in ozone abundance can be seen in Northern Europe. The usage of a climatological a priori covariance matrix with a minimum variance of 10% (Fig. 9c) decreased the amount of tropospheric ozone everywhere. These results highlight the fact that changes in the tropospheric ozone abundances caused by one modification can be canceled out by another modification. On average, the usage of the TpO3 climatology decreased the tropospheric ozone abundance by 0.6 % while the albedo modification decreased it slightly more, by 3.3 %.

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Whereas, the usage of climatological a priori covariance matrix decreased the ozone abundance by 11.3% (Table 4).

As a next step, we compared all these versions of the algorithm to the IASI retrieval for the same day, as presented in Fig. 10 and in Table 4. First, we did the compar-₅ ison with the operational retrieval (Fig. 10a). The operational version overestimates the amount of ozone at the outskirts of Europe and slightly underestimates in Eastern Europe where IASI detected the high values. The usage of the TpO3 climatology (Fig. 10b) improves the correspondence in Western Europe while changing the small underestimation in Eastern Europe into a slight overestimation, thus, the new climatology did not improve the agreement for the highest ozone values in Eastern Europe. Then, we changed the albedo assumptions so that a linear albedo, instead of a second order polynomial, was used in the UV2 channel (Fig. 10c). Based on the plot, this modification does not clearly improve the agreement, although, the averaged values in Table 4 indicate some improvement (from overestimation of 23% to 19%). As a last step, we replaced the a priori covariance matrix with the climatological version (Fig. 10d). The usage of the new error covariance matrix clearly improves the agreement with IASI all over Europe (overestimation down to 12%) except for the highest ozone values in Eastern Europe. This implies that the better correspondence with IASI was achieved by decreasing ozone abundances in regions with low ozone values and that the OMO3PR retrieval is not able to capture the high ozone values as well as IASI even after a number of modifications.

5 Conclusions

In order to find ways to improve the retrieval of tropospheric ozone from OMI measurements, we assessed the sensitivity of the OMO3PR algorithm to several a priori assumptions. The studied assumptions were: stray light correction, surface albedo parametrization and a priori ozone climatologies. We found that stray light correction is essential for the retrieval but it mainly affects the stratospheric layers. Surface albedo

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parametrization also has a significant effect on the retrieved ozone profile but mainly in the layers close to the surface. The selection of the a priori ozone profile climatology had a relatively small effect on the retrieved ozone profiles while the usage of climatological variance values in the a priori covariance matrix increased the differences significantly. Based on these sensitivity studies we modified the OMO3PR algorithm to improve the accuracy of tropospheric ozone retrievals. This was done by replacing the operational ozone climatology (LLM) with TpO3 climatology, by using climatological variance values in the a priori covariance matrix and by changing the wavelength dependency of surface albedo in the UV2 channel from a second order polynomial to linear. Our studies showed:

- 1. a priori covariance matrix basically defines the vertical distribution of degrees of freedom for a retrieval.
- 2. We were able to increase degrees of freedom significantly in the troposphere by increasing a priori errors in the troposphere and lower stratosphere, and by decreasing them at higher altitudes.
- 3. All the studied versions of the OMO3PR algorithm were equally effective in reducing uncertainty in the retrieved ozone profiles. This implies that the posterior error values depend mostly on the chosen a priori errors.
- 4. When compared with the IASI measurements presented by Eremenko et al. (2008) the new version produced over 10 % smaller ozone abundances in the troposphere over Europe which reduces the systematic overestimation of the OMO3PR algorithm.

Acknowledgements. The Dutch-Finnish built Ozone Monitoring Instrument (OMI) is part of the National Aeronautics and Space Administration (NASA) Earth Observing System (EOS) Aura satellite payload. The OMI project is managed by the Netherlands Space Office (NSO) and the Royal Netherlands Meteorological Institute (KNMI). OMI vertical ozone profile data were processed at and obtained from the NASA Goddard Earth Sciences (GES) Data and Information Services Center (DISC). We thank Maxim Eremenko for providing us with the IASI data.

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Table 1. The agreement between OMO3PR and other ozone profile observations in percents. Summarized from Kroon et al. (2011). The instruments used in the study are the Microwave Limb Sounder (MLS), the Tropospheric Emission Spectrometer (TES), the Stratospheric Aerosol and Gas Experiment (SAGE II; McCormick et al., 1989), the Halogen Occultation Experiment (HALOE; Russell et al., 1993), the Optical Spectrograph and Infrared Imager System (OSIRIS; Llewellyn et al., 2004), the Global Ozone Monitoring by the Occultation of Stars (GOMOS; Bertaux et al., 2010) and balloon-borne ozonesondes (ECC).

	Tropical	Mid-latitude	Polar
OMI – MLS [%]	±10	±10	±30
OMI – TES [%]	±20	±30	±60
OMI – SAGE II [%]	10-60	20-40	
OMI – HALOE [%]	> 30	> 30	
OMI – OSIRIS/GOMOS [%]		±10	-15-20
OMI – ECC [%]	5–80	5–60	10–20

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Table 2. Content of the Optimal estimation state vector and the related a priori information used in the OMO3PR retrieval.

Optimal estimation state vector		a priori information	a priori error	
Species	Number of elements			
O ₃ profile	18	LLM climatology (McPeters et al., 2007)	20% (For ozonehole conditions 60% (21–50 km) and 30% for other altitudes.)	
Surface albedo (surface or cloud)	6	OMI surface albedo climatology (Kleipool et al., 2008)	100%	
Stray light	6	0	100 %	

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Table 3. Mean a priori and posterior errors in percents for three retrievals: operational, with ML ozone climatology and with TpO3 climatology. Mean unweighted errors are presented for the whole profile and for lower atmosphere (> 100 hPa). The values are calculated from the data presented in Fig. 5.

a priori	Operational	ML	ТрО3
Whole column	19.8 %	20.6 %	16.6 %
Lower atmosphere (> 100 hPa)	19.5 %	30.7 %	28.8 %
posterior			
Whole column Lower atmosphere (> 100 hPa)	10.5 %	12.0 %	10.0 %
	15.5 %	21.0 %	19.2 %

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Table 4. Differences in tropospheric (up to 400 hPa ~ 6 km) ozone abundances between IASI and OMI retrievals for 17 July 2007 in Europe. Four different versions of the algorithm are used: operational is the operational version, TpO3 refers to a version that uses the TpO3 climatology, TpO3_alb refers to a version that uses the TpO3 climatology and linear albedo in the UV2 channel, and TpO3_alb_covar_10 refers to a version that uses the TpO3 climatology, linear albedo in the UV2 channel and a climatological error covariance matrix. Average difference in Dobson units (Ave diff), average relative difference (Ave rel diff) and average standard deviation (Ave std) are presented. In addition, the difference between the operational OMI retrieval and the other versions (Ave rel diff with ope) in percents are given.

17 Jul 2007	operational	ТрО3	TpO3_alb	TpO3_alb_covar_10
Ave diff [DU]	-5.06	-5.19	-4.32	-2.72
Ave rel diff	-0.23	-0.23	-0.19	-0.12
Ave std	4.50	3.57	3.59	3.79
Ave rel diff with ope [%]		0.6	4.7	11.3

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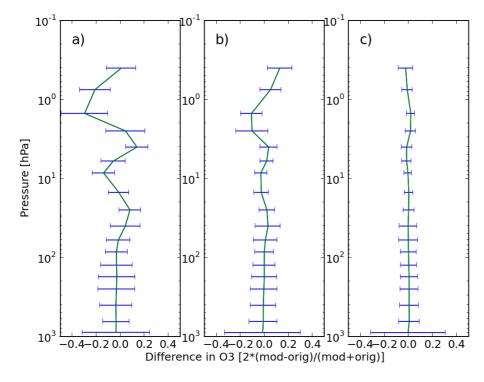


Fig. 1. Effect of stray light corrections. Change in the ozone profiles when the retrieval has no stray light correction (a), only L1 correction (b), and only L2 correction (c). Error bars show the standard deviation of the difference.





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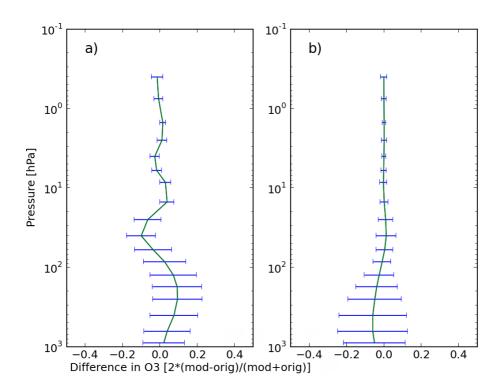


Fig. 2. Effect of albedo assumptions. (a) assuming linear albedos at both UV1 and UV2 channels instead of second order polynomials. (b) assuming constant albedo at UV2 channel instead of second order polynomial. Error bars show the standard deviation of the difference.

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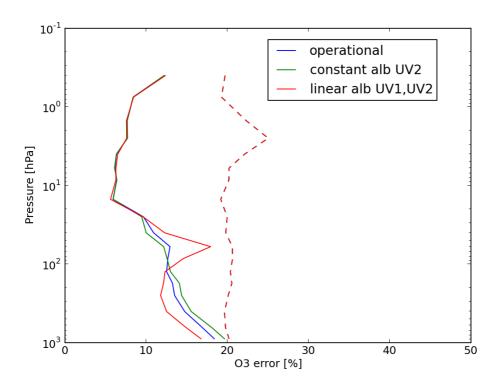


Fig. 3. Effect of albedo assumptions on posterior errors (solid lines). Operational retrieval is shown in blue, linear albedos at both UV1 and UV2 channels instead of second order polynomials shown in red, and constant albedo at UV2 channel instead of second order polynomial is shown in green. Dashed lines show the a priori errors.

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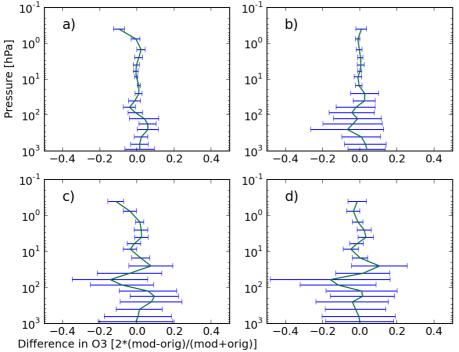


Fig. 4. Effect of a priori ozone climatologies. Change in ozone profiles when ML (a) and TropO3 (b) are used with the operational error covariance matrices. Change in ozone profiles when ML (c) and TropO3 (d) are used with climatological covariance matrices. Error bars show the standard deviation of the difference.

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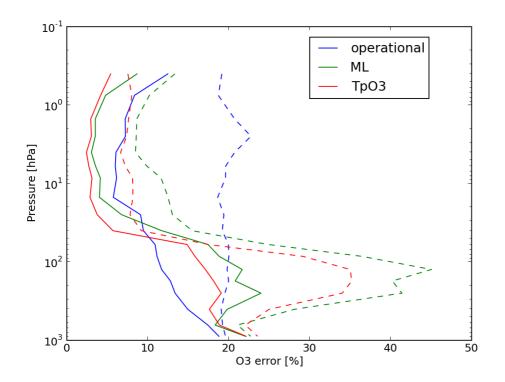


Fig. 5. Effect of ozone climatologies on a priori (dashed lines) and posterior errors (solid lines). Operational retrieval is shown in blue, ML climatology in green and TpO3 climatology in red. The a priori errors for ML and TpO3 climatologies are based on the ozone variability reported in the respective climatologies.

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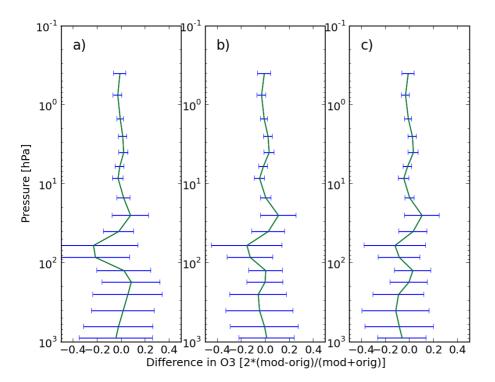


Fig. 6. Effect of albedo assumptions with the TropO3 climatology. **(a)** assuming linear albedos at both UV1 and UV2 channels instead of second order polynomials. **(b)** assuming constant albedo at UV2 channel. **(c)** assuming linear albedo at UV2 channel. Error bars show the standard deviation of the difference.

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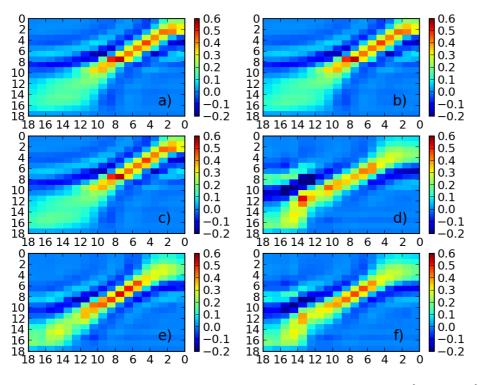


Fig. 7. Mean averaging kernels from 34 pixels on the 17 July 2007 (45-55° N, 20-30° E) for the operational retrieval (a), with the TpO3 climatology (b), with TpO3 and linear albedo in UV2 channel (c), with TpO3, linear albedo in the UV2 channel and a climatological error covariance matrix (d), with a climatological error covariance matrix and minimum of 20% variance (e) and with a climatological error covariance matrix and minimum of 10% variance (TpO3 alb covar 10), (f). Axis are the layer indexes, 18 being the layer closest to the surface.

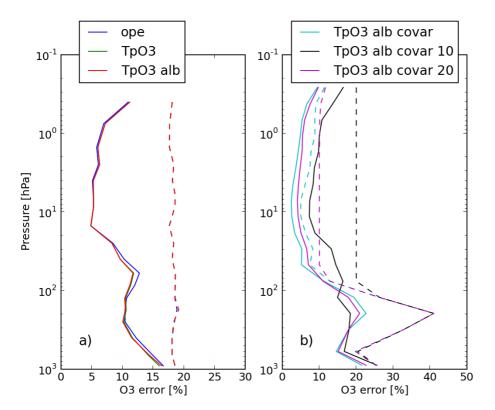


Fig. 8. Effect of a priori covariance matrix assumptions on a priori (dashed lines) and posterior errors (solid lines). Operational retrieval is shown in blue, TpO3 climatology in green, TpO3 with linear UV2 albedo in red (a), TpO3 with climatological a priori errors in cyan, TpO3 with climatological a priori errors with minimum of 20 % in black and TpO3 with climatological a priori errors with minimum of 10% in magenta (b).

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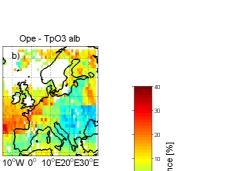


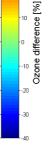


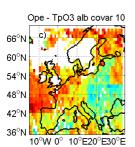
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Ope - TpO3

10°W 0° 10°F20°F30°F

66°N **a**)

60°N

54°N

48°N

42°N

36°N

Fig. 9. Difference in tropospheric ozone abundances (up to 400 hPa) between operational (ope) and modified OMO3PR retrievals on 17 July 2007. Modified versions of the OMI retrieval algorithm are: TropO3 climatology and operational albedo **(a)**, TropO3 climatology and linear albedo in UV2 **(b)**, and TropO3 climatology, linear albedo in UV2 and climatological error covariance matrix **(c)**. Daily data averaged on 1 × 1 grid. Averaged differences are given in Table 4.

66°N

60°N

54°N

48°N

42°N

36°N

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7, 1835-1869, 2014

Sensitivity of the OMI ozone profile retrieval (OMO3PR) to a priori assumptions

AMTD

T. Mielonen et al.



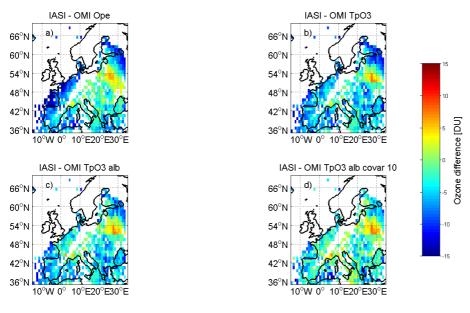


Fig. 10. Difference in tropospheric ozone abundances (up to 400 hPa) between IASI and OMI on 17 July 2007. Four different versions of the OMI retrieval algorithm are used: operational (a), TropO3 climatology and operational albedo (b), TropO3 climatology and linear albedo in UV2 (c), and TropO3 climatology, linear albedo in UV2 and climatological error covariance matrix (d). Daily data averaged on 1 × 1 grid. Statistics of the differences are given in Table 4.