Response to Reviewer #1:

Review

First of all, we thank the referee for his positive general comments about the paper. We acknowledge him for his useful corrections and suggestions, which have helped us clarifying several points and improving the manuscript.

Below we provide our point-by-point responses to his individual comments. Before each response, the reviewer comments have been quoted between []. Corresponding information and corrections have been added to the revised version of the manuscript. Technical comments are also included in the revised version.

General Comments:

[1) The paper also discusses the use of daily vs. monthly ozone median averages in the trend analysis. This is a less frequently used approach. It has its positive and negative sides for understanding short and long-term variability in time series. The advantage of using daily median ozone values in the upper stratosphere makes sense as there is a physical process that relate Solar flux (SF) and ozone variability on the daily bases, but it cannot be clearly separated in layers below upper stratosphere. It will be good to have discussion on significance of the daily vs monthly SF contribution to the trend analyses for all layers (section 4.3.1 discusses only upper stratosphere layer trends).]

We thank the referee for pointing out this missing aspect of our analysis. It is true that most of the solar cycle variation of ozone occurs in the stratosphere, even in the lowermost stratosphere (e.g. Soukharev and Hood, 2006). However, the use of daily median ozone values in the layers below (UTLS and MLT) is justified in our analysis by the coarse vertical resolution of IASI (full-width at half-maximum of the averaging kernels) which is such that upper and lower atmospheric levels contribute to each other.

As expected, the use of daily medians mainly helps in reducing the uncertainty associated with the trends (i.e. in discriminating between a linear trend and the solar flux effect) in the UST column where the ozone hole recovery is clearly identified, but it also reduces the uncertainty in the lower layers (cfr Table 2 of the manuscript), principally in the MLST and the UTLS where the solar cycle is an important driver (cfr Figure 8 of the manuscript). This is now specifically mentioned in Sections 4.3.2 of the revised version:

"We show that the daily and monthly trends in all layers and all latitude bands fall within each other uncertainties, but that the use of daily median strongly helps in reducing everywhere the uncertainty associated with the trends for the reasons discussed above (Section 4.3.1). This is particularly observed in the UST where the ozone hole recovery has been identified from previous studies, but also in the MLST and the UTLS where the solar cycle variation of ozone is the largest (see Figure 8). As a consequence, …"

This result is important as it tends to indicate that daily data should be preferred to monthly data for deriving significant trends. This gives in our opinion convincing evidence of the benefit of IASI in terms of frequency sampling for the assessment of O_3 trends.

[2) The paper proposed the use of daily data for separation of the Solar signal from the trend contained in the 6-years long time series, but it is not clear from the text that it improves the model fit in all layers and latitude bands (i.e. residuals). This should be discussed in more details in the paper, including showing results in other than US layers.]

In the example provided in Figure 9 of the manuscript (UST in 30°S-50°S), we show that the daily data considerably improves the regression model in terms of residuals (44% in daily *vs* 60% in monthly data) and of trend uncertainty (1.74±0.77 in daily vs1.21±1.30 in monthly data). This translates to larger relative differences between the regression with and without the linear term in daily data (17%) than in monthly data (10%), which indicates a larger compensation effect in the latter. The averaged relative residuals are now indicated in the middle panels in the revised Figure 9, which also illustrates the differences between the two regression models (with and without the linear trend term), as suggested by the referee in one of the technical comments. The offset between the two models is observed for most of layers and latitudinal bands particularly where either O_3 recovery or solar effect signal is important, which is consistent with the decrease in trend uncertainty.

We present in Figure 1 below (same as Figure 9 of the manuscript) one additional example (MLST in 30°S-50°S) characterized by a large significant negative trend in both daily and monthly data (see Table 2 of the manuscript) with a large offset between the two regression models. With this example, we show that even if both residuals and trends are similar in daily and monthly data (-2.17±0.58DU/Yr in daily data vs -2.36±1.80 DU/Yr in monthly data, see Table 2 of the manuscript), the higher co-linearity of the linear and the solar flux terms in monthly data in comparisons with daily data translates to a much larger trend uncertainty (a factor of ~3 in this example).

This is now better explained in Sections 4.3.1 of the revised version:
- "This effective co-linearity of the linear and the monthly solar flux terms translates to larger model fit residuals (44% in daily averages *vs* 60% in monthly averages), to larger relative differences between the two regression models (with and without the linear term) (17% in daily vs 10% in monthly data), and to larger uncertainty on the trend coefficients when using the monthly data in comparison with the daily data."
- "The same conclusions can be drawn from the fits in other layers and latitude bands, especially those where the solar cycle variation of ozone is largest (MLST and UTLS) or where the ozone recovery occurs (UST). A larger trend uncertainty associated with monthly data *vs* daily data is found in all situations (see Table 2, Section 4.3.2)."

[3) One note, the "US" abbreviation for upper stratosphere in the text was confusing to me, as it is typically used for geographical domain of the United States. I would have preferred to have the "UST" abbreviation. "MLS" is also an acronym commonly used for the satellite (Microwave Limb Sounder) ozone data, it therefore it would be better to change it to "MLST".]
We thank the referee for pointing that out. These acronyms have been changed to UST and MLST throughout the revised version of the manuscript.

Specific comments:

[1) P. 12, lines 260-261. Can you please provide more details on how the correction for the autocorrelation is applied to uncertainties of the fit?]

We calculated the uncertainty of the fitted parameters by computing the standard error with an effective sample size (n*) of independent information based on the lag-1 autocorrelation coefficient correlation of the noise residual ($n^* = n \cdot \frac{1-\Phi}{1+\Phi}$) as in Santer al. (2000).This is now better explained in Section 3.1 of the revised version:

"The constant term (Cst) and the coefficients a_n, b_n, x_j are estimated by least-squares method and their standard errors (σ_e) are calculated from the covariance matrix of the coefficients and corrected to take into account the uncertainty due to the autocorrelation of the noise residuals as discussed in Santer et al. (2000) and references therein:

$$\sigma_e^2 = (Y^T Y)^{-1} \cdot \frac{\sum_t [O_3(t) - yY(t)]^2}{n-m} \cdot \frac{1+\Phi}{1-\Phi} \qquad (3)$$

Where Y is the matrix with the covariates ($trend, \cos(n\omega t), \sin(n\omega t), X_{norm,j}$) sorted column wise, y is the vector of the regression coefficients corresponding to the columns of Y, n is the number of daily (or monthly) data points in the time series, m is the number of fitted parameters, and Φ, the lag-1 autocorrelation of the residuals."

[2) P.14, lines 301-303. It is clear from the paper that the IASI has information in the MLT layer, which is between surface and _ 8 km. On the other hand, IASI sensitivity to ozone variability below 4 km is not clearly discussed. Figure 4 suggests 20-40 % total error of the retrieval at the bottom of each of 3 plots for different atitude bands. Figure 5 shows that about 20-40 % ozone variability is observed in the lowest 4 km, with the exception of tropical region. AKs for 0-4 km altitude likely have large contribution from layers above. Is it possible to discern actual day-to-day ozone variability below 4 km and trend that is above the retrieval noise? The information on the AP contribution in MLT (similar to the Figure 2 discussion) can be discussed in this section to help with the sensitivity assessment. This section needs to expand the discussion on information in the MLT.]

The referee is right; AKs below 4 km altitude suggest a large contribution from the upper layers. Based on AKs profile shapes, one should generally better not consider analyzing the ground-300hPa tropospheric column separately in sub-layers since each of them contributes to each other, nor analyzing the lowermost troposphere because of the sharp decrease of sensitivity down to the surface which is inherent to nadir thermal IR sounding in cases of low surface temperature or low thermal contrast (see Figure 4 of the manuscript). As a result, the variability can hardly be discussed independently below 4 km and this is why no trends were given for the lowermost troposphere.

However, one exception is found in spring-summer 30°N-50°N latitude band where the detected variability below 4 km (between ~30% and ~45%, see Figure 5 of the manuscript) is larger than the retrieval error (lower than 25%, see Figure 4 (b) of the manuscript). As mentioned in the paper, this variability could potentially be linked to photochemical production of O_3 associated with anthropogenic precursor emissions. The a priori contribution in the ground-700hPa column, as suggested from Figure 2 of the manuscript for the ground-300hPa column, is the lowest in this region and during that period. It has been estimated to 10-20% over the continental regions.

This is now better explained in both the revised Section 2 and Section 4.1, and some words of caution about the detectable ozone variability in the lower troposphere have been added as well.

[3) P.14 lines 314-315. Please clarify the statement "The fact that the patterns are similar in _10 km mainly reflects the low sensitivity of IASI to that level compared to the others." This is in regards to Figure 6. It would be good to explain a bit more about the patterns. Otherwise reader is left to guess if it is about seemingly no variability in the tropics (blue color indicates low concentrations), or similarity to results at 20 km, or something else. Figure 5 shows high relative ozone variability at 10 km level, but the range in absolute ozone concentrations might be small.]
This sentence has been changed to "The fact that the patterns in ~10km are similar to those in ~20 km mainly reflects the low sensitivity of IASI to that level compared to the others"

[4) P. 21,
a) lines 452-456, statement that ". . .linear term is not compensated by solar flax in daily averages" is not completely true, because the SF fitted signal from the model with and without the liner term (blue and orange lines shown in the bottom left panel of Figure 9) are not exactly the same (positive and negative coefficients).
b) Also, the difference between the orange and blue SF signal can be fitted with the linear slope.
c) Besides Figure 8, it will be useful to have a tabulated summary of the variables in the statistical model that were kept after iterative backward selection, and fitting uncertainties for all layers and latitude bands. Otherwise it is hard to get these numbers from the figure. It can be added in the Supplemental materials.]
a) The sentence has been corrected.
b) Exactly. This results from the exclusion of the linear term trend in the regression model. This is precisely what we expect from using daily data and daily solar signature instead of monthly ones: the offset when using daily data corresponds well to a trend over the IASI period. It results from the fact that, in daily data, the solar flux cannot completely compensate the linear trend (LT) term in the regression model because of its strong daily signature, while it largely compensates the LT in monthly data. See responses to comment 5a) and b) below. We now better explain Figure 9 in the revised Section 4.3.1.
c) We now provide in the revised version of the Supplementary Materials and here below, the Table S1 which summarizes the proxies retained in the stepwise backward elimination approach that are significant at the 95% level for each latitude band and for each partial column. Summarizing in one Table the fitted uncertainties for each retained fitted parameters, each latitude band and each layer is difficult and we have preferred to keep Figure 8 as it is. Nevertheless, to help the readers, the proxies which become statistically non-significant when accounting for the autocorrelation in the noise residuals at the end of the elimination process (with an uncertainty larger than its associated estimate; i.e. larger than 100% corresponding to an error bar overlapping the zero line) are indicated between parentheses in Table S1. This has been now mentioned in the revised version in Section 4.2.

[5) Additional Figure 9 comments:

a) The information in the middle panel is not very clear. It is stated that the deseasonalized IASI ozone data are plotted. Can you please explain the process of deseasonalization for data, such as how the seasonal cycle was derived – from data averages or from the model fit?
b) Whereas the model fit with the linear term included (light blue line) seems to follow the de-seasonalized IASI ozone data (dark blue), the model fit without the linear term (orange) is clearly low-biased from the data (dark blue line). It is not clear how the model fit can be done with the resulting mean offset from the data. Is it possible that the wrong constant term is used to calculate the model time series (orange) for this plot. My understanding of the discussion is that two separate models were used to obtain the data fit: one is with (blue) and another one is without (orange) the linear term. Please make corrections to the text if the single model is used, but the model result is plotted with and without the linear term.
c) On the other hand, in the case of the model fit without the linear term the SF signal contribution to the model fit for monthly mean data is much larger as compared to SF term in the daily data fit model. Is it due to the fact that solar flux seems to increase from 2008 to 2013, and for the analyzed time period seems to be comprise of a liner trend and the day-to-day variability that has significantly increased by 2011?]
a) Deseasonalized IASI data were obtained by subtracting from the IASI ozone time series the seasonal cycle derived from the model fit. This has been now mentioned in the revised version.
b) and c) See responses to general comment 2).
As discussed in Section 4.3.1, two separate models have indeed been used, one with and one without the linear term. The offset (which is actually not constant but increasing over the time period, i.e. representative of a trend) between the both models precisely results from the fact that, in daily data, the solar flux cannot completely compensate the linear term trend because of its strong daily signature. In monthly data, the solar flux and linear trend terms are less distinguishable and we observe a much larger compensation effect: the SF signal is indeed much larger and the offset is consequently not as high as in daily data. This is now better explained in Section 4.3.1 of the revised version.

[6) p.22, lines 486-488. When comparing to the previous publications of the trend analysis, please mention the difference in the time period analyzed. I would replace “in agreement with previous studies” with “comparable to the results published in the previous studies”]
Thanks for pointing that out. It has been changed.

[7) p. 22,line 497 “change ‘was’ to ‘were’] It has been changed.

[8) p.23, line 506, change ‘conducting’ to “leading”] It has been changed.

[9) p.23, line 507-508, add at the end of the sentence “in winter (Table 3)”. Remove the next sentence.] It has been changed.

[10) P. 23, lines 508-510 add “NH” after “in summer”, and “SH” after “in winter”.] It has been changed.

[11) P. 23 lines 511-512. The discussion of the effects of the upper stratosphere temperature trends is important for the trend analysis. Can you please comment on the correlations between daily ozone and Solar flux, ozone and temperature, and possibility to discern temperature contribution to ozone variability from Solar flux in upper layers.]

Previous studies have shown that the various chemical production and loss mechanisms respond to the annual cycles of temperature and of different trace gases (i.e. stratospheric temperature is the main driver of ozone loss within the polar vortex, and this chemical destruction further favours low total ozone and thus less ozone radiative heating and lower stratospheric temperatures) and that all these effects are correlated: Temperature changes are linked to changes in the frequency of stratospheric warmings (e.g. due to QBO-induced secondary circulation, decreasing CO_2 cooling,…); Solar cycle plays a very clear influence on both ozone and stratospheric temperatures variations that are also correlated with the QBO. Please refer for example to Steinbrecht et al. (ACP, 2006) which reported results from a multiple linear regression analysis of both long-term total ozone TOMS observations and long-term temperature reanalyses, accounting for the 11-year solar cycle and QBO effects amongst others.

As mentioned throughout the manuscript, the complexity of the dynamical and chemical processes makes it difficult to unambiguously define simple and independant predictor in a statistical model (e.g. Mäder et al., 2007; Harris et al., 2008). We now mention in the conclusions that effects of changing stratospheric temperatures as well as changes in the Brewer-Dobson circulation should be investigated in a further study.

[12) P.25, lines 553-556.
a) This section discusses the MLT layer (ground-300 hPa). Please clarify what is meant by "As for the upper layers, ...". It is possible that the subject of the discussion has changed, and then it would be better to have a new paragraph.
b) Also, Tables 2 and 3 show negative trend in the IASI MLT layer , but it is stated here that it is in agreement with increases in ozone found in Arctic (Kivi et al, 2007) following changes in Arctic Oscillation. This statement needs further explanation how the negative ozone trend is related to the Arctic Oscillation during 2008-2013 time period.
c) Table 3 title has missing information about the second row of trend results. Please add after daily " (top) and monthly (bottom)", similar to the title in Table 2.]
a) and b) We thank the referee for pointing that mistake. This sentence has been deleted in the revised version.
c) "Same as Table 2" has been indicated in the title instead of repeating the description of Table 2 to shorten the Table 3 title.

[Supplemental material: The discussion on the tropospheric ozone variability (MLT) is largely concerned with the stratospheric origin of the tropospheric ozone which is tracked by means of the difference between total and ozone tagged by modeled NOx tracer (Figures S2 and S3). And this is a wonderful addition to the data analysis. However, the reader would also like to understand the contribution of the stratospheric ozone due to the shape of the AK, which is not discussed at all. It should be possible to assess this retrieval error by using truncated AK (zero weights for stratospheric ozone) for smoothing MOZART -4 profiles and then comparing it to the full IASI AK smoothed profiles.]
As suggested by referee #3, we now illustrate in the revised Supplement (Figure S4(a)) the fit of MOZART-4 O_3 and of $O_3^{tagged\_NOx}$ time series, in addition to the stratospheric contribution (Figure S4(b)), without accounting for the IASI sensitivity, to evaluate the effect of the smoothing error from the observational system. We prefer to adopt this approach instead of truncating the AK in the stratosphere, since residual stratospheric contributions will still be

reflected in AK from lower layers. Note also that the smoothing error $[(A-I)Sa(A-I)^T]$ can be evaluated from the a priori contribution [Xa- (AXa)] provided in Figure S5(b) since they are both correlated; i.e., if the IASI sensitivity is low in the MLT, the smoothing error will be large as well as the contributions from the a priori and from the upper layers. When comparing Fig. S4(b) and Fig.S5(b), the differences suggest that the limited vertical sensitivity of IASI contributes a smaller part (~10%-20%) to the IASI stratospheric contribution than the natural stratospheric influence (~20% to 45%). In addition, the contribution of the real natural variability (originating from both the troposphere and the stratosphere through STE processes) into the MLT O_3 columns is also now illustrated in an additional figure (Fig. S6(a) and Fig.2 here below) and is estimated to be larger than 50% everywhere. For example, we interestingly show that in the 30N-50N band where the DOFS is the largest (See Fig.2(b)), this contribution reaches ~85% from which only ~30% originate from the stratosphere (Fig. S4(b)) and ~55% from the troposphere (Fig. S6(b)). This is now specifically mentioned in the last paragraph of Section 2 of the revised manuscript and in the Supplementary materials.

This further supports the findings that IASI is able to detect a large part of the real variability of O_3 in the N.H. troposphere, and that the increase in the observed concentrations and variability in the mid-latitudes N.H. during spring-summer likely indicate a photochemical production of O_3 associated with anthropogenic precursor emissions (Cfr. Section 4.1 of the manuscript).

Technical comments:

[Figure 1 – add a few minor ticks to the altitude axes]
It has been added.

[Figure 5 – “1*sigma” – is it correct expression, or it should be defined as sigma/ (median ozone value)*100?]
It should indeed be defined as [1σ(daily median O_3)/(daily median O_3)*100]. This has been corrected.

[Figure 9. It would be better to separate middle panel into two – for the model fit with and without the linear term. It would then allow for space in the plot to show the residual for both fits separately.]
We prefer to keep the middle panel as it is to more easily compare the regression models with the linear term trend included or not in the regression model and to highlight the increasing offset between the both models. But as suggested by the referee, we have added in the revised Figure 9 a panel illustrating the difference between the two regression models and the averaged relative residuals (%) have been indicated as well.

Table 1 List of the proxies retained in the stepwise backward elimination approach which are significant at the 95% level (see text for details) for each 20-degree latitude bands and for each partial column. Proxies are indicated for Solar flux (blue), QBO10 (green), QBO30 (orange), ENSO (red) and NAO (pink)/AAO (purple). Symbols indicated between parentheses refer to proxies which are not significant statistically when accounting for the autocorrelation in the noise residuals.

Proxies	**Ground-300hPa (Troposphere)**	**300-150hPa (UTLS)**	**150-25hPa (MLST)**	**25-3hPa (UST)**	**Total columns**
70°N-90°N	**(O) (O) O O**	**O(O) O (O)**	**O (O) (O)O O**	**(O)O (O) O**	**O (O) O O O**
50°N-70°N	**O (O) (O) O (O)**	**O O (O) O**	**O (O) (O)O O**	**(O)(O)**	**O (O) (O)O O**
30°N-50°N	**(O) (O) (O)O**	**O(O)(O)O**	**O (O) O O(O)**	**O (O) (O)(O)**	**O (O) (O)O O**
10°N-30°N	**(O) (O) (O)**	**(O)O(O) O (O)**	**(O)(O)(O)O O**	**O (O) (O)**	**O (O) O O**
10°S-10°N	**(O) O (O) (O)(O)**	**O O O O O**	**(O) OO(O)(O)**	**O O O**	**(O) O O O(O)(O)**
30°S-10°S	**(O) (O)(O) (O)**	**(O)O(O) O (O)**	**O (O) O O (O)**	**(O)O O O (O)**	**(O) (O) O O O**
50°S-30°S	**(O) (O)(O) O (O)**	**(O)O(O) O O**	**O O O (O)**	**(O)O O O (O)**	**(O) (O) O O (O)**
70°S-50°S	**O (O) (O)**	**(O)O (O) O**	**(O)(O) O (O) O**	**(O)O O (O)**	**(O) (O) O O O**
90°S-70°S	**(O) O O**	**(O) O(O) O**	**(O)(O)(O)(O)(O)**	**O(O) O**	**(O)(O)(O)(O)(O)**

Figure Caption

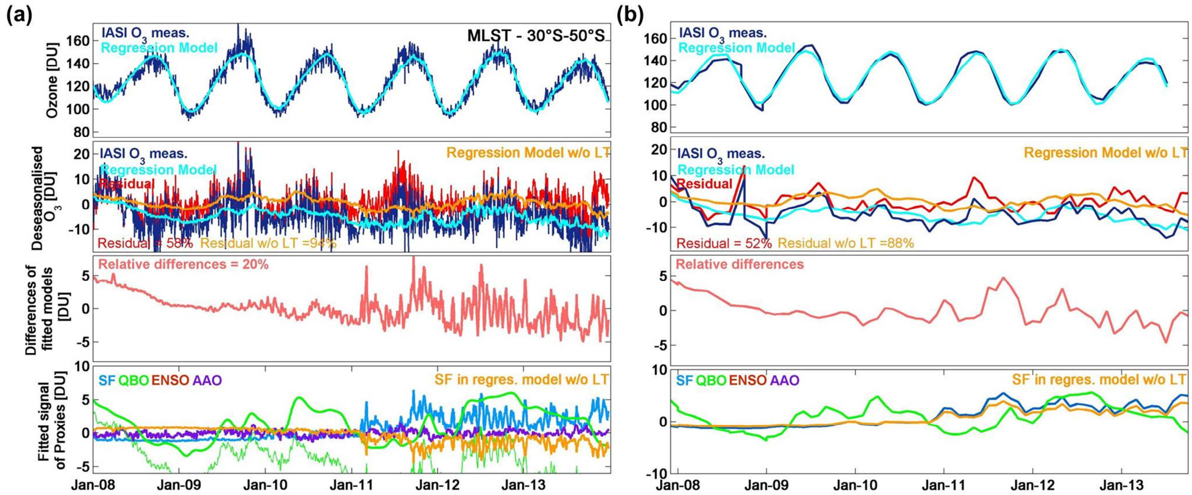

Figure 1. Daily (a) and monthly (b) time series of O_3 measurements and of the fitted regression model in the UST in the 30°S-50°S latitude band (top row), of the deseasonalised O_3 (2^d row), of the difference of the fitted models with and without the linear term (3^d row), and of the fitted signal of proxies ([regression coefficients*Proxy]): SF (blue), QBO (QBO^{10} + QBO^{30}; green), ENSO (red) and AAO (purple) (bottom) (given in DU). The averaged relative residuals are also indicated (%).

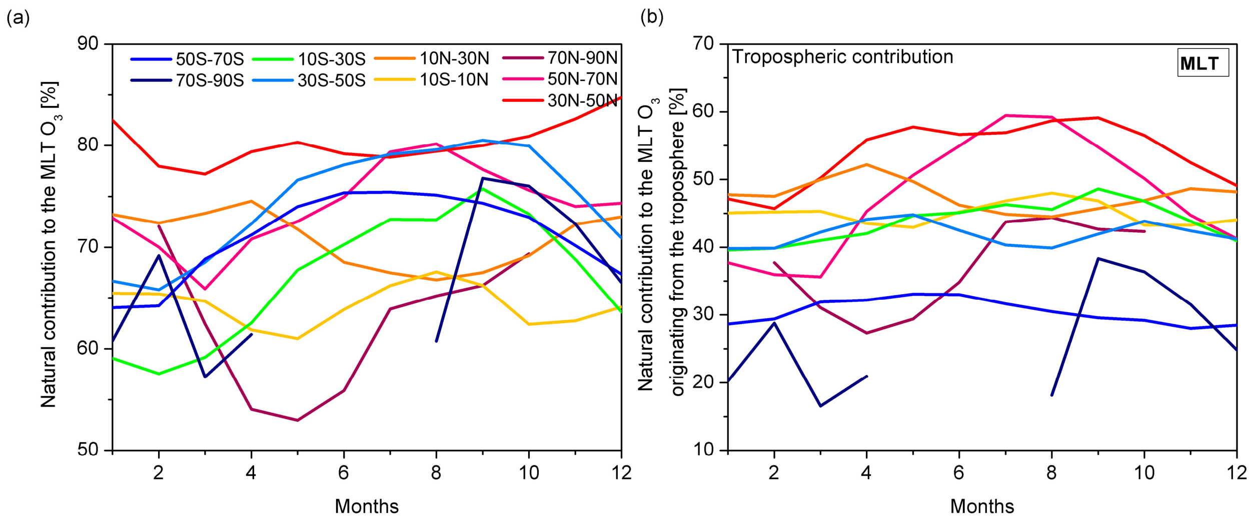

Figure 2: Contribution to the IASI MLT O_3 columns (%) (a) of the natural variability (troposphere and stratosphere) and (b) from the troposphere.