Response to Reviewer #2:

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 are developed 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) This investigation is well done in general but fails to account for lags between the geophysical quantities such as QBO and ENSO and ozone variability in non-local regions.]
A typical time-lag relation between ozone and ENSO from 0 to 4 months was examined before the submission of the paper, but it did not show any improvement in terms of residuals and uncertainty of the fitted parameters. We interpret this by a weak apparent contribution of ENSO to the IASI ozone time series in zonal bands outside the Niño region 3.4. This is now specifically mentioned in Section 3.2.

Optimizing the regression model by including time-lags in different geophysical variables could be investigated in the future but this is beyond the scope of this study.

We acknowledge, however, in the revised Section 4.2 that the missing representation of time lags in the proxy times series may lead to underestimating the influence of some geophysical parameters on the O_3 variations, in particular that of ENSO and QBO in zonal bands outside the regions where these geophysical quantities are measured (i.e. Niño region 3.4 for ENSO and Singapore for QBO). The fact that the regression model could be improved this way is now also mentioned in the Conclusion.

[2) Following this analysis, the authors make a fairly convincing argument that the density of the IASI measurements allows for trend analysis even over short timescales, though the comparison between FTIR instruments and IASI should be more carefully done.]
We thank the referee for his suggestion. As one can expect from the FTIR measurements illustrated on Figure 10 of the manuscript, IASI time series with the same sampling as the ones of the FTIR leads to non-significant trends. This has been tested by applying the regression models on a subsampled IASI dataset with the same temporal resolution as that of the FTIR datasets. In all cases, we observe that the fitted trends inferred from IASI and FTIR are within the uncertainties of each other and that the uncertainties associated with the subsampled IASI datasets are significantly larger than the ones with the full daily dataset. These trend values have been added in Table 4 of the manuscript and this is mentioned in the revised Section 4.3.3.

We now conclude this section 4.3.3 with:

"Even if validating the IASI fitted trends with independent datasets is challenging due to the short-time period of available IASI measurements and the insufficient number of usable correlative measurements over such a short period, the results obtained for trends inferred from IASI *vs* FTIR tend to confirm the conclusion drawn in subsections 4.3.1 and 4.3.2, that the high temporal sampling of IASI provides good confidence in the determination of the trends even on periods shorter than those usually required from other observational means."

[3) The paper includes supplementary information with a model analysis aimed at quantifying how much the tropospheric ozone signal seen by IASI is "contaminated" by stratospheric ozone. It is unclear to me, however, that their approach quantifies how much of the tropospheric ozone that IASI sees is stratospheric due to the "smearing" of the averaging kernels (what they are trying to quantify) versus stratospheric air that has actually been transported into the troposphere.]

There are indeed two combined effects in the stratospheric contribution as seen by IASI in the troposphere: 1) the stratospheric influence simply calculated as the difference between the total simulated O_3 and the $O_3^{tagged\_NOx}$ and 2) an additional part due to the limited vertical sensitivity of IASI in the troposphere; the latter is accounted for by applying the IASI averaging kernels on the MOZART4 stratospheric contribution. This simulates the stratospheric part as observed by IASI in the troposphere. This is now better explained in the Supplement.

In addition, as suggested by both referees #1 and #3, we now illustrate in the revised Supplement (Figure S4a), the fit of MOZART-4 O_3 and of $O_3^{tagged\_NOx}$ time series, in addition to the stratospheric contribution (Figure S4b), without accounting for the IASI sensitivity to evaluate the effect of the smoothing error from the observational system (effect 2)). 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. S6a) 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.3b), this contribution reaches ~85% from which only ~30% originate from the stratosphere (Fig. S4b) and ~55% from the troposphere (see Fig. S6(b) or Fig. 1 here below). 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 in the 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).

Specific comments:

[1) Abstract
a) Page 27576, Line 3: Should there be a "the" before MetOp-A?
b)Page 27576, Line 5: "time development" is a little awkward – "temporal evolution" would be better.
c) Page 27576, Lines 15-19: The attribution of the trends is somewhat overstated in the abstract as compared to the paper. Perhaps "which is consistent with other studies suggesting a turnaround for stratospheric O3 recovery" and "possibly linked to the impact of decreasing ozone precursor emissions" would be more appropriate.]
a), b) and c) All corrections have been made.

[2) Introduction
a) Page 27577, Line 7: Suggest replacing "present" with "undergoes".
b) Page 27578, Lines 17-18: The wording is difficult to follow. I suggest "by the possibility of using IASI measurements to discriminate O3 distributions."]
a) and b) All corrections have been made.

[3) Section 2
a) Page 27579, Line 16: Given IASI's 9:30 / 21:30 overpass time, I would not expect it to be as sensitive to changes in precursor emissions as instruments with afternoon overpass times. Have you used a model to assess this and what implications the overpass time might have in terms of quantifying the true trend associated with decreases in precursor emissions?
b) Page 27581, Lines 8-17: Please see comments on the supplementary material. It is unclear to me whether the model analysis the authors performed actually quantifies how much of the tropospheric ozone that IASI sees is stratospheric due to the "smearing" of the averaging kernels (what they are trying to quantify) versus stratospheric air that has actually been transported into the troposphere.]
a) The overpass time should not have such an impact due to the long lifetime of O_3 in the mid- and upper troposphere where the IASI sensitivity is the largest.
b) See general comment 3). This is now better explained in the Supplement.

[3) Section 3
a) Page 27581, Lines 20-21: Why just ODS-driven trends here? The authors are not specifically talking about stratospheric ozone, and in fact address tropospheric ozone trends driven by precursor emissions. Also, in both instances in this sentence, "trend" should be preceded by "the" or should be plural.
b) Page 27582, Lines 9-14: It would be very helpful if the authors could provide more detail here. Why was the time lag for the autocorrelation of the residuals assumed to be 1 day or 1 month? Given the lifetime of ozone, might it not be longer? What method was used to correct the coefficient estimates by accounting for this autocorrelation?
c) Page 27582, Lines 14-15: "robust" would be more accurate than "adequate"
d) Page 27583: In the analysis using geophysical variables, the zonal wind at 10 and 30hPa are used to represent the QBO. However, it is the QBO shear rather than the zonal wind itself that strongly affects the zonal distribution of ozone, which responds primarily to the anomalous QBO thermal wind circulation cell driven by the zonal wind gradient. While the temporal evolution of the shear is generally consistent with the temporal evolution of the zonal wind itself, there can be

important differences when the descent of a particular QBO phase is delayed or occurs faster than usual. The authors may want to compare the time series of the shear to that of the individual wind components to determine whether using the shear time series might make an important difference in their results.
e) Page 27583, Line 17: What is meant by “both components” here?
f) Page 27583, Line 27: “there is a” is needed before “predominance of easterlies”
g) General comment on Section 3: For many of the geophysical variables considered here, there may be important lags between the geophysical quantity and the ozone response in particular parts of the atmosphere. Lower stratospheric ozone in midlatitudes, for example, does show a QBO signal, but it lags the QBO winds in the tropics by a few months. Likewise, midlatitude ozone does not respond within a month to ENSO changes in the tropics. Optimizing the regressions including the possibility of time lags may be too involved for this paper, but the authors should at least acknowledge that they may be underestimating the role of geophysical variables in regions outside the location of the geophysical quantity for QBO, ENSO, NAO, and AAO.]
a) This sentence has been rewritten:
” In order to characterize the changes in ozone measured by IASI and to allow a proper separation of trend, …”

b) 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 with the Eq. (3).

The autocorrelation coefficients at various lags were examined for each layer in each latitude bands (cfr example for the 30N-50N band provided in Table below) and lag 1 appeared to be the most important in all cases. As a consequence, a time lag of one day (or one month) has been preferred to account for the correlation of the noise residuals.

30N-50N	*lag1*	*lag2*	*lag3*	*lag4*	*lag5*
MLT	**0.65**	0.54	0.49	0.47	0.44
UTLS	**0.62**	0.48	0.40	0.36	0.29
MLST	**0.75**	0.64	0.56	0.50	0.44
UST	**0.80**	0.68	0.62	0.56	0.49
TOTAL	**0.78**	0.69	0.62	0.58	0.54

c) This has been changed.
d) We thank the referee for his suggestion.
However, to the best of our knowledge, the time series of monthly averaged wind speed measurements done by the ground-station in Singapore is the most widely used proxy for the quasi-biennial oscillation (QBO). Previous studies on QBO have shown that the longitudinal differences in the phase of the QBO are small (e.g. Baldwin et al., 2001 and references therein), and, as a consequence, the dataset used in this study is supposed to represent well the equatorial belt.

We agree with the referee that analyzing the structure of the QBO by comparing the QBO shear with the zonal wind would be interesting. We feel however that this is beyond the scope of this study.

[Baldwin, M.P., L.J. Gray, T.J. Dunkerton, K. Hamilton, P.H. Haynes, W.J. Randel, J.R. Holton, M.J. Alexander, I. Hirota, T. Horinouchi, D.B.A. Jones, J.S. Kinnersley, C. Marquardt, K. Sato, and M. Takahashi, 2001: The Quasi-Biennial Oscillation. Reviews of Geophys., 39, 179-229.]

e) This is now specified: "The difficulty in discriminating solar flux and linear trend terms…".
f) It has been added.
g) See responses to General comment 1).

[4) Section 4
a) Page 27587, Line 16-18: Why don't we see ozone depletion in the Southern hemisphere stratosphere?
b) Page 27587, Lines18-19: There appears to be a difference between the Northern and Southern hemispheres in terms of how similar the 10 km panel is to the 19 km panel. Is this the case? What is the reason for the asymmetry?
c) Page 27587, Lines 10-26: What is the source of the noise at high latitudes?
d) Page 27588, line 3: It would be much clearer to say "color contours" rather than "colorscale"
e) Page 27588, lines 8-14: High midlatitude lower stratospheric ozone values in 2010 have been linked to the combined Easterly shear QBO and El Nino (Neu et al., Nature Geosci., 2014), and the failure to reproduce them with the regression may also be due to 1) the use of the zonal wind as a QBO proxy rather than the shear in the zonal wind and / or 2) the failure to account for lags between the QBO and ENSO and the response of midlatitude tratospheric ozone.
f) Page 27589, Lines 20-21: Is it clear that the spring maxima solely reflect STE, or is there also a seasonal dependence of the averaging kernels that makes the lower atmosphere more sensitive to the layers above during spring?
g) Page 27591, Lines 5-14: The QBO results for the midlatitudes may differ substantially if a lag were considered. The authors should at least acknowledge that by using zonal winds rather than shear and not considering a lag, they likely underestimate the QBO's importance in regions not directly impacted by the QBO winds.
h) Page 27591, Lines 15-22: The negative ENSO coefficient in the tropical UTLS is consistent with results from Neu et al., Nature Geosci., 2015.
i) Page 27592-27593, Lines 24-1: I don't understand what the percentages refer to here.
j) Page 27594, Line 14: need an "increase" in "reports a factor of two in"
k) Section 4.3.3: I am not sure I agree with the conclusions presented here. The authors show that total column trends from FTIR data and from IASI over different time periods do not agree and use this to argue that the sampling of IASI provides confidence in the determination of trends. However, to actually reach this conclusion, they would need to show that trends between IASI sub-sampled at the same times and locations as the FTIR measurements are consistent with those from the FTIRs (i.e. non-significant) for the overlapping period of the data (2008-2012). Only by doing so can they demonstrate the advantage of IASI sampling and show that it explains the difference between IASI and the FTIR measurements.]
a) As mentioned in Section 2, only daytime observations which are characterized by a good vertical sensitivity to the troposphere are used in this study. As a result, we do not include in this

study the variations due to the ozone hole formation (also mentioned in Section 4.2), which is otherwise well monitored by IASI using night-time measurements.

b) We observe indeed an asymmetry between the contribution of the MLST into the UTLS in the N.H. and the S.H. A deeper analysis of the correlation between the MLST and the UTLS layers as a function of IASI sensitivity and dynamics should be investigated to explain this observation. The stratosphere-troposphere exchanges could be part of the answer since they are expected to be larger in the N.H. than in the S.H. There are also significant hemispheric differences in the strength and behavior of the Brewer-Dobson circulation, which is usually the largest in the N.H., because of hemispheric differences in wave forcings. The use of a stratospheric chemical and transport model could help in quantifying this effect, but this is beyond the scope of the study.

c) This is because of the low temporal sampling of daytime IASI measurements in high latitudes.

d) It has been changed.

e) We thank the referee for pointing that out. These possible explanations have been added in the revised paper.

f) It is more probably related to the STE processes. There is indeed a seasonal dependence of the averaging kernels but the sensitivity is the lowest in the winter (implying more information coming from the stratosphere), not in spring.

g) See responses to General comment 1). This is now specified in this Section 4.2.

h) We thank the referee for pointing this reference which has been added.

i) The percentages represent the averaged fitted model residual relative to the IASI O_3 time series. This is now better explained.

j) It has been changed.

k) The influence of the FTIR temporal sampling on IASI is now tested in the revised paper. See general comment 2).

[5) Tables and Figures

Table 2: Caption is very unclear – it should say "based on daily (top values) and monthly (bottom values)"]

It has been changed.

[6) Supplementary Material

a) Lines 19-21: Why use constant emissions, particularly when the goal is investigate variations in ozone with time? Also, it is not clear whether the emissions are truly constant or simply have no interannual variability.

b) Lines 30-32: Are representative averaging kernels used, or scene-dependent averaging kernels?

c) Lines 37-40: This goes back to the previous question – if the averaging kernels underestimate the stratospheric influence on the partial column this could produce a bias.

d) Lines 47-48: One might expect the model to perform better in the southern hemisphere since the fire emissions do vary with time, but it isn't clear that this is the case. Why not?

e) Section S.3: Given the large difference between the model and IASI, are the IASI averaging kernels (which must depend on ozone amount) still applicable to the model profiles? Also, the authors are using the difference between total ozone and the tagged ozone in MOZART (with the IASI averaging kernels) as a measure of the stratospheric influence on tropospheric ozone as seen by IASI. However, this term includes both stratospheric air that has been transported to the troposphere and the "smearing" of the stratospheric signal into the troposphere by the averaging kernels. I understood the point of this analysis to be to understand how much the IASI tropospheric column is “contaminated” by stratospheric ozone due to the averaging kernels rather than to understand how much stratospheric ozone matters for tropospheric ozone variability. In this case, wouldn’t a better measure be the difference in the difference between total ozone and tagged ozone with and without the IASI averaging kernels?

f) Line 76: The sentence beginning “This method allows” should be reworded.

g) Line 110: Again, I am confused as to whether the emissions have no variability at all or simply no interannual variability.

h) Lines 120-122: I don’t think it’s clear from this analysis that the decrease in tropospheric ozone is “much more important than what we estimate from IASI”. The emissions-driven portion of it may be, but tropospheric ozone is a mixture of the emissions-driven ozone and ozone that enters through STE, and the authors have not convincingly shown that the “true” trend in this mixture of air is significantly larger than that observed by IASI (because they have not fully separated the influence of STE and the averaging kernels)]

a) As mentioned in the Supplement, the anthropogenic emissions have no variability at all, they are constant over years. On the contrary, biomass burning and biogenic emissions vary seasonally. This is now clearly mentioned in the revised supplement. A reference for the biogenic emissions has been added.

We have preferred the POLMIP emission dataset in this study because it has been built to represent as well as possible the anthropogenic emissions, particularly in the N.H.. The use of the POLMIP dataset in POLMIP models, amongst with MOZART-4, has also been extensively evaluated recently (Monks et al., ACP, 2014; Arnold et al., ACP, 2014; Emmons et al., ACP, 2015). The description of the dataset has been extended in the Section S1 of the revised Supplement.

b) We used the scene-dependent averaging kernels. Similarly to Wespes et al. (2012), the AvK of the different IASI observations contained in each MOZART grid have been considered to smooth the gridded MOZART profile interpolated on the IASI profile. This is now indicated in the Supplement.

c) See general comment 3). Because the IASI sensitivity is excellent in the stratosphere and smaller in the troposphere, the averaging kernels rather overestimate the stratospheric influence on the MLT column. This is now better explained in the Supplement and illustrated with Figures S4 and S5.

d) Large fire emissions are also observed in the N.H. (e.g; the fire NO emissions in the N.H. represent more than 60% of the global NO fire emissions in the POLMIP dataset; Siberia, South-East Asia and China regions account for ~30% of the total NO fire emissions). We relate the large positive bias of MOZART-4 in the MLT in equatorial belt (~25%) and extra-tropical regions in the Supplement to possible issues with horizontal transport in the model or overestimated ozone production efficiency at these latitudes.

e) See general comment 3). The stratospheric influence without accounting for the IASI averaging kernels is now shown in Fig. S4 of the Supplement and the MLT variability originating from the troposphere as seen by IASI is now quantified and illustrated in Fig. S6 and in Fig.1 here below.

f) This has been changed.

g) See comment 6a) above.

h) See general comment 3) and specific comment 6e) above. The conclusions in the Supplement have been rewritten accordingly.

Figure caption

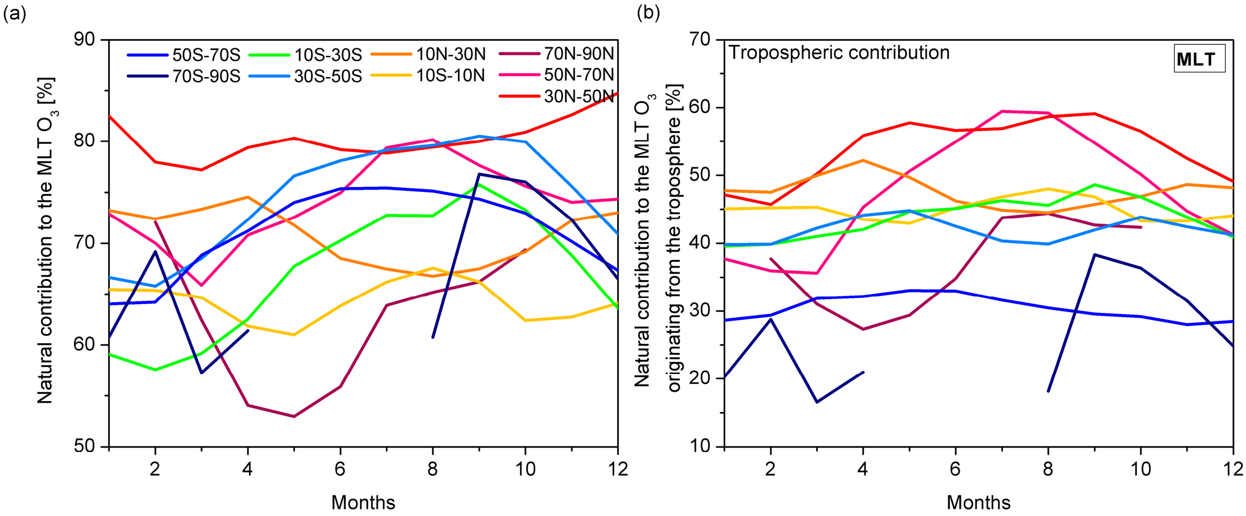

Figure 1: Contribution to the IASI MLT O_3 columns (%) (a) of the natural variability (troposphere and stratosphere) and (b) from the troposphere.