Response to Referee #2:

We thank the referee for her/his comments. Responses to individual comments that have been quoted […] are given here below.

[There are major differences between IASI trends (both for this submitted paper and in the TOAR) and trends measured from other independent sources of tropospheric ozone. The reported negative trends (in both NH and SH) for IASI tropospheric column ozone do not appear to be reproduced by other key data sources of tropospheric ozone. This paper does not compare IASI trends with several key studies on trends and also does not compare IASI trends directly with those derived from other independent data. The current paper will require major analysis/changes by the authors – they should compare more extensively with other studies on tropospheric ozone trends and reconcile differences. In addition the authors should compare with either ECC sondes or aircraft measurements (or even both) to evaluate the trends from IASI. As a note, just like the ECC sondes, aircraft data from MOZAIC+IAGOS for 1994-recent are public domain and can be compared on a region-by-region basis with IASI trends. A recent paper by Petetin et al. (2016) examined the long record of MOZAIC+IAGOS aircraft tropospheric ozone for 1994-2012 and did not measure negative trends in any season as reported here for IASI. Here is a paper that describes the MOZAIC and IAGOS ozone instruments and shows that the two time series can be joined for trend studies: Instrumentation on commercial aircraft for monitoring the atmospheric composition on a global scale: the IAGOS system, technical overview of ozone and carbon monoxide measurements Philippe Nédélec, Romain Blot, Damien Boulanger, Gilles Athier, Jean-Marc Cousin, Benoit Gautron, Andreas Petzold, Andreas Volz-Thomas & Valérie Thouret Tellus B: Chemical and Physical Meteorology Vol. 68 , Iss. s1,2016. Near-daily MOZAIC/IAGOS ozone profiles are available above Frankfurt since 1994. These profiles extend from the surface to 12 km and cover the full depth of the troposphere at the latitude of Frankfurt.

There is no drift in the observations as these instruments are routinely calibrated. In terms of data quality and sampling frequency, Frankfurt is the world's best data record of tropospheric ozone profiles and it is ideal for evaluating monthly satellite tropospheric ozone products. The MOZAIC/IAGOS data are open access. Monthly mean profiles on pressure surfaces can be easily provided by Herve Petetin. He can also limit the analysis to the portions of profiles measured below the tropopause. Papers by Hervé Petetin: Herve.Petetin@aero.obs-mip.fr, Laboratoire d'Aérologie, Université de Toulouse, CNRS, UPS, France. The following paper shows no tropospheric ozone trend at Frankfurt for 1994-2012 in any season, except for winter where ozone has actually increased. The TOAR-Climate provides an update with data through 2013 and gets similar results. Petetin, H., V. Thouret, A. Fontaine, B. Sauvage, G. Athier, R. Blot, D. Boulanger, J.-M. Cousin, and P. Nédélec (2016), Characterizing tropospheric ozone and CO around Frankfurt between 1994–2012 based on MOZAIC-IAGOS aircraft measurements, Atmos. Chem. Phys., 16, 15147-15163, doi:10.5194/acp-16-15147-2016. https://www.atmos-chem-phys.net/16/15147/2016/
The following paper demonstrates that many profiles are available at Frankfurt during the morning, around the time of the IASI overpass:
Petetin, H., et al. (2016), Diurnal cycle of ozone throughout the troposphere over Frankfurt as measured by MOZAIC-IAGOS commercial aircraft, Elem. Sci. Anth., 4:129, DOI: http://doi.org/10.12952/journal.elementa.000129/.]

We thank the referee for his/her comments and suggestion about reconciling the trend divergence as recorded from independent datasets, but, at the same time, we feel that this is well strongly beyond the scope of the present paper. Also the discussion on the upper tropospheric O_3 falls outside of the manuscript which focuses, on purpose, on the middle-low tropospheric O_3 column from IASI. We would like to draw the attention of the referee to the following:

- As co-authors of that TOAR-Climate paper lead by A. Gaudel and O. Cooper, we are of course aware of the trend divergence between the TOAR datasets, which is for now an open question and which deserves further investigation (that huge piece of work will be attempted if there is a TOAR-2 project). We hope that the reviewer will appreciate that such a multi-instrument analysis is completely beyond the scope of this paper. In the present study, we use the dataset from a single instrument (IASI) and we intend to go further in the analysis of the ozone time series and, specifically, we seek to derive significant trends in tropospheric O_3 by applying to the IASI data record a full multilinear regression (MLR) model - instead of the straightforward but over-simplistic least-squares single linear regression (SLR) method used in the TOAR-climate. In fact the shortcomings from the SLR over the MLR are specifically discussed in the dedicated section 4.3 of the present manuscript, which demonstrates the interest of the MLR for better determining accurate/realistic trends and for further resolving trend biases between independent datasets (see also the conclusion section).

- We also would like to recall here that the TOAR-climate assessment report has identified as possible causes for the trend bias in TOCs the differences in the tropopause calculation (same tropopause definition but different temperature profiles are used) and in the instrument vertical sensitivities and sampling. This is described in the TOAR-climate report at the end of the Tropospheric Ozone Burden Section 5.7 (Ozone trend estimation): "… This can be taken into account by sampling and applying the AKs of each measurement type to a common model simulation with a known trend in tropospheric column ozone to find the resulting trend bias, if any. These validation and model sampling exercises will be the focus of future intercomparisons of remotely sensed tropospheric column ozone data products." This is an important but huge piece of work which will be attempted in the follow-on TOAR project.

  The difficulty in comparing, because of the lack of homogeneity between the existing datasets and between the methodologies, the trends from our analysis with those reported in the TOAR-climate assessment report, as required by the referee, is now better underlined in the revised manuscript, especially in the introductory Section 4.3 (L.416-426):
  "… Substantial effort in homogenizing independent tropospheric O_3 column (TOCs) datasets have been performed in the TOAR-climate assessment report (Gaudel et al., submitted to Elementa), but large SLR trend biases remain between the TOAR datasets, in particular, between the satellite datasets. The lack of homogeneity in terms of tropopause calculation (same tropopause definition but different temperature profiles are used), of instrument vertical sensitivities and of spatial sampling has been specifically pointed as possible causes for the trend divergence.
  Reconciling trend biases between the datasets (e.g. by applying the vertical sensitivity of each measurement type to a common platform, as proposed in the TOAR-climate assessment report) is beyond the scope of this study, but the improvement in using a MLR instead of a SLR model for determining more accurate/realistic trends is explored here …"

- A last point that we would like to highlight here is that, from an instrumental point of view, there is no drift in the IASI radiance data. This can easily be assessed as there are currently 2 IASI flying which show similar radiance measurements. IASI is the reference instrument used in the Global Space-based Inter-Calibration System (GSICS). Its instrumental design (based on the Michelson interferometry which spreads and, hence, attenuates the effect of the degradation, if any, over the whole spectral range, as opposed to UV sounders) prevents any instrumental degradation/drift and assure a very good radiometric accuracy and stability. The good performance of IASI is indeed confirmed from the excellent stability in the recorded radiances that are monitored daily at the

EUMETSAT ground segment, and from a series of successful validation studies which are mentioned in Section 2 of the manuscript.

However, it is true that two recent validation experiments lead by Arno Keppens/BIRA-IASB and Anne Boynard/LATMOS that were not available at the time of the submission but that are now submitted to this QOS special issue (and listed in the reference section) suggest a drift between IASI and the sonde data. Actually, the drift has been demonstrated in Boynard et al. (this issue) to result from a "jump" in the IASI O_3 time series between the period before and after September 2010. The reasons for this jump are still unclear. It translates to an "artificial" negative drift of around ~2.8 DU/dec in the N.H. (cfr Boynard et al., this issue) and, more particularly, of around ~2.7 DU/dec in the mid-latitudes of the N.H. (based on the stations characterized by the better temporal sampling). The amplitude of that drift is lower than the one of the averaged negative trend derived from the MLR in the N.H. (~5 DU/dec on average in summer; i.e. the drift cannot fully explain the trends reported in the present study). Furthermore, the drift strongly decreases (<|1| DU/dec on average) after the jump and becomes even non-significant for most of the stations (significantly positive drifts are also found for some stations) over the periods before or after the jump.

For overcoming the drift issue and avoiding any potential overestimation of the amplitude of the negative trends derived from the whole IASI dataset, the constant term used in the MLR model has been split into two components: one covering the period before the jump and one after the jump. We show that the resulting trends are quite similar to the previous ones. In particular, the band-like pattern of negative trends in the N.H. in summer is still clearly observed (i.e. the impact of the jump was likely compensated by the adjustments of other covariates in the previous model regression). The only major difference between the regression results is that significant negative trends that were detected in the high latitudes of the S.H. are now turning non-significant (cfr Figure 1 here below which compares the distribution of O_3 trends derived from the two regression models). These new results are incorporated in the paper. The changes that have been made to address the reviewer's concern include the following:

1. The drift reported in the two companion papers is now clearly mentioned in the revised manuscript:

- In Section 2, L.118-124: "Note, however, that a drift in the N.H. MLT O_3 over the whole IASI dataset is reported in Keppens et al. (this issue) and Boynard et al. (this issue) from comparison with O_3 sondes. This drift (~2.8 DU/dec in the N.H.) is shown in Boynard et al. (this issue) to result from a discontinuity ("jump" as called in Boynard et al., this issue) in September 2010 in the IASI O_3 time series, for reasons that are unclear at present. Furthermore, the drift strongly decreases (<|1| DU/dec on average) after the jump and it becomes even non-significant for most of the stations (significant positive drift is also found for some stations) over the periods before or after the jump, separately."
- In Section 2, L.137-140: "In order to take account of the observed "jump" properly, we modified the previously used MLR model so that the LT term is split into two components covering the periods before and after the September 2010 "jump, separately."

2. The figures 1 to 6 and 8 of the manuscript have therefore been reprocessed and they depict now the results derived from the improved regression model (including two constant terms to

account for the “jump” in Sep 2010 instead of only one constant term over the whole IASI period).

3. Finally, some words of caution have been added in the conclusion section about a possible impact of the reported drift on the trend estimates: “Nevertheless, it is worth noting that there could be a possible impact of the sampling (because of the cloud and quality filters applied) and of the “jump” in September 2010 that has been identified in the IASI dataset (see Section 2), in both MLR and SLR trends.”

[In addition, the following paper in ACPD shows evidence that UT ozone has actually increased across the NH mid-latitudes from 1995 to 2013:
Cohen, Y., et al. (2017), Climatology and long-term evolution of ozone and carbon monoxide in the UTLS at northern mid-latitudes, as seen by IAGOS from 1995 to 2013, ACPD, https://www.atmos-chem-phys-discuss.net/acp-2017-778/acp-2017-778.pdf/.]

We thank the referee for pointing out our attention on this recent paper. It should be noted, however that the present study is restricted to the tropospheric O_3 column from the ground to 300hPa, and hence avoids the UTLS. Finding difference in trends in these two layers is not a surprise. Indeed, we show in a companion paper (cfr Wespes et al., ACP, 2016; see Table 2) that the UTLS O_3 from IASI (defined as the partial column ranging from 300 to 150 hPa) is characterized by a significant positive trend in the mid-and high latitudes of the northern hemisphere – in agreement with Cohen et al., 2017 – while the lower tropospheric O_3 column features a significant negative trends in the 30°N-50°N band. This finding, in particular, demonstrates the possibility to decorrelate the troposphere and the UTLS from the IASI measurements.

[In your paper you also mention negative trends in the SH from IASI that are hard to explain. You reference an ACPD paper (Zeng et al., now published in ACP, 2017) that combined ozonesondes with a Chemistry-Climate Model for evaluating ozone trends for Lauder, New Zealand during 1987-2014. The Zeng et al. study found evidence of negative trends for 9-12 km column ozone, but no trends in upper tropospheric ozone (6-9 km) and distinctly positive trends for the lower troposphere (0-6 km). For most of the mid-latitude troposphere (i.e., 0-9 km) the trends that they measure for Lauder actually appear as positive rather than negative. It is also not certain how much their 9-12 km layer ozone is impacted by decadal decreases in lower stratospheric ozone. Shown below is a comparison that includes ozonesondes, Umkehr, and FTIR ozone at Lauder (this figure appears in the supplement to TOAR-Climate). While IASI-FORLI shows a strong ozone decrease at this location, the sondes, FTIR, and Umkehr data show no trends since 2000. There seem to be substantial discrepancies in IASI trends in not just the NH but also in the SH as well that the authors will need to reconcile.]

On the contrary to the highly vertically resolved ozone sonde profiles, IASI exhibits only one full information level in the troposphere (meaning that there is no decorrelation between the sub-layers in the troposphere). The column ranging from the surface to 300 hPa was initially chosen (cfr Wespes et al., 2016 and 2017) to limit as much as possible the influence of the stratosphere, but also to include the altitude of the maximum sensitivity of IASI in the troposphere. At Lauder, this altitude is typically around 6-8 km and the stratospheric contribution to the tropospheric columns (due to the IASI limited sensitivity and the natural portion from the stratosphere) is estimated to range between 40 and 50% (see the Supplementary materials in Wespes et al., 2016). In other words, we cannot expect to reproduce the exact same trends as those derived by Zeng et al. in specific 3 km sub-layers. Note finally that negative trends in the UTLS and in the low stratosphere were also derived from IASI in the 30°S-50°S band (see Table 2 in Wespes et al., 2016), and, hence, that the negative trends that we calculate in the mid-latitudes of the S.H likely originate from the stratosphere. This assumption is also suggested from the O_3-CO correlation study in Section 4.4 of the present paper and it would be in line with the explanation of Zeng et al. (2017). It is clearly mentioned in Section 2 and 4.1 of the manuscript.

The Zeng et al. reference has been updated.
Please, see our response to the first comment of Referee #2 above about reconciling trends.

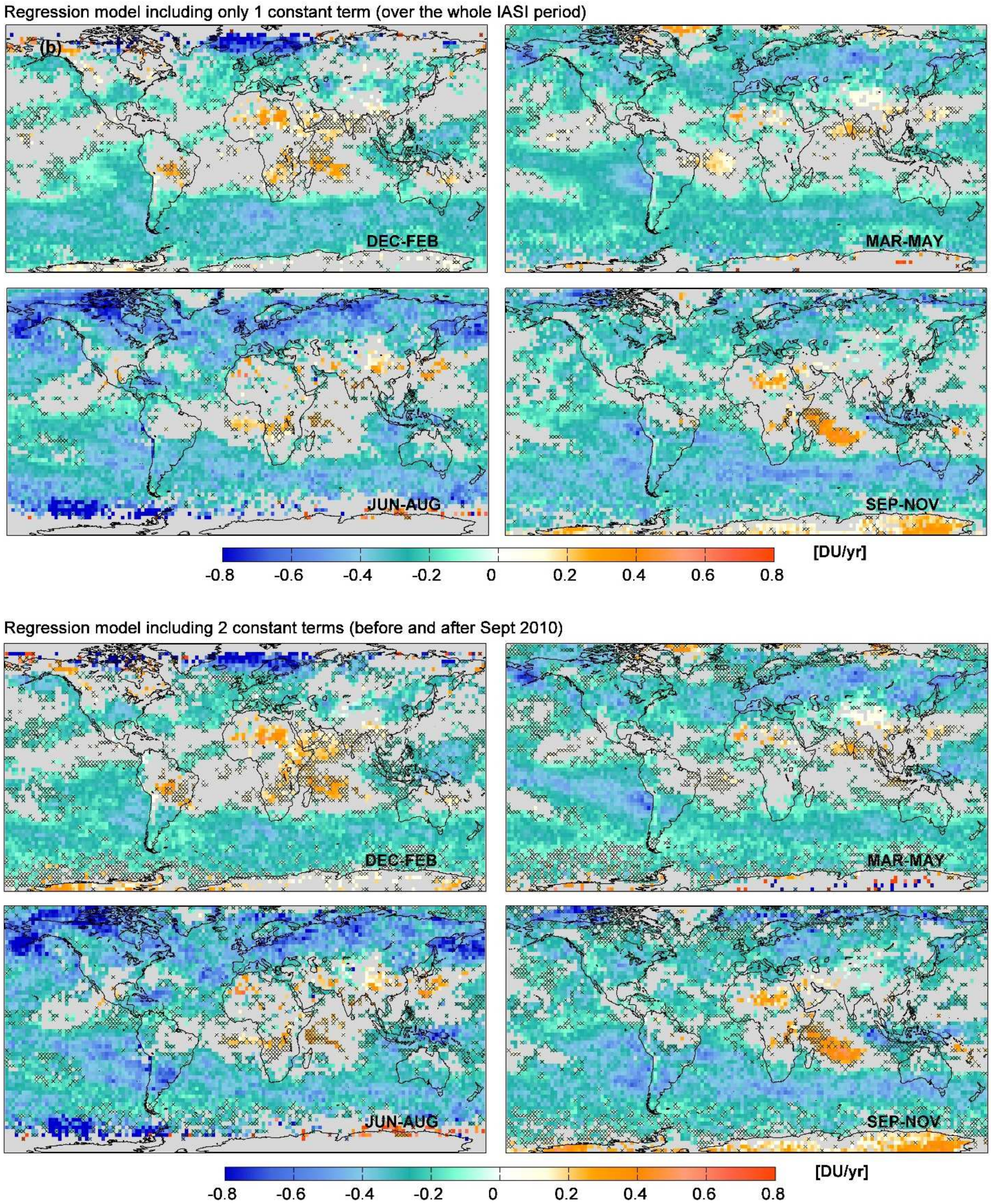

Fig.1: Comparison between the seasonal distributions of the adjusted trends (in DU/yr) obtained from the MLR model including one constant term (over the whole IASI period) *vs* those obtained from the MLR model including two constant terms (one before and one after Sept 2010).