Response to Referee #1:

We are grateful to the referee for her/his careful reading of the manuscript and for her/his comments and suggestions. Responses to individual comments that have been quoted […] are given here below.

Major comments

[The authors have performed a careful statistical analysis of a particular dataset. However, the manuscript does not discuss potential issues associated with the dataset that was used. The authors cite a number of papers that deal with characterization and validation of the IASI-FORLI dataset, but do not discuss any of the potential issues with the dataset that these works may have raised. Of particular relevance for this work on trends is the previous work by Boynard et al. [2016] (a paper on which all authors in this work were involved and cited in this work in the list of papers that deal with characterization of the IASI-FORLI dataset) that has shown that the IASI-FORLI dataset may have its own issues in terms of drifts with time. Figure 15 in Boynard et al. [2016] shows comparisons of IASI-FORLI with sondes over time. The figure appears to show a distinct negative drift in the IASI-FORLI surface-300 hPa ozone compared to sondes over the 2008-2015 time period. This is highly relevant to the results reported in this work, but was not discussed.]

We thank the referee for pointing that important feature out.

At the time of the initial submission, no drift in the IASI dataset was reported in previous papers (neither in Boynard et al., 2016) or in IASI quality assessment reports ever. From the Boynard et al 2016 paper, it was not obvious that IASI-sondes comparison was showing a drift between the 2 datasets. It was obviously not our intent to leave that feature out of the discussion.

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:

- 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 constant term is split into two components covering the periods before and after the September 2010 "jump, separately."
  - In Section 2, L.428-430: "Note that the constant term in the SLR is split into two components (covering the periods before and after the September 2010 "jump") to take account of the observed "jump (see Section 2)."

- The figures 1 to 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).

- 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."

[Also, in considering trends from the IASI-FORLI ozone dataset, the influence of clouds on sampling ought to at least be mentioned somewhere. If I understand correctly, the IASI-FORLI retrievals are only performed for relatively clear-sky cases. We might expect there to be changes in cloudiness over time, and this could potentially impact trend estimates for thermal-IR ozone.]

Actually, changes in cloudiness over time are not suspected to directly impact on the trend estimates. Only the FORLI retrievals with a cloud fraction in the field-of-view lower than 13%, i.e. only the clear or almost-clear scenes, are analyzed in this study. The maximum threshold of 13% for the cloud cover has been shown in previous studies to be good enough to consider the IASI pixel as clear for the O_3 retrievals (i.e. the atmosphere can be treated as a non-scattering medium in the radiative transfer code; cfr Clerbaux et al.,

2009; Hurtmans et al., 2012). It is now clearly mentioned in the revised Section 2 (L.104-106) that the cloud contaminated IASI scenes are filtered out:
"… measurements (defined with a solar zenith angle to the sun < 80°) which are characterized by a good spectral fit (determined here by quality flags on biased or sloped residuals, suspect averaging kernels, maximum number of iteration exceeded,…) and which correspond to clear or almost-clear scenes (a filter based on a fractional cloud cover below 13% has been applied; cfr Clerbaux et al., 2009; Hurtmans et al., 2012)…".

Note also that the use of quality flags (e.g. based on large residuals …) that are specified in Section 2 further helps in filtering the cloud contaminated IASI scenes.

We agree, however, that the use of a cloud filter (and of other quality flags) might influence the sampling of the dataset and, hence, that it might impact on the trend estimates. The effect of the temporal and of the spatial samplings on the trend biases was already mentioned in Section 4.3. It is also now indicated in the conclusion Section (L.570-573):
"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."

[There is no substantive discussion of how the trends from this analysis of IASI-FORLI data compare with those reported from radiosondes or from other satellite datasets. The authors do have some discussion in the introduction about difficulties and limitations associated with previous trend studies, and some rather vague, qualitative statements in Section 4.1 about how the trends determined from this work are consistent with findings in the literature However, there is some implication here, from this paragraph in the introduction, and from the lack of specific discussion of results from other studies in the conclusions, that the trends reported here from this IASI-FORLI analysis provide definitive and absolute answers. I felt that there ought to be some more discussion of these results in the context of the recent Gaudel et al. paper associated with the Tropospheric Ozone Assessment Report. (This paper, for which the authors of this work were also involved as co-authors, had previously been available for public comment and is currently in review for Elementa.) I appreciate that a reconciliation of the differences in the trends from different satellite datasets reported in the Gaudel et al. TOAR paper is outside the scope of this manuscript, and I appreciate that the Gaudel et al. paper used a linear regression approach rather than the more rigorous multivariate approach advocated for in this work. Nonetheless, I feel strongly that the point that there are discrepancies between trends from different datasets included in Gaudel et al. ought to be raised more prominently in this manuscript.]
We apologize if it is felt from reading the paper that we were so definitive in our conclusions. We are well aware that the accurate trend determination is a difficult task and we wanted to make the point that our results, in particular the comparison between MLS and SLR trends in the dedicated Section 4.3 which clearly highlights large differences in trend estimates, open perspectives for better determining accurate and realistic trends and for further resolving the trend biases between the existing datasets. Some clarifications have been brought in the last paragraph of the conclusion Section (L.582-587):
"This study supports overall the importance of using (1) high density and long term homogenized satellite records, such as those provided by IASI, and (2) complex models with predictor functions that describe the O_3-regressors dependencies for a more accurate determination of trends in tropospheric O_3 - as required by the scientific community, e.g. in the Intergovernmental Panel on Climate Change (IPCC, 2013) - and for further resolving trend biases between independent datasets (Payne et al., 2017; the TOAR report) …"

We understand the concern of the reviewer considering the different results/conclusions presented here in comparison to those from TOAR. However, as it is clearly stated in the manuscript (e.g. in the introduction, in the introductory paragraph of Section 4.1 and of Section 4.3), the lack of homogeneity in terms of time-varying instrumental biases, of measurement periods, of spatial and temporal samplings, of boundaries of the O_3 columns and of vertical sensitivity and resolution of the measurements (cfr the TOAR-climate assessment report), combined with differences in the methodology used (MLR vs SLR) makes impossible to "quantitatively" compare our results with those from previous/parallel studies. The best we can do is to "qualitatively" discuss them with respect to the recent published findings that, furthermore, mostly focus on changes in O_3 precursor emissions. It is what we have specifically done in Section 4.1.

It is true that direct comparisons between SLR trends obtained from a series of available independent measurements (among others, IASI) using the same period and the same tropopause definition to limit the possible sources of discrepancies have been performed in the TOAR-climate assessment report. Nevertheless, large trend biases were reported between the different datasets and, more particularly, between the satellite datasets. 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 …"

Understanding/reconciling the trend biases is still an open question which deserves further investigation. That huge piece of work could be attempted if there is a TOAR-2 project.

[The authors raise some interesting speculative points about attribution of trends in tropospheric ozone, but since no rigorous attempts at attribution were made in this work, some care is needed with the language associated with these statements. Specific examples are provided in the minor comments below.]
As required by the referee, we have now taken care to avoid making too strong statements in the sections specified in the minor comments below (see the minor comments below related to this comment for the changes made in the revised version).

Please, note that, by presenting our results in light of recent reported studies and by exploiting the simultaneous O_3 and CO measurements from IASI, we have investigated, as much as possible, the potential of IASI to derive trends and to help in understanding the origins of the air masses. The only way to more rigorously attempt to attribute trends would be to use a chemistry-transport model which would allow to trace back the sources of the transported air masses. The use of a CTM is beyond the scope of this paper and could be interestingly explored in a future study.

[The discussion of attribution of trends (Section 4) is largely limited to changes in emissions. Why is this? What about long-term variations in stratosphere-tropopshere exchange and the influence on tropospheric ozone? I see that Section 4.1 mentions interannual variability in stratosphere-troposphere exchange in the discussion of trends in IASI-FORLI tropospheric ozone in the SH tropical region, but I did not understand

why this was not mentioned in the context of other regions. Presumably this could also be an important factor in mid-latitudes? (e.g. as per Verstraeten et al., 2015)?]

The influence of the stratosphere-troposphere processes on the tropospheric O_3 trends is specifically discussed in Section 4.1 (for the S.H. tropical region and the mid-high latitudes of the S.H.) and in Section 4.4 where one of the objectives is specifically to help in discriminating the tropospheric from the stratospheric air masses at a global scale by using simultaneous O_3 and CO measurements from IASI. The contamination from the stratosphere was indeed shown to be largest in the mid-high latitudes of both hemispheres (see figure 9 of the manuscript). We agree that the air masses identified, with the O_3/CO correlation analysis, as mainly originating from the troposphere may also reflect some minor stratospheric contributions and, hence, that the associated trends might be to some extent influenced by the variability in the stratosphere-troposphere exchanges. This influence is now specifically mentioned in the paragraph related to the trends calculated in the N.H. in the revised Section 4.1 and some values quantifying the influence of the stratosphere into the IASI MLT columns (taken from the supplementary materials in Wespes et al., 2016, which estimates, with a global CTM, the stratospheric portion into the tropospheric O_3 columns from IASI) have been added in Sections 4.1 and 4.4:

- Section 4.1, L.316-318: "We should also note that, even if these latitudes are characterized by the lowest stratospheric contribution (~30-45%; see supplementary materials in Wespes et al., 2016), it might partly mask/attenuate the trends in the tropospheric O_3 levels."
- Section 4.4, L.488-492 : '…the negative correlations for the high latitude regions might also reflect air masses originating from/characterizing the stratosphere due to natural intrusion or to artificial mixing with the troposphere introduced by the limited vertical sensitivity of IASI in the highest latitudes (stratospheric contribution varying between ~40% and 65%; see supplementary materials in Wespes et al., 2016)."

The study of Verstraeten et al. (2015) has also been added in the reference list and referred to in the revised Section 4.1 (L.271-275):
"… the tropospheric O_3 increases which have been shown to mainly result from a strong positive trend in the Asian emissions over the past decades (e.g. Zhao et al., 2013; Cooper et al., 2014; Zhang et al., 2016; Cohen et al., 2017; Tarasick et al., 2017; and references therein) but also from a substantial change in the stratospheric contribution (Verstraeten et al., 2015) …"

Minor comments

[In general, the paper would benefit from editing by an English language service. There are small eccentricities in grammar throughout the paper. They are so numerous that I have not attempted to list issues of grammar in the minor corrections below. However, in most cases, these are not an impediment to understanding.]

We have carefully proofread the paper in order to track down those grammatical eccentricities. We have found and corrected several of these errors in the revised version. In addition, an English language service will be provided by ACP during the proofreading phase before the final submission of the manuscripts to correct any grammar mistakes/incorrect word usages left in the revised manuscript.

[The authors have chosen to describe the quantity of interest (tropospheric column from ground to 300 hPa) as tropospheric ozone columns (TOCs) in this work. In the previous Wespes et al. [2016] companion paper, the authors had referred to this ground-300hPa quantity as middle-low troposphere (MLT) ozone. The Gaudel et al. TOAR-Climate paper states that the IASI-FORLI TCO used in that study relies on the WMO definition of the daily tropopause height. It seems confusing to refer to the ground-300 hPa values as TOCs.]

We thank the referee for highlighting that issue and we agree that it might be confusing. For consistency with those previous papers, we have substituted "TOC" for "MLT" through the revised manuscript.

[Abstract, line 23-24: "This finding supports the reported decrease of O3 precursor emissions in recent years". It would be more appropriate to say that the finding "is consistent with", rather than "supports".]
It has been changed in the revised version.

[Line 76-77: What is menat by "trend characteristics"? Consider an alternative choice of wording?]
"Trend characteristics" meant both the sign and the amplitude of a trend. It has been replaced by "trend parameters" in the revised version.

[Lines 103-104: "These profiles are characterized by a good vertical sensitivity to the troposphere and the stratosphere". I am not sure exactly what the authors mean here. Please consider an alternative choice of wording.]
This sentence has been corrected: "These profiles are characterized by a good vertical sensitivity IN the troposphere and the stratosphere".

[Figure 2: What is the difference between gray areas and crosses in Figure 2? This is not clear from either the manuscript text or the figure caption. Also, the crosses in Figure 2 are almost impossible to see. The crosses are also tough to see in Figure 5, but are a bit more visible in that figure, possibly because of the lighter colour scale. Please find a way to make the crosses more visible.]
As in Wespes et al. (2016), the grey areas indicate that the covariate (here the linear trend term) is not retained by the stepwise backward elimination process in the grid cell, while the crosses indicate that the regression coefficient of the covariate (which is retained by the elimination process) is turning non-significant in the 95% confidence limits when accounting for the autocorrelation in the noise residual at the end of the elimination procedure (cfr Section 3, L.174-177 of the manuscript). The meaning of the grey areas is now given in the revised manuscript (Section 3, L.185-186):
"The grey areas in the LT panels refer to the LT terms rejected by the stepwise backward elimination process".

The size of the black crosses in the Fig.2 and 5 is limited by the resolution of the grid cells (2.5° lat x 2.5° lon). The resolution of the figures has been improved in the revised version to make the crosses more visible.

[Lines 216-224: This is difficult to follow, possibly because the authors are trying to make a general statement that covers all eventualities. I was not sure what the main point of this paragraph should be.]
Lines 216-224 refer to the titles of Sections 4 and 4.1 and the description of Figure 5 (annual and seasonal distributions of the MLR trends). We think that the referee refers to the next lines (L.224-235). For clarifying the main point of that paragraph, the sentence has been rewritten (L.247-248 of the revised manuscript):

"… As a result, comparing/reconciling the adjusted trends with independent measurements, even on a qualitative basis, remains difficult. …"

[Lines 246-251: I think the wording of this statement is too strong, given the scope of this study. I was also surprised that this section does not mention stratosphere-troposphere exchange.]
We are not sure how to interpret this comment considering that Lines 246-251 of the original manuscript refer to:

"The large O_3 enhancement of ~0.33±0.23 DU/yr (i.e. 3.1±2.2 DU over the whole IASI period) stretching from southern Africa to Australia over the north-east of Madagascar during the austral winter-spring likely originates from large IAV in the subtropical jet-related stratosphere–troposphere exchanges which have been found to primarily contribute to the tropospheric O_3 trends over that region (Liu et al., 2016; 2017). Nevertheless, this finding should be mitigated by the fact that the trend value in the S.H. tropics is of the same magnitude as the *RMSE* of the regression residuals (~2-4.5 DU; see Fig.1)."

The impact of the stratosphere-troposphere exchange is here clearly mentioned in line with what was found in previous studies, and our results have been also treated carefully by considering the amplitude of the *RMSE* of the regression residuals.

Note that, as required by the referee in his/her last major comment, the influence of the variability in the stratosphere-troposphere exchanges on the MLT trends has been specifically mentioned in the revised paragraphs related to the trends derived in the N.H. and over the South-East Asia in Section 4.1. (see the related changes made in the revised version in the response to the last major comment above).

[Lines 272-281: I found the idea that the annual and summer trends for 2008-2016 are "amplified" relative to the trends for 2008-2013 hard to reconcile with the language about "leveling off". Can the authors please revise this paragraph for clarity?]

What we meant is that the amplitude of the negative trend calculated from IASI is larger over 2008-2016 than over 2008-2013, which supports the recent assumption of a levelling off of tropospheric O_3 and, further, suggests a possible decrease in the tropospheric O_3 levels.

The sentence has been rewritten for clarity (L.307-313):

"This finding is in line with previous studies which point out a possible leveling off of tropospheric O_3 in summer due to the decline of anthropogenic O_3 precursor emissions observed since 2010-2011 in North America, in Western Europe and also in some regions of China (e.g. Cooper et al., 2010; 2012; Logan et al., 2012; Parrish et al., 2012; Oltmans et al., 2013; Simon et al., 2015; Archibald et al., 2017; Miyazaki et al., 2017). It even goes a step further by suggesting a possible decrease in the tropospheric O_3 levels".

[Line 433 (and also line 523): I do not think it makes sense to talk about "air masses" in the context of seasonal means. Consider changing "air masses" to "outflow regions"?]

We thank the referee for that suggestion. "Air masses" have been changed to "outflow regions" and "patterns".

[Lines 471-482: I found this paragraph difficult to follow. China is not the only place where ozone precursor emissions have been decreasing in recent years. Perhaps it would be better just to say that the pollution outflow from Eastern Asia shows a stronger positive O3-CO relationship than the outflow from either the Eastern US or Europe and leave it at that? It does not seem that there is enough information here to make definitive statements about attribution.]

Actually the O_3-CO covariance ($COV_{O3\text{-}CO}$) that is discussed and analyzed in that paragraph provides additional information to $R_{O3\text{-}CO}$ and dO_3/dCO (that are discussed in the paragraphs above in the manuscript). It describes the joint variability of O_3 and CO, and clearly allows to identify North-East of India and East of China in summer as the regions in the N.H. characterized by the largest O_3-CO variability and, hence, by the most intense pollution episodes (in comparison with Eastern US and Europe). For avoiding possible misunderstandings, some clarifications have been made in the revised version (L.524-531):

"… To conclude, the particularly strong positive O_3-CO relationship in terms of $R_{O3\text{-}CO}$, dO_3/dCO and $COV_{O3\text{-}CO}$ measured over and downwind North-East India/East China in summer in comparison with the ones measured downwind East US and over Europe indicate that South-East Asia experiences the most intense pollution episodes of the N.H. with the largest O_3-CO variability ($COV_{O3\text{-}CO} > 40\times10^{33}$ mol^2.cm^{-4}) and the largest O_3 enhancement ($dO_3/dCO > 0.5$) over the last decade. The strong O_3-CO relationship in that region is associated with the significant increase that is detected in the IASI O_3 levels downwind East of Asia (see Section 4.1)…”

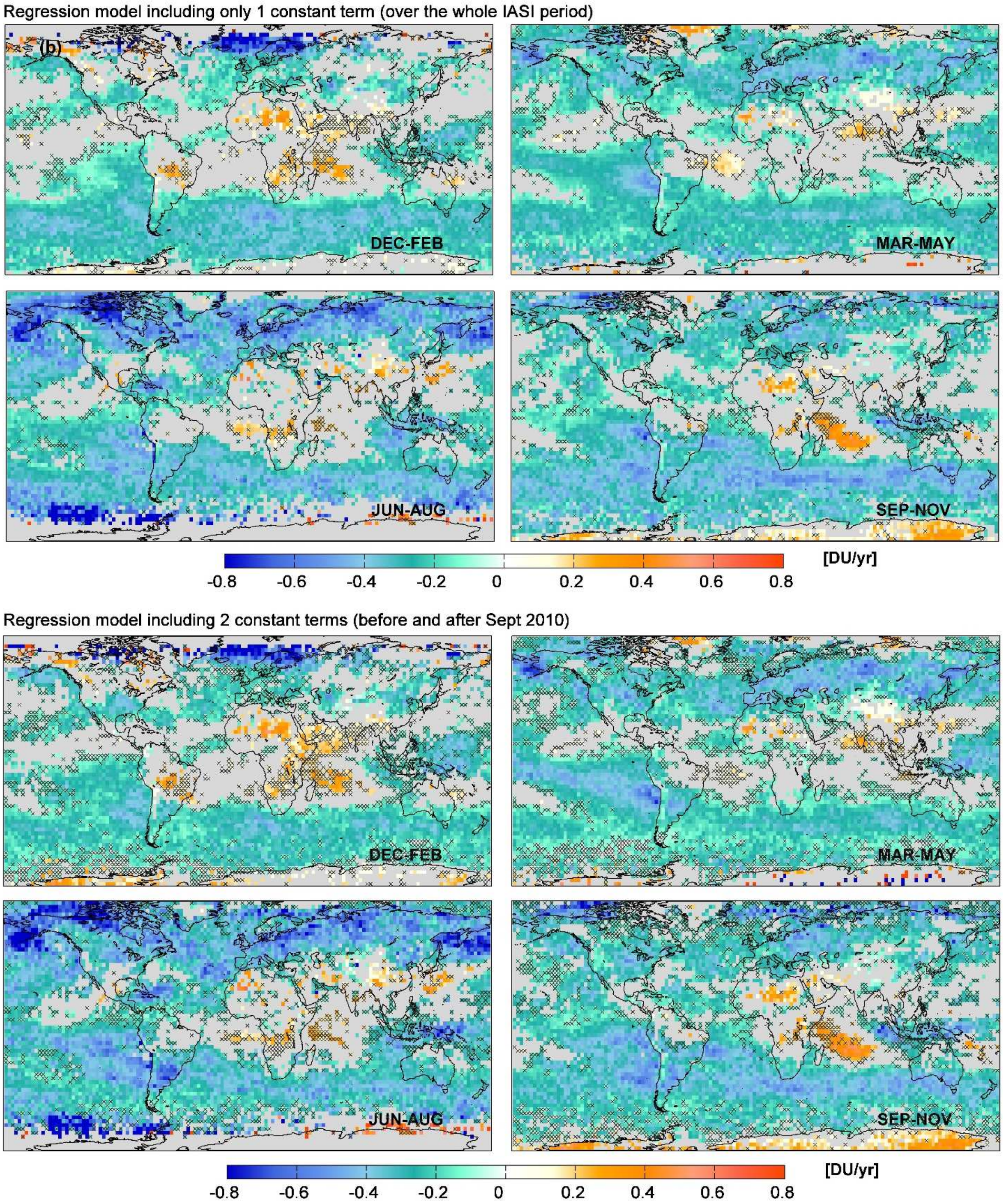

Fig.1: Comparison between the seasonal distributions of the adjusted trends (in DU/yr) obtained from the MLR model including one constant term (over the whole IASI period) *vs* those obtained from the MLR model including two constant terms (one before and one after Sept 2010).

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 constant term is split into two components covering the periods before and after the September 2010 "jump, separately."
- In Section 2, L.428-430: "Note that the constant term in the SLR is split into two components (covering the periods before and after the September 2010 "jump") to take account of the observed "jump."

2. The figures 1 to 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 (L.570-573) 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.

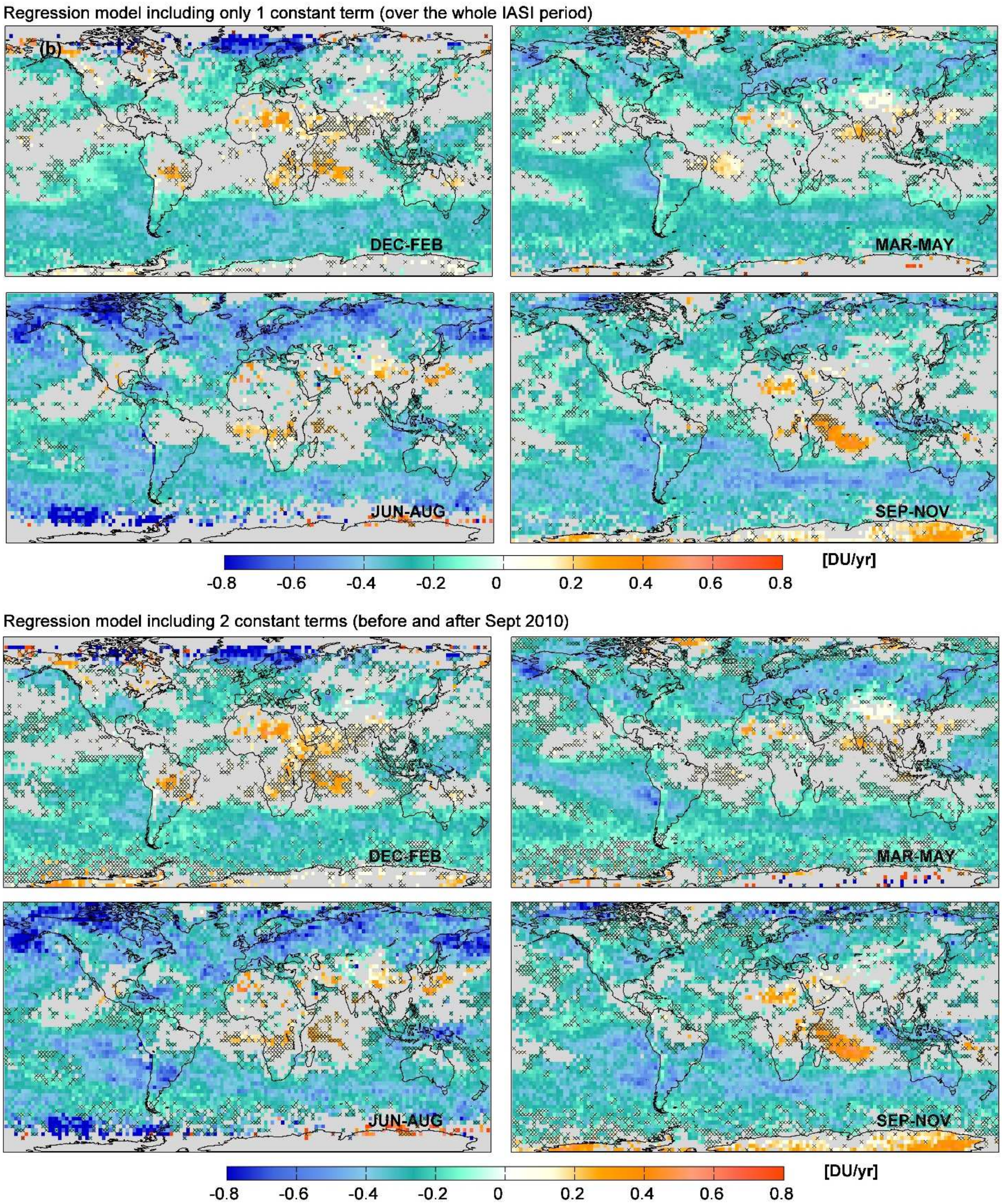

Fig.1: Comparison between the seasonal distributions of the adjusted trends (in DU/yr) obtained from the MLR model including one constant term (over the whole IASI period) *vs* those obtained from the MLR model including two constant terms (one before and one after Sept 2010).

List of relevant changes made in the manuscript:

Section 2:

- **L. 101-106:** "daytime measurements (defined with a solar zenith angle to the sun < 80°) which are characterized by a good spectral fit (determined here by quality flags on biased or sloped residuals, suspect averaging kernels, maximum number of iteration exceeded,…) and which correspond to clear or almost-clear scenes (a filter based on a fractional cloud cover below 13% has been applied; cfr Clerbaux et al., 200; Hurtmans et al., 2012)."

- **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 (~2.8DU/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."

- **L. 126-127:** "we focus on a tropospheric column ranging from ground to 300 hPa (called MLT – Middle-Low Troposphere – in this study)."

- **L. 137-140:** "In order to take account of the observed "jump" properly, we modified the previously used MLR model so that the constant term is split into two components covering the periods before and after the September 2010 "jump", separately."

Section 3:

- **L. 185-186:** "The grey areas in the LT panels refer to the LT terms rejected by the stepwise backward elimination process."

Section 4.1:

- **L. 247-248:** "As a result, comparing/reconciling the adjusted trends with independent measurements, even on a qualitative basis, remains difficult."

- **L. 271-275:** "This tends to indicate that the tropospheric O_3 increases which have been shown to mainly result from a strong positive trend in the Asian emissions over the past decades (e.g. Zhao et al., 2013; Cooper et al., 2014; Zhang et al., 2016; Cohen et al., 2017; Tarasick et al., 2017; and references therein) but also from a substantial change in the stratospheric contribution (Verstraeten et al., 2015) …"

- **L. 316-318:** "We should also note that, even if these latitudes are characterized by the lowest stratospheric contribution (~30-45%; see supplementary materials in Wespes et al., 2016), it might partly mask/attenuate the variability in the tropospheric O_3 levels."

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 where 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 the trend biases between the datasets 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 by …"

- **L. 428-430:** "Note also that the constant term in the SLR is split into two components (covering the periods before and after the September 2010 "jump") to take account of the observed "jump" (see Section 2)."

Section 4.4:

- **L. 490-492:** "by the limited vertical sensitivity of IASI in the highest latitudes (stratospheric contribution varying between ~40% and 65%; see supplementary materials in Wespes et al., 2016)."

- **L. 529-531:** "The strong O_3-CO relationship in that region is associated with the significant increase that is detected in the IASI O_3 levels downwind East of Asia (see Section 4.1)"

Conclusion Section:

- **L. 570-573:** "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."
- **L. 582-585**: "This study supports overall the importance of using (1) high density and long term homogenized satellite records, such as those provided by IASI, and (2) complex models with predictor functions that describe the O_3-regressors dependencies for a more accurate determination of trends in tropospheric O_3…"

Throughout the manuscript:

- TOC → MLT
- Figures 1 to 8 have been updated. Hence, the values referring to these figures have been changed accordingly in the text.

References:

- **L.1015-1017:** "Verstraeten, W.W., J.L. Neu, J.E. Williams, K.W. Bowman, J.R. Worden and K.F. Boersma: Rapid increases in tropospheric ozone production and export from China, Nature Geosciences, doi: 10.1038/NGEO2493, 2015."

Decrease in tropospheric O_3 levels of the Northern Hemisphere observed by IASI

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Abstract

In this study, we describe the recent changes in the tropospheric ozone (O_3) columns ~~(TOCs)~~ measured by the Infrared Atmospheric Sounding Interferometer (IASI) onboard the Metop satellite during the first 9 years of the IASI operation (January 2008 to May 2017). Using appropriate multivariate regression methods, we discriminate significant linear trends from other sources of O_3 variations captured by IASI. The geographical patterns of the adjusted O_3 trends are provided and discussed on the global scale. Given the large contribution of the natural variability in comparison with that of the trend (25-85% *vs* 15- 50%, respectively) to the total O_3 variations, we estimate that additional years of IASI measurements are generally required to detect the estimated O_3 trends with a high precision. Globally, additional 6 months to 6 years of measurements, depending on the regions and the seasons, are needed to detect a trend of |5| DU/decade. An exception is interestingly found during summer ~~in the~~at mid-high latitudes of the North Hemisphere (N.H.; ~ 40°N-75°N) where the large absolute fitted trend values (~|0.5| DU/yr on average) combined with the small model residuals (~10%) allow the detection of a band-like pattern of significant negative trends. This finding ~~supports~~ is consistent with the reported decrease in O_3 precursor emissions in recent years, especially in Europe and US. The influence of continental pollution on that latitudinal band is further investigated and supported by the analysis of the O_3-CO relationship (in terms of correlation coefficient, regression slope and covariance) that we found to be the strongest at ~~the~~ northern mid-latitudes in summer.

1 Introduction

O_3 plays a key role throughout the whole troposphere where it is produced by the photochemical oxidation of carbon monoxide (CO), non-methane volatile organic compounds (NMVOCs) and methane (CH_4) in the presence of nitrogen oxides (NO_x) (e.g. Logan et al., 1981). O_3 sources in the troposphere are the in situ photochemical production from anthropogenic and natural precursors, and the downwards transport of stratospheric O_3. Being a strong pollutant, a major reactive species and an important greenhouse gas in the upper troposphere, O_3 is of highest interest for air quality, atmospheric chemistry and radiative forcing studies. Thanks to its long lifetime (several weeks) relatively to transport timescales in the free troposphere (Fusco and Logan, 2003),

O_3 also contributes to large-scale transport of pollution far from source regions with further impacts on global air quality (e.g. Stohl et al., 2002; Parrish et al., 2012) and climate. Monitoring and understanding the time evolution of tropospheric O_3 at a global scale is, therefore, crucial to apprehend future climate changes. Nevertheless, a series of limitations make O_3 trends particularly challenging to retrieve and to interpret.

Since the 1980s, while the O_3 precursors anthropogenic emissions have increased and shifted equatorward in the developing countries (Zhang et al., 2016), extensive campaigns and routine in situ and remote measurements at specific urban and rural sites have provided long-term but sparse datasets of tropospheric O_3 (e.g. Cooper et al., 2014 and references therein). Ultraviolet and Visible (UV/VIS) atmospheric sounders onboard satellites provide tropospheric O_3 measurements with a much wider coverage, but they result either from indirect methods (e.g. Fishman et al., 2005) or from direct retrievals which are limited by coarse vertical resolution (Liu et al., 2010). All these datasets also suffer from a lack of homogeneity in terms of measurement methods (instrument and algorithm) and spatio-temporal samplings (e.g. Doughty et al., 2011). Those limitations, in addition to the large natural inter-annual variability (IAV) and decadal variations in tropospheric O_3 levels (due to large-scale dynamical modes of O_3 variations and to changes in stratospheric O_3, in stratosphere-troposphere exchanges, in precursor emissions and in their geographical patterns), introduce strong biases in trends determined from independent studies and datasets (e.g. Zbinden et al., 2006; Thouret et al., 2006; Logan et al., 2012 ; Parrish et al., 2012 and references therein). As a consequence, determining accurate trends requires a long period of high density and homogeneous measurements (e.g. Payne et al., 2017).

Such long-term datasets are now becoming obtainable with the new generation of nadir-looking and polar-orbiting instruments measuring in the thermal infrared region. In particular, about one decade of O_3 profile measurements, with a good sensitivity in the troposphere independently from the layers above, is now available from the IASI (Infrared Atmospheric Sounding Interferometer) sounder aboard the European Metop platforms, allowing to monitor regional and global variations in tropospheric O_3 levels (e.g. Dufour et al., 2012; Safieddine et al., 2013; Wespes et al., 2016).

In this study, we examine the tropospheric O_3 changes behind the natural IAV as measured by IASI over January 2008-May 2017. To that end, we use the approach described in Wespes et al. (2017), which relies on a multi-linear regression (MLR) procedure, for accurately differentiating trends from other sources of O_3 variations; the latter being robustly identified and quantified in that companion study. In Section 2, we briefly review the IASI mission and the tropospheric O_3 product, and we shortly describe the multivariate models (annual or seasonal) that we use for fitting the daily O_3 time series. In Section 3, after verifying the performance of the MLR models over the available IASI dataset, we evaluate the feasibility to capture and retrieve significant ~~trend~~ trend parameters ~~characteristics~~, apart from natural O_3 dependencies, by performing trend sensitivity studies. In Section 4, we present and discuss the global distributions of the O_3 trends estimated from IASI in the troposphere. The focus is given in summer over and downwind anthropogenic polluted areas of the N.H. where the possibility to infer significant trends from the first ~9 years of available IASI measurements is demonstrated. Finally, the O_3-CO correlations, enhancement ratios and covariance are examined for characterizing the origin of the air masses in regions of positive and negative trends.

2 IASI O_3 measurements and multivariate regression

The IASI instrument is a nadir-viewing Fourier transform spectrometer that records the thermal infrared emission of the Earth-atmosphere system between 645 and 2760 cm^{-1} from the polar Sun-synchronous orbiting meteorological Metop series of satellites. Metop-A and –B have been successively launched in October 2006 and September 2012. The third and last launch is planned in 2018 with Metop-C to ensure homogeneous long-term IASI measurements. The measurements are taken every 50 km along the track of the satellite at nadir and over a swath of 2200 km across track, with a field of view of four simultaneous footprints of 12 km at nadir, which provides global coverage of the Earth twice a day (at 9:30 AM and PM mean local solar time). The instrument presents a good spectral resolution and a low radiometric noise, which allows the retrieval of numerous gas-phase species in the troposphere (e.g. Clerbaux et al., 2009, and references therein; Hilton et al., 2012; Clarisse et al., 2011).

In this paper, we use the FORLI-O_3 profiles (Fast Optimal Retrievals on Layers for IASI processing chain set up at ULB; v20151001) retrieved from the IASI-A (aboard Metop-A) daytime measurements (defined with a solar zenith angle to the sun < 80°) which ~~result from~~are characterized by a good spectral fit (determined here by quality flags on biased or sloped residuals, suspect averaging kernels, maximum number of iteration exceeded,…) and which correspond to clear or almost-clear scenes (a filter based on a fractional cloud cover below 13% has been applied; cfr Clerbaux et al., 200; Hurtmans et al., 2012). These profiles are characterized by a good vertical sensitivity in ~~to~~ the troposphere and the stratosphere (e.g. Wespes et al., 2017). The FORLI algorithm relies on a fast radiative transfer and retrieval methodology based on the Optimal Estimation Method (Rodgers, 2000) and is fully described in Hurtmans et al. (2012). The FORLI-O_3 profiles, which are retrieved ~~on~~at 40 constant vertical layers from surface up to 40 km and an additional 40-60 km one, have already undergone thorough characterization and validation exercises (e.g. Anton et al., 2011; Dufour et al., 2012; Gazeaux et al., 2012; Hurtmans et al., 2012; Parrington et al., 2012; Pommier et al., 2012; Scannell et al., 2012; Oetjen et al., 2014; Boynard et al., 2016; Wespes et al., 2016; Keppens et al. 2017; Boynard et al., 2017). They demonstrated a good degree of accuracy, of precision and of vertical sensitivity with no instrumental drift, to capture the large-scale dynamical modes of O_3 variability in the troposphere independently from the layers above (Wespes et al., 2017), with the possibility to further differentiate long-term O_3 changes in the troposphere (Wespes et al., 2016). 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 (~2.8DU/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.

For the~~is~~ purpose of this work, we focus on a tropospheric column ranging from ground to 300 hPa (called MLT – Middle-Low Troposphere – in this study) that includes the altitude of maximum sensitivity of IASI in the troposphere (usually between 4 and 8 km altitude), which limits as much as possible the influences of the stratospheric O_3 and that was shown in Wespes et al. (2017) to exhibit independent deseasonalized anomalies/dynamical processes from those in the stratospheric layers. The stratospheric contribution into the tropospheric O_3 columns have been previously estimated in Wespes et al. (2016) as ranging between 30% and 65% depending on the region and the season with the smallest contribution as well as the largest sensitivity in the northern mid-latitudes in spring-summer where the O_3 variations, hence, mainly originate from the troposphere. We use almost the same MLR model (in its annual or its seasonal formulation) as the one developed in the companion paper (see Eq.1 and 2; Section 2.2 in Wespes et al., 2017), which includes a series of geophysical variables in addition to a linear trend (LT) term. In order to take account of the observed "jump" properly, we modified the previously used MLR model so that the constant term is split into two components covering the periods before and after the September 2010 "jump", separately. The MLR which is performed using an iterative stepwise backward elimination approach to retain the most relevant explanatory variables (called "proxies") at the end of the iterations (e.g. Mäder et al., 2007) is applied on the daily IASI O_3 time series. The main selected proxies used to account for the natural variations in O_3 are namely the QBO (Quasi-Biennial Oscillation), the NAO (North Atlantic Oscillation) and the ENSO (El Niño–Southern Oscillation) (cfr Table 1 in Wespes et al. (2017) for the exhaustive list of the used proxies). Their associated standard errors are estimated from the covariance matrix of the regression coefficients and are corrected to take into account the uncertainty due to the autocorrelation of the noise residual (see Eq. 3 in Wespes et al. (2016)). The common rule that the regression coefficients are significant if they are greater in magnitude than 2 times their standard errors is applied (95% confidence limits defined by 2σ level). The MLR model was found to give a good representation of the IASI O_3 records in the troposphere over 2008-2016, allowing us to identify/quantify the main O_3 drivers with marked regional differences in the regression coefficients. Time-lags of 2 and 4 months for ENSO are also included hereafter in the MLR model to account for a large but delayed impact of ENSO on mid- and high latitudes O_3 variations far from the Equatorial Pacific where the ENSO signal originates (Wespes et al., 2017).

3 Regression performance and sensitivity to trend

In this section, we first verify the performance of the MLR models (annual and seasonal; in terms of residual errors and variation explained by the model) to globally reproduce the time evolution of O_3 records over the entire studied period (January 2008 – May 2017). Based on this, we then investigate the statistical relevance for a trend study from IASI in the troposphere by examining the sensitivity of the pair IASI-MLR to the retrieved LT term.

Figure 1 presents the seasonal distributions of tropospheric O_3 measured by IASI averaged over January 2008 – May 2017 (left panels), along with the root-mean-squared error of the seasonal regression fit (*RMSE*, in DU; middle panels) and the contribution of the fitted seasonal model into the IASI O_3 time series (in %; right panels), calculated as $\frac{\sigma\left(O_3^{\text{Fitted\_model}}(t)\right)}{\sigma\left(O_3(t)\right)}$ where σ is the standard deviation relative to the regression models and to the IASI O_3 time series. These two statistical parameters help to evaluate how well the fitted model explains the variability in the IASI O_3 observations. The seasonal patterns of O_3 measurements are close to those reported in Wespes et al. (2017) for a shorter period (see Section 2.1 and 3.1 in Wespes et al. (2017) for a detailed description of the distributions) and they clearly show, for instance, high O_3 values over the highly populated areas of Asia in summer. The distributions from Fig.1 show that the model reproduces between 35% and 90% of the daily O_3 variation captured by IASI and that the residual errors varies between 0.01 DU and 5 DU (i.e. the *RMSE* relative to the IASI O_3 time series are of ~15% on global average and vary between 10% in the N.H. in summer and 30% in specific tropical regions). On an annual basis (data not shown), the model explains a large fraction of the variation in the IASI O_3 dataset (from ~45% to ~85%) and the *RMSE* are lower than 4.5 DU everywhere (~3 DU on the global average). The relative *RMSE* ~~are~~is less than 1% in almost all situations indicating the absence of bias.

The seasonal distributions of the contribution to the total variation~~s~~ in ~~TOCs~~the MLT from the adjusted harmonic terms and explanatory variables, which account for the "natural" variability, and from the LT term are shown in Fig. 2 (left and right panels, respectively). The grey areas in the LT panels refer to the LT terms rejected by the stepwise backward elimination process. The crosses ~~in the LT panels~~ indicate that the trend estimate in the grid cell is non-significant in the 95% confidence limits (2σ level) when accounting for the autocorrelation in the noise residual at the end of the elimination procedure. While the large influence of the seasonal variations and of the main drivers - namely ENSO, NAO and QBO - on the IASI O_3 records has been clearly attested in Wespes et al. (2017), we demonstrate in ~~with~~ Fig.2 that the LT also contributes considerably to the O_3 variations detected by IASI in the troposphere. The LT contribution generally ranges from 15% to 50%, with the largest values (~30-50%) being observed at mid-high latitudes ~~of~~in the S.H. (30°S-70°S) and ~~of~~in the N.H. (~45°N-70°N) in summer. In the S.H., they are associated with the smallest tropospheric O_3 columns (Fig.1; left panels) and the smallest natural contributions (<25%; left panels), while in the N.H. summer, they interestingly correspond to large MLT O_3

columns~~TOCs~~, large natural contributions (~50-60%) and the smallest *RMSE* (<12 % or <3 DU). From the annual regression model, the natural variation and the trend contribute respectively for 30-85% and up to 40% to the total variation in ~~TOCs~~the MLT.

In Fig.3, we further investigate the robustness of the estimated trends by performing sensitivity tests in regions of significant trend contributions (e.g. in the N.H. mid-latitudes in summer; cfr Fig.2). The ~9-year time series of IASI O_3 daily averages (dark blue) along with the results from the seasonal regression model with and without ~~including~~ the LT term included in the model (light blue and orange lines, respectively) are represented in the top row panel for one specific location (Fig.3a and b; highlighted by a blue circle in the JJA panel in Fig.4). The second row panel provides the deseasonalised IASI (dark blue line) and fitted time series (calculated by subtracting the adjusted seasonal cycle from the time series) resulting from the adjustment with and without ~~including~~ the LT term included in the MLR model (light blue and orange lines, respectively). The differences between the fitted models with and without LT are shown in the third rows (pink lines). They match fairly well the adjusted trend over the IASI period (3[d] row panel, grey lines; the trend and the *RMSE* values are also indicated) and the adjustment without LT leads to larger residuals (e.g. $RMSE_{\_JJA\_w/o\_LT}$ =3.37~~25~~ DU *vs*. $RMSE_{\_JJA\_with\_LT}$ =3.21~~14~~ DU in summer). This result demonstrates the possibility to capture trend information from ~9 years of IASI-MLR with only some compensation effects by the other explanatory variables, contrary to what was observed when considering a shorter period of measurements or a lesser temporal sampling (i.e. monthly dataset; e.g. Wespes et al., 2016). It is also worth to mention that the O_3 changes calculated over the whole IASI dataset in summer are larger than the *RMSE* of the model residuals (increase of 5.39±1.86 DU *vs RMSE* of 3.21~~14~~ DU), underlying the statistical relevance of trend estimates.

The robustness of the adjusted trend is verified at the global scale in Fig.4 which represents the seasonal distributions of the relative differences in the *RMSE* with and without ~~including~~ the LT included in the MLR model, calculated as $[(RMSE_{\_w/o\_LT} - RMSE_{\_with\_LT})/RMSE_{\_with\_LT} \times 100]$ (in %). An increase in the *RMSE* when excluding LT from the MLR is observed almost everywhere in regions of significant trend contributions (Fig.2), especially in mid-high latitudes of the S.H. and of the N.H. in summer where it reaches 10%. This result indicates that adjusting LT improves the performance of the model and, hence, that a trend signal is well captured by IASI at a regional scale in the troposphere. From the annual model, the increase in the *RMSE* only reaches 5% at mid-high latitudes of the S.H. (data not shown). In regions of weak or non-significant trend contribution (see crosses in Fig.2), no improvement is logically found.

4 O_3 trend over 2008-2017

4.1 Annual and seasonal trends

The annual and the seasonal distributions of the fitted LT terms which are retained in the annual and the seasonal MLR models by the stepwise elimination procedure are respectively represented in Fig. 5 (a) and (b) (in DU/yr). Generally, the mid-high latitudes of both hemispheres and, more particularly, the N.H. mid-latitudes in summer reveal significant negative trends, while the tropics are mainly characterized by non-significant or weak significant trends. Even if trends in emissions have already been able to qualitatively explain measured tropospheric O_3 trends over specific regions, the magnitude and ~~the patterns of~~ the trend~~s~~ estimates considerably vary between independent measurement datasets (e.g. Cooper et al., 2014; the TOAR report – Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation, Lead Authors: A. Gaudel and O.R. Cooper – coordinated by the International Global Atmospheric Chemistry Project and available on http://www.igacproject.org/activities/TOAR and submitted to Elementa; and references therein) for the reasons discussed in Section 1 and they are not reproduced/explained by model simulations (e.g. Jonson et al., 2006; Cooper et al., 2010; Logan et al., 2012; Wilson et al., 2012; Hess et al., 2013; and references therein). As a result, ~~interpreting~~ comparing/reconciling the adjusted trends ~~at the global scale~~ with independent measurements, even on a qualitative basis, remains difficult. Nevertheless, several of the statistically significant features observed in Fig.5 show, interestingly, qualitative consistency with respect to recent published findings:

- The S.H. tropical region extending from the Amazon to tropical eastern Indian Ocean seems to indicate a general increase with, for example, a~~n~~ ~~annual~~ DJF trend of ~0.23~~13~~±0.18~~10~~ DU/yr (i.e. ~~1.20~~2.09±~~0.93~~1.70 DU over the IASI measurement period), despite the large IAV in the MLT~~TOCs~~ which characterizes the tropics and which likely explains the high frequency of non-significant trends. Enhanced O_3 levels over that region have already been analysed for previous periods (e.g. Logan et al., 1985, 1986; Fishman et al., 1991; Moxim et al., 2000; Thompson et al., 2000, 2007; Sauvage et al., 2006, 2007; Archibald et al., 2017). For instance, the large~~r~~ O_3 enhancement of ~0.36~~3~~±0.25~~3~~ DU/yr (i.e. 3.3~~1~~±2.3~~2~~ DU over the whole IASI period) stretching from southern Africa to Australia over the north-east of Madagascar during the austral winter-spring likely originates from large IAV in the subtropical jet-related stratosphere–troposphere exchanges which have been found to primarily contribute to the tropospheric O_3 trends over that region (Liu et al., 2016; 2017). Nevertheless, this finding should be mitigated by the fact that the trend value in the S.H. tropics is of the same magnitude as the *RMSE* of the regression residuals (~2-4.5 DU; see Fig.1).

- The trends over the South-East Asia are mostly non-significant and vary by season. In spring-summer, some grid cells in India, in mainland China and eastwards downwind China exhibit significant positive trends reaching ~0.45 DU/yr (i.e. ~4.2 DU over the IASI measurement period). This tends to indicate that the tropospheric O_3 increases which have been shown to mainly result~~ing~~ from a strong positive trend in the Asian emissions over the past decades (e.g. Zhao et al., 2013; Cooper et al., 2014; Zhang et al., 2016; Cohen et al., 2017; Tarasick et al., 2017; and references therein) but also from a substantial change in the stratospheric contribution (Verstraeten et al., 2015) persists through 2008-2017 despite the recent decrease in O_3 precursor emissions recorded in China after 2011 (e.g. Duncan et al., 2016; Krotkov et al., 2016; Miyazaki et al., 2017; Van der A et al. 2017). This would indicate that this decrease is probably too recent/weak to recover the 2008 O_3 levels over the entire region. Note, however, that this finding has to be taken carefully given the large model residuals (*RMSE* of ~2-4 DU; cfr Section 3, Fig.1) over that region. Finally, the large uncertainty in trend estimates~~ion~~ over the South-East Asia might reflect~~s~~ the large IAV in the biomass-burning emissions and lightning NO_x sources, in addition to the recent changes in emissions.

- The mid- and high latitudes of the S.H. show clear patterns of negative trends, all over the year and in a more pronounced manner during winter-spring, with larger amplitudes than those of the *RMSE* values (~-0.33~~5~~±0.14~~1~~ DU/yr on average in the 35°S-65°S band; i.e. a trend amplitude of ~|3.~~3~~1|±1.~~0~~3 DU over the studied period *vs* a *RMSE* value of ~2.5 DU). These significant negative trends in the S.H. are hard to explain but, considering the stratospheric contribution into the tropospheric columns (natural and artificial due to the limited IASI vertical sensitivity) in the mid-high latitudes of the S.H. (~40-60%; see supplementary materials in Wespes et al., 2016) and the negative significant trends previously reported from IASI in the UTLS/low stratosphere in the 30°S-50°S band, they could be in line with those derived by Zeng et al. (2017) in the UTLS for a clean rural site of the S.H. (Lauder, New Zealand), ~~and~~ which mainly originate from increasing tropopause height and O_3 depleting substances.

~~-~~ In the N.H., a band-like pattern of negative trends is observed in the 40°N-75°N latitudes covering Europe and North America, especially during summer. Averaged annual trend of –0.31~~7~~±0.17~~8~~ DU/yr and summer trend of –0.47±0.22~~16~~ DU/yr (i.e. ~~–3.42~~2.87±~~0.65~~1.57 DU and -4.36~~1~~±2.02~~1.47~~ DU, respectively, from January 2008 to May 2017) are estimated in that latitudinal band. These trend values are significantly larger than the *RMSE* of the MLR model (<3.5 DU in JJA; cfr Section 3, Fig.1). Interestingly, both the annual and summer trends are amplified ~~in comparison with~~relative to the ones calculated in the mid-latitudes of the N.H. over the 2008-2013 period of IASI measurements (-0.19±0.05 DU/yr and -0.30±0.10 DU/yr for the annual and the summer trends, respectively, calculated in the 30°N-50°N band; see Wespes et al. (2016)). This finding is line with ~~is in agreement with~~ previous studies which point out a possible leveling off of tropospheric O_3 in summer due to the decline of anthropogenic O_3 precursor emissions observed since 2010-2011 in North America, in Western Europe and also in some regions of China (e.g. Cooper et al., 2010; 2012; Logan et al., 2012; Parrish et al., 2012; Oltmans et al., 2013; Simon et al., 2015; Archibald et al., 2017; Miyazaki et al., 2017). It even goes a step further by suggesting a possible decrease in the tropospheric O_3 levels. Archibald et al. (2017) recently reported a net decrease of ~5% in the global anthropogenic NO_x emissions in the 30°N-90°N latitude band, which is consistent with the annual significant negative trend of -0.2732±0.158 DU/yr for O_3 estimated from IASI in that band. We should also note that, even if these latitudes are characterized by the lowest stratospheric contribution (~30-45%; see supplementary materials in Wespes et al., 2016), it might partly mask/attenuate the variability in the tropospheric O_3 levels.

4.2 Expected year for trend detection

In this section, we further verify that it is indeed possible to infer, from the studied IASI period, the significant negative trend derived in the 40°N-75°N band in summer (~|0.5| DU/yr on average, see Section 4.1) by determining the expected year from which such a trend amplitude would be detectable at a global scale. This is achieved by estimating the minimum duration (with probability 0.90) of the IASI O_3 measurements that would be required to detect a trend of a specified magnitude, and its 95% confidence level, following the formalism developed in Tiao et al. (1990) and in Weatherhead et al. (1998):

$$N^* \approx \left[\frac{3.3 \cdot \sigma_\varepsilon}{|\tau_{yr}|} \cdot \sqrt{\frac{1+\Phi}{1-\Phi}} \right]^{2/3} \qquad \text{Eq (1)}$$

$$CL_{N^*} = \left[N^* \cdot e^{-B}; N^* \cdot e^{+B} \right] \qquad \text{Eq (2)}$$

Where N^* is the number of the required years, σ_ε is the standard deviation of the autoregressive noise residual ε_t, τ_{yr} is the magnitude of the trend per year, Φ is the lag-1 autocorrelation of the noise. The magnitude of the variation and of the autocorrelation in the noise residuals are taken into account for a better precision on the trend estimate. Given that large variance (σ_ε^2) and large positive autocorrelation Φ of the noise induce small signal-to-noise ratio and long trend-like segments in the dataset, respectively, these two parameters increase the number of years that would be required for detecting a specified trend. CL_{N^*} is the 95% confidence limits which is not symmetric around N^* and depends on B, an estimated uncertainty factor calculated as $\frac{4}{3\sqrt{D}}\sqrt{\frac{1+\Phi}{1-\Phi}}$, with D the number of days in the IASI datasets, which accounts for the uncertainty in Φ (the uncertainty in σ_ε being negligible given that only a few years of data are needed to estimate it; cfr Weatherhead et al. (1998)). As a result, based on the available IASI-A and proxies datasets and assuming that the MLR model used in this study is accurate, we estimate, in Fig. 6 (a) and (b), the expected year when an O_3 trend amplitude of |5| DU per decade (i.e. τ_{yr} = |0.5| DU/yr which corresponds to the averaged absolute value of the fitted negative trends in the N.H. summer; see Fig.5b) is detectable, and its associated maximal confidence limit, respectively. The results in

Fig. 6 clearly demonstrates the possibility to infer, from the available IASI dataset, such significant trends in the mid- and high latitudes of the N.H. in summer and fall (trend detectable from ~2016-2017 with an uncertainty of ~6-9 months; cfr Fig.6b). On the contrary, the tropical regions and the N.H. in winter-spring would require additional ~ 6 months to 6 years of measurements to detect an amplitude of |0.5| DU/yr (trend significant only from ~ 2017 – 2023 or after depending on the location and the season). Note also that the strongest negative trends (up to -0.85 DU/yr, i.e. τ_{yr} = |0.85| DU/yr, see Fig.5b) observed in specific regions of the N.H. mid-latitudes would only require ~6 years of IASI measurements for being detected. The mid- and high latitudes of the S.H. would require the shortest period of IASI measurement for detecting a specified trend, with only ~7 years ± 6 months of IASI measurements to detect a |0.5| DU/yr trend (trend detectable from ~2015). That band-like pattern in the S.H. corresponds to the region with the weakest IAV and contribution from large-scale dynamical modes of variability in the IASI MLT columns ~~TOCs~~ (see Section 3, Fig.1 and 2), which translates into small σ_{ε}^{2} and Φ. Note however that an additional few months of IASI data are required to confirm the smaller negative trend of ~-0.35 DU/yr measured on average in the S.H. (see Fig.5; a period ~9 years ± 6 months being necessary to detect a trend amplitude of |3.5| DU/dec). Given that large σ_{ε} means large noise residual in the IASI data, the regions of short or long required measurement period coincide, as expected, well with the small or high *RMSE* values of the regression residuals (see Section 3, Fig. 1).

The regions of the longest measurement periods required in the tropics for a trend detection (up to ~16 years of IASI data) correspond to known patterns of widespread high O_3: (a) above intense biomass burnings in Amazonia and eastwards across tropical Atlantic (Logan et al., 1986; Fishman et al., 1991; Moxim et al., 2000; Thompson et al., 2000, 2007; Sauvage et al., 2007), (b) eastwards Africa across the South Indian Ocean which is subject to large variations in the stratospheric influences during the winter-spring austral period (JJA-SON) (Liu et al., 2016; 2017), (c) Eastwards Africa across the North Indian Ocean to India likely due to large lightning NO_x emissions above central Africa during the wet season associated with the northeastward jet conducting a so-called "O_3 river" (Tocquer et al., 2015) and (d) above regions of positive ENSO "chemical" effect in Equatorial Asia/Australia and eastwards above northern and southern tropical regions (Wespes et al., 2016) explained by reduced rainfalls and biomass fires during El Niño conditions (e.g. Worden et al., 2013). In fact, most of these patterns (a, b and d) are closely connected with strong El- Niño events which extend the duration of the dry season and cause severe droughts, producing intense biomass burning emissions, for instance, over South America (e.g. Chen et al., 2011; Lewis et al., 2011) and South Asia/Australia (e.g. Oman et al., 2013; Valks et al., 2014; Ziemke et al., 2015), and which ~~perturb~~ alter the tropospheric circulation by increasing the transport of stratospheric O_3 into the troposphere (e.g. Voulgarakis et al., 2011; Neu et al., 2014) and the transport of biomass burning air masses to the Indian Ocean (Zhang et al., 2012). In summary, these large-scale indirect ENSO-related variations in tropospheric O_3 and the lightning NO_x impact on O_3, which are not accounted for in the MLR by specific representative proxies, are misrepresented in the regression models. They induce large noise residuals, i.e. large σ_{ε}, and, hence, extends the time period needed to detect a trend of any given magnitude.

Figure 6, finally, suggests that a long duration is also required, especially in summer, above and east of China to quantify the anthropogenic impact on the local ~~TOC~~ changes in the MLT: additional 3±1.5 years or 5±1.5 years for a given |5| or |3.5| DU/dec trend are respectively calculated. This result could be explained by large perturbations in ~~TOCs~~the MLT columns induced by recent decreases after decades of almost constant increases in surface emissions in China (e.g. Cohen et al., 2017; Miyazaki et al., 2017).

4.3 Multi-linear *vs* single linear model

Even if MLR have already been used for extracting trends in stratospheric and total O_3 columns (e.g. Mäder et al., 2007; Frossard et al., 2013; Rieder et al., 2013; Knibbe et al., 2014), single linear regressions (SLR) without discriminating the natural (chemical and dynamical) factors describing the O_3 variability are still commonly used (e.g. Cooper et al., 2014; the TOAR report – Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation, Lead Authors: A. Gaudel and O.R. Cooper – submitted to Elementa, and references therein). They are, however, suspected to contribute to trend biases retrieved between independent measurements. In addition to the time-varying instrumental biases, trend biases can also be related to differences in the spatial and the temporal samplings (e.g. Doughty et al., 2011; Saunois et al., 2012; Lin et al., 2015), in the measurement period, in the upper boundary of the O_3 columns, in the algorithm and in the vertical sensitivity of the measurements. The latter artificially alters the characteristics of the sounded layer by contaminations from the above and the below layers leading to a mixing of the trend but also of the natural characteristics originating from these different layers (e.g. troposphere and stratosphere). The differences in the studied period, in the tropopause definition and in the spatio-temporal sampling might also imply differences in the natural influence on the measured O_3 variations. While the impact of the natural contribution is taken into account in the MLR model, it might introduce an additional bias in the trend determined from SLR, making further challenging to compare trends estimated from a series of inhomogeneous independent measurements. 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 where 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 the trend biases between the datasets 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 Tthe improvement limitation in using a MSLR instead of a MSLR model for determining more accurate/realistic trends is explored here by comparing the seasonal distributions of the trends estimated from MLR (see Fig. 5 (b) in Section 4.1.) and from SLR (presented in Fig.7). Note that the constant term in the SLR is split into two components (covering the periods before and after the September 2010 "jump") to take account of the observed "jump" (see Section 2). The highest differences in the fitted trends derived from the two methods are found in the tropics and in some regions of the mid-latitudes of the N.H. They likely result from overlaps between the LT term and other covariates. For instance, the regions of thewith high significant SLR trends (~0.3-0.56 DU/yr over the tropical western and middle Pacific) during the period extending from September to May match the regions of with strong El Niño/Southern Oscillation influence (cfr Fig. 8 and 12 in Wespes et al., 2016). On the contrary, the MLR model lends generally weak significant negative or non-significant trends in the Pacific Niño region during that period and it would even need additional ~3 to 4 years of IASI measurements for detecting the fitted SLR trends (see Section above). The effect of ENSO in overestimating the fitted SLR trend is further illustrated on Fig. 8 which represents the time series of O_3 observed by IASI and adjusted by the annual MLR model (top row) along with the deseasonalized times series (middle row) and the fitted SLR and MLR trends (bottom row). The fitted signal of the ENSO proxy from the MLR regression (calculated as $x_j X_{norm,j}$ following Wespes et al. (2017)) is also represented (bottom row). That example clearly shows that the ENSO influence is considerably compensated by the adjustment of the linear trend in the SLR regression (annual trend of -0.2948±0.0286 DU/yr from SLR vs -0.1323±0.092 16 DU/yr from MLR for that example). Finally, differences between the SLR and the MLR models are also observed in the region ofwith strong positive NAO influence over the Icelandic/Arctic region during MAM (see Wespes et al. (2016) for a description of the NAO-related O_3 changes). On the contrary, the sub-tropical S.H. exhibit negative trends from both the SLR and the MLR. It results from the weak natural IAV and contributions in tropospheric O_3 above that region (see Section 3, Fig. 1 and 2), which, hence, limits the compensation effects.

In summary, if despite the fact that considering a long period of measurements is usually recommended in SLR study to pass over overcome the dynamical cycles and, hence, to help in discriminating their influences from trends, we show that it is not accurate enough considering that some dynamics have irregular or no particular periodicity (e.g. NAO, ENSO) it is not accurate enough. Furthermore, accurate satellite measurements of tropospheric O_3 at a global scale are quite recent, limiting the period of available and comparable datasets (e.g. Payne et al., 2017). As a consequence, we support here that using a reliable multivariate regression model based on geophysical parameters and adapted for each specific sounded layer is a robust method for differentiating a “true” trend from any other sources of variability and, hence, that it should help in resolving trend differences between independent datasets.

4.4 Continental influence

In this section, we use the capabilities of IASI to simultaneously measure O_3 and CO in order (1) to differentiate tropospheric and stratospheric air masses, (2) to identify the regions influenced by the continental export/intercontinental transport of O_3 pollution and (3) to evaluate that continental influence on tropospheric O_3 trends as observed by IASI. Similar tracer correlations between CO and O_3 have already been used to give insight into the photochemical O_3 enhancement in air pollution transport (e.g. Parrish et al., 1993; Bertschi et al., 2005). However, there are only a few studies using global satellite data for this purpose (Zhang et al., 2006; Voulgarakis et al., 2010; Kim et al., 2013) and the analysis of the O_3-CO relationship for better understanding the origin of O_3 trends in the troposphere has, to the best of our knowledge, never been explored.

~~We provide in~~ Fig.9a and 9b show the seasonal patterns of the O_3-CO correlations (referred as R_{O3-CO}) and of the dO_3/dCO regression slopes calculated in the troposphere (from the surface to 300 hPa) over the studied IASI period (January 2008 – May 2017). The dO_3/dCO regression slopes, which represent the so-called O_3-CO enhancement ratio, is used to evaluate the photochemical O_3 production in continental outflow regions. The R_{O3-CO} and the dO_3/dCO distributions are similar and clearly show regional and seasonal differences in the strength of the O_3-CO relationships. ~~Regions of~~ The patterns of positive and negative correlations allows to discriminate ~~air masses~~the outflow regions characterized by photochemical O_3 production from precursors (including CO) or CO destruction (both identified by positive R_{O3-CO}) from ~~those~~ the regions characterized by O_3 loss (chemical destruction or surface deposition) or by strong stratospheric contaminations (both identified by negative R_{O3-CO}). Negative R_{O3-CO} and dO_3/dCO are measured ~~in the~~at high latitudes of both hemispheres all over the year, but more specifically at high latitudes of the S.H. in summer-fall (with R_{O3-CO} <-0.25 on averages in DJF and MMA). ~~If the~~Given that high latitudes experience more O_3 destruction than the low latitudes due to a lack of sunlight, the negative correlations for the high latitude regions might also reflect air masses originating from/characterizing the stratosphere due to natural intrusion or to artificial mixing with the troposphere introduced by the limited vertical sensitivity of IASI in the highest latitudes (stratospheric contribution varying between ~40% and 65%; see supplementary materials in Wespes et al., 2016). These processes are likely at the origin of the band-like pattern of negative trends in the S.H. discussed in Sections 3 and 4.1. Negative R_{O3-CO} and dO_3/dCO are also found above the Caribbean, the Arabic Peninsula and the North Indian Ocean in JJA/SON and the South Atlantic in DJF. They are in line with Kim et al. (2013) and they likely reflect the influence of lightning NO_x which produce O_3 but also OH oxidizing CO (e.g. Sauvage et al., 2007; Labrador et al., 2004).

Strong positive correlations are identified in both R_{O3-CO} and dO_3/dCO patterns over the tropical regions and for mid-latitudes of both Hemispheres during the peak of photochemistry in summer. Maxima (R_{O3-CO} >0.8 and dO_3/dCO>0.5) are detected in continental pollution outflow regions in the N.H., especially downwind South-East Asia and over the South Africa/Amazonia/South Atlantic region. These O_3-CO correlation patterns from IASI are fully consistent with those measured by TES (Zhang et al., 2006; 2008; Voulgarakis et al., 2010) and by OMI/AIRS (Kim et al., 2013), which have been interpreted with global CTM's as originating from Asian pollution influence and from combustion sources including biomass burning, respectively. The high positive R_{O3-CO} found in JJA at mid-latitudes of the N.H. are detected in a lesser extent in DJF reflecting the decreasing photochemistry. It is also worth ~~to~~ pointing out in Fig. 9 the clear hemispheric differences in the R_{O3-CO} and dO_3/dCO values at mid-high latitudes and, more particularly, the shift of positive R_{O3-CO} and dO_3/dCO towards higher latitudes of the N.H. during summer (e.g. R_{O3-CO}= 0.24 in summer *vs* 0.038 in spring in the 35°N-55°N band). As a consequence, these results suggest that the band-like pattern of negative trends measured by IASI in summer might substantially reflect the continental pollution influence and, hence, that it could result from the decline of anthropogenic O_3 precursor emissions. Nevertheless, interpreting O_3-CO correlations in the free troposphere, especially in photochemically aged pollution plumes far from the emission sources towards the highest latitudes, remains complicated by the mixing of the continental combustion outflow with stratospheric air masses, in addition to the background dynamic and photochemistry (e.g. Liang et al., 2007).

Finally, we also provide in Fig.9c ~~global~~ the seasonal patterns of O_3-CO covariances (COV_{O3-CO}). They confirm the band-like pattern of the weak natural variation captured in the S.H. mid-latitudes ~~S.H.~~ (see Sections 3 and 4.1) and help identifying the region downwind East China, the northern mid-latitudes ~~pollution~~ outflow region and the South tropical region as the ones with the highest pollution variability, in addition to the strongest O_3-CO correlations. ~~Finally~~To conclude, the particularly strong positive O_3-CO relationship in terms of R_{O3-CO}, dO_3/dCO and COV_{O3-CO} measured over and downwind North-East India/East China in summer in comparison with the ones measured downwind East US and over Europe indicate that ~~the~~ South-East Asia experiences the most of the intense pollution episodes of the N. with the largest O_3-CO variability (COV_{O3-CO} > 40×10^{33} $mol^2.cm^{-4}$) ~~with~~and the largest O_3 enhancement (dO_3/dCO > 0.5) over the last decade. ~~,~~ The strong O_3-CO relationship in that region ~~explaining~~ is associated with the significant increase that is detected in the IASI O_3 levels downwind East of Asia (see Section 4.1) despite the net decrease in O_3 precursor emissions recorded in China after 2011 (e.g. Cohen et al., 2017; Miyazaki et al., 2017).

5 Conclusions

In this study, we have explored, for the first time, the possibility to infer significant trends in tropospheric O_3 from the first ~10 years (January 2008 – May 2017) of ~~the~~ IASI daily measurements at a global scale. To this end, MLR analyses have been performed by applying a multivariate regression model (annual and seasonal formulations), including a linear trend term in addition to chemical and dynamical proxies, on gridded mean tropospheric ozone time series. This work follows on the analysis of the main dynamical drivers of O_3 variations measured by IASI, which was recently published in Wespes et al. (2017). We have first verified the performance of the MLR models in explaining the variations in daily time series over the whole studied period. In particular, we have shown that the model reproduces a large part of the O_3 variations (>70%) with small residuals errors (*RMSE* of ~10%) ~~in the~~at northern latitudes in summer. We have then performed O_3 trend sensitivity tests to verify the possibility to capture trend characteristics independently from natural variations. Despite the weak contribution ~~from~~ of~~the~~ trends to the total variation in ~~TOCs~~ the MLT O_3 columns at ~~the~~ aglobal scale, the results demonstrate the possibility to discriminate significant trends from the explanatory variables, especially in summer at mid-high latitudes of the N.H. (~45°N-70°N) where the contribution and the sensitivity of ~~the~~ trends are the largest (contribution of ~30-50% and a ~10% increase in the *RMSE* excluding the LT in the model). We then focused on the interpretation of the global trend estimates. We have found an interesting significant positive trends in the S.H. tropical region extending from the Amazon to the tropical eastern Indian Ocean and over South-East Asia in spring-summer which should however be carefully considered given the high *RMSE* of the regression residuals in these regions. The MLR analysis reveals a band-like pattern of high significant negative trends in the N.H. mid-high latitudes in summer (–0.47±0.16 DU/yr on average in the 45°N-70°N band). The statistical significance of such trend estimates is further verified by estimating, based on the autocorrelation and on the variance of the noise residuals, the minimum number of years of IASI measurements that are required to detect a trend of a |5| DU/dec magnitude. The results clearly demonstrate the possibility to determine such a trend amplitude from the available IASI dataset and the used MLR model at northern mid-high latitudes in summer, while much larger measurement periods are necessary elsewhere. In particular, the regions ~~of~~with the longest required period ~~highlight~~, in the tropics, highlight a series of known processes that are closely related to the El-Niño dynamic, which underlies the lack of associated parameterizations in the MLR model. The importance of using reliable MLR models in understanding large-scale O_3 variations and in determining trends is further explored by comparing the trends inferred from MLR and from SLR, the latter being still commonly used by the international community. The comparison has clearly highlighted the gain of MLR in attributing the trend-like segments in natural variations, such as ENSO, to the right processes and, hence, in avoiding misinterpretation of "apparent" trends in the measurement datasets. 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. Finally, by exploiting the simultaneous and vertically-resolved O_3 and CO measurements from IASI, we have provided and used the O_3-CO correlations in the troposphere to help ~~in~~ determining the origins/characteristics of ~~the air masses with~~ patterns of negative or positive trends. The distributions have allowed us to identify, in particular, strong positive O_3-CO correlations, regression slopes and covariance in the N.H. mid-latitudes and northward during summer, which suggests a continental pollution influence in the N.H. band-like pattern of high significant negative trends recorded by IASI and, hence, a direct effect of the policy measures taken to reduce emissions of O_3 precursor species.

This study supports overall the importance of using (1) high density and long term homogenized satellite records, such as those provided by IASI, and (2) complex models with predictor functions that describe the O_3-regressors dependencies for a more accurate~~ly~~ determination~~ing~~ of trends in tropospheric O_3 - as required by the scientific community, e.g. in the Intergovernmental Panel on Climate Change (IPCC, 2013) - and for further resolving trend biases between independent datasets (Payne et al., 2017; the TOAR report – Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation, Lead Authors: A. Gaudel and O.R. Cooper). Determination, with IASI, of robust trends ~~at the global scale~~ in tropospheric O_3 at the global scale will be achievable in the near future by merging the homogeneous O_3 profiles from the three successive instruments onboard Metop-A (2006); -B (2012) and –C (2018) platforms and from the IASI-Next Generation instrument onboard the Metop Second Generation series of satellites (Clerbaux and Crevoisier, 2013; Crevoisier et al., 2014). A long record of tropospheric O_3 measurements will be also assured by the Cross-track Infrared Sounder (CrIS) onboard the Joint Polar Satellite System series of satellites.

Acknowledgments

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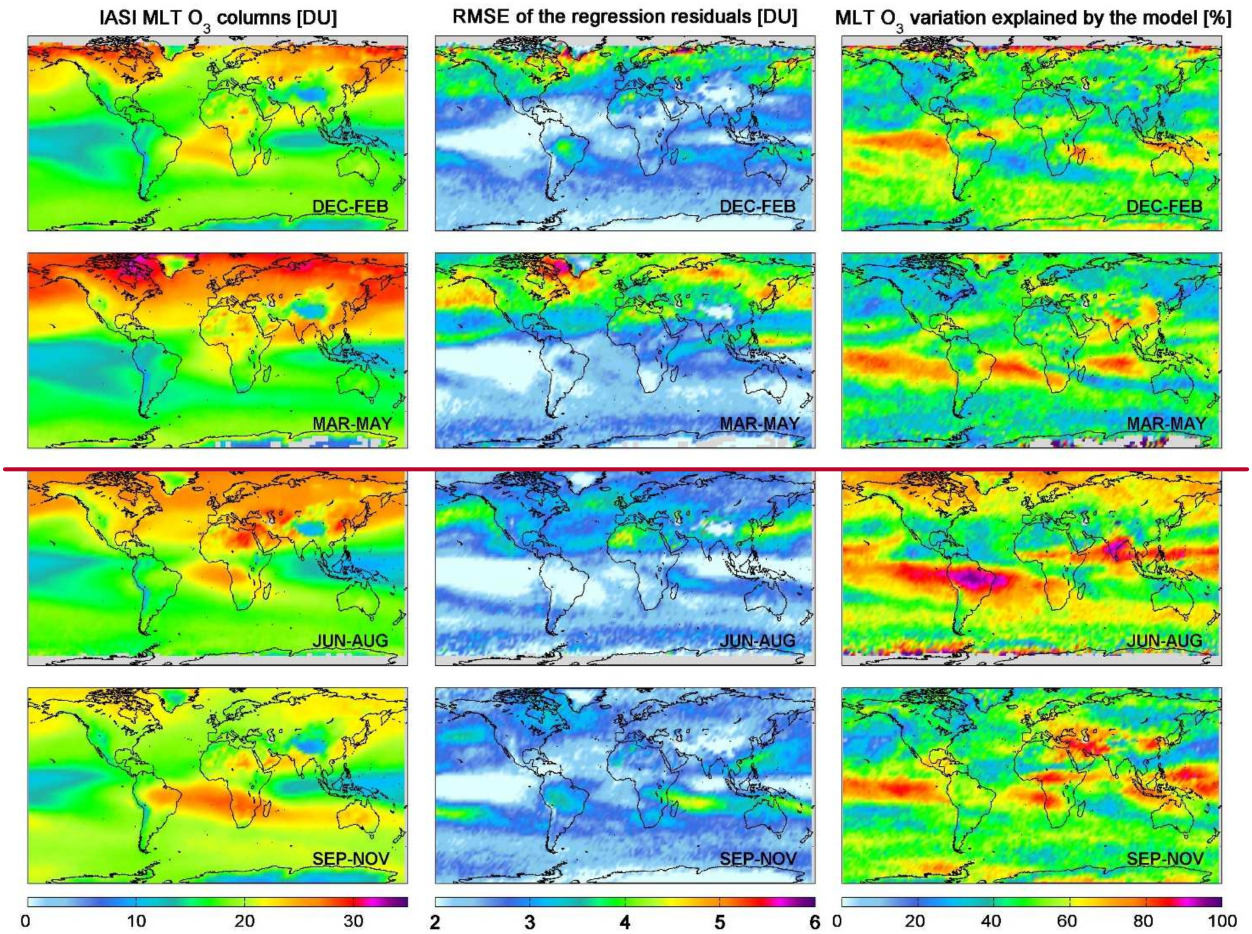
Figure captions



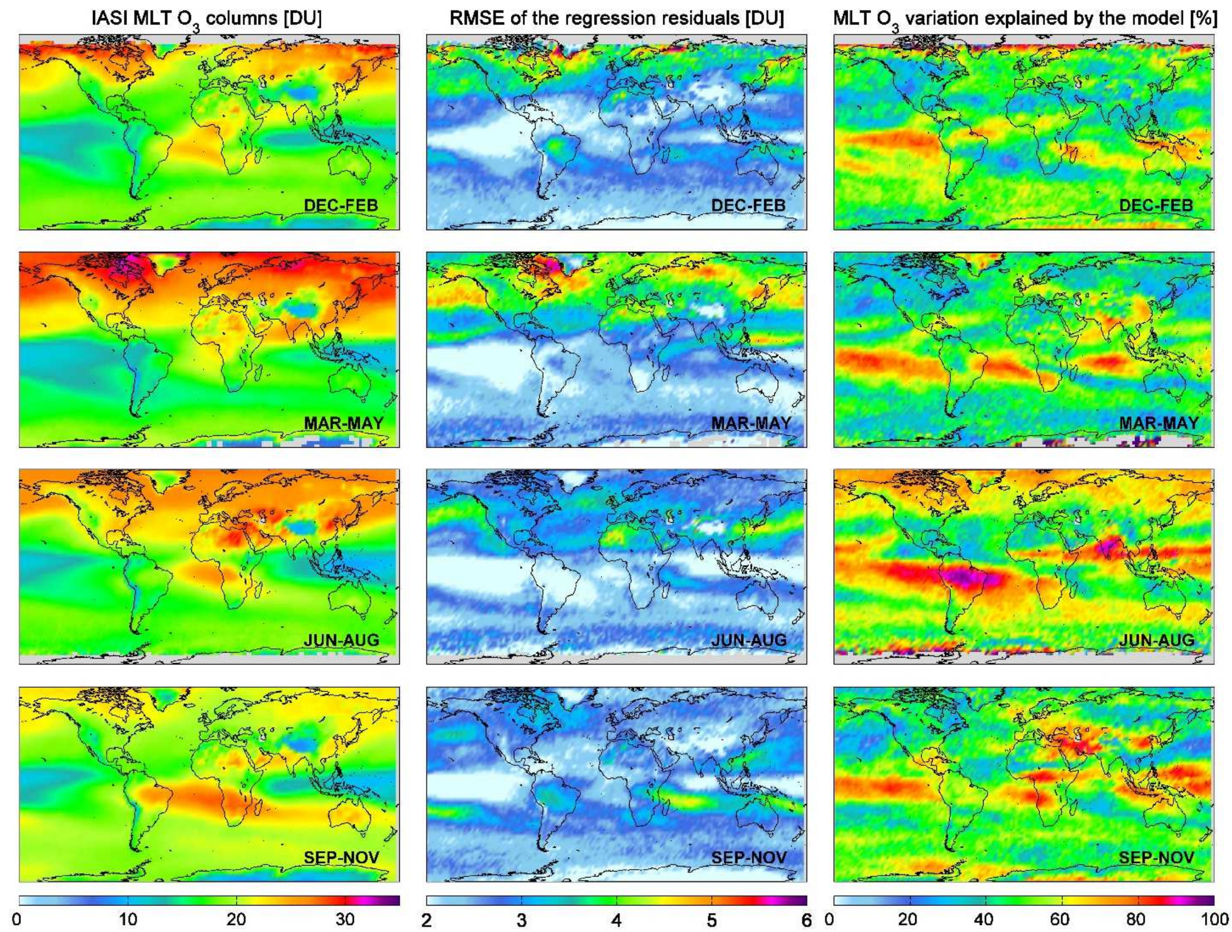
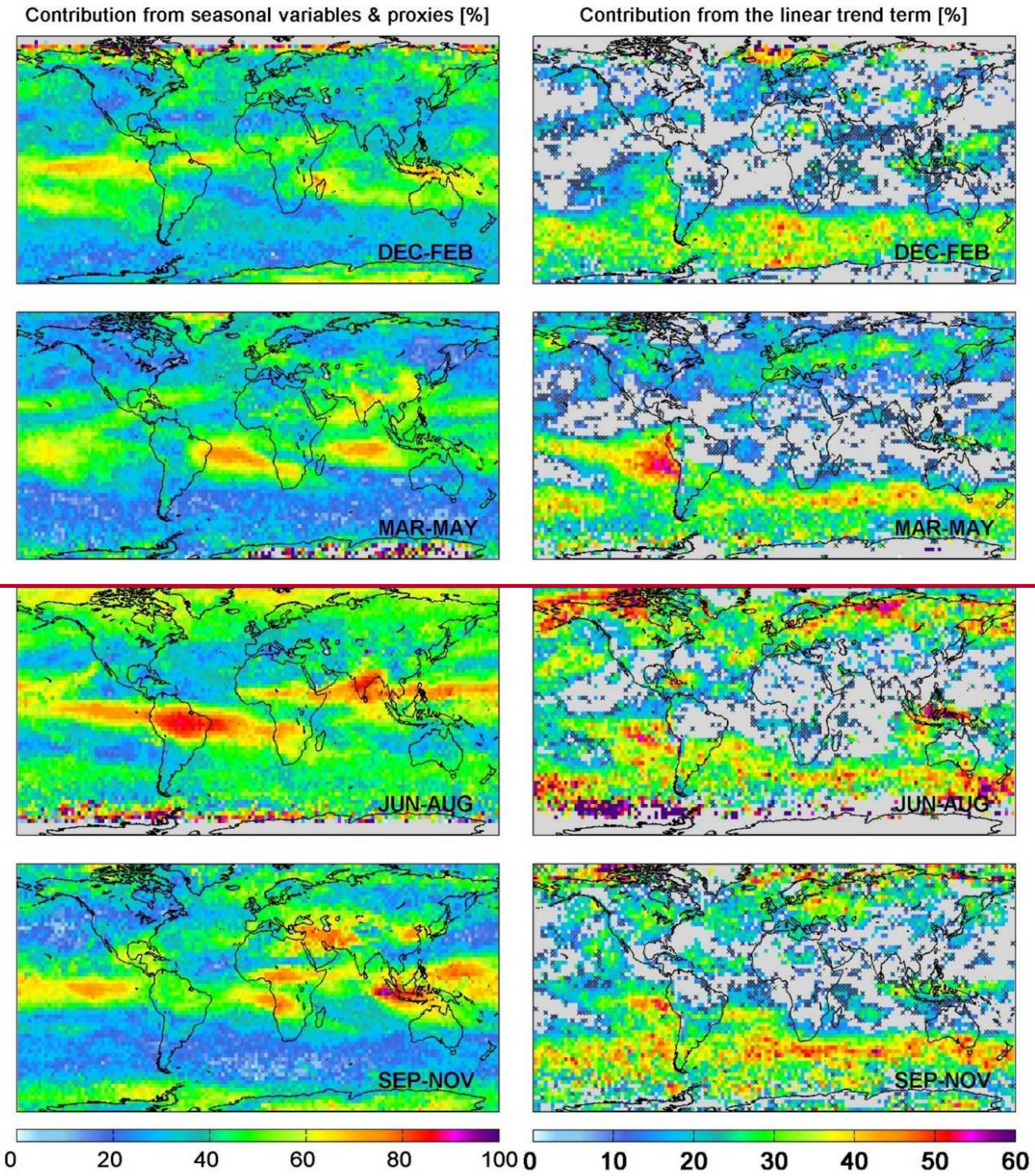


Fig.1. Seasonal distribution of O_3 tropospheric columns (in DU, integrated from ground to 300hPa) measured by IASI and averaged over January 2008 – May 2017 (left panel), of the *RMSE* of the regression fits (in DU, middle panel) and of the fraction of the variation in IASI data explained by the regression model, calculated as $\left[100\times\left(\sigma\left(O_3^{Fitted\_model}(t)\right)/\sigma\left(O_3(t)\right)\right)\right]$ (in %, right panel). Data are averaged over a 2.5°x2.5° grid box.


Contribution from seasonal variables & proxies [%]
Contribution from the linear trend term [%]
DEC-FEB
DEC-FEB
MAR-MAY
MAR-MAY
JUN-AUG
JUN-AUG
SEP-NOV
SEP-NOV
0 20 40 60 80 100
0 10 20 30 40 50 60

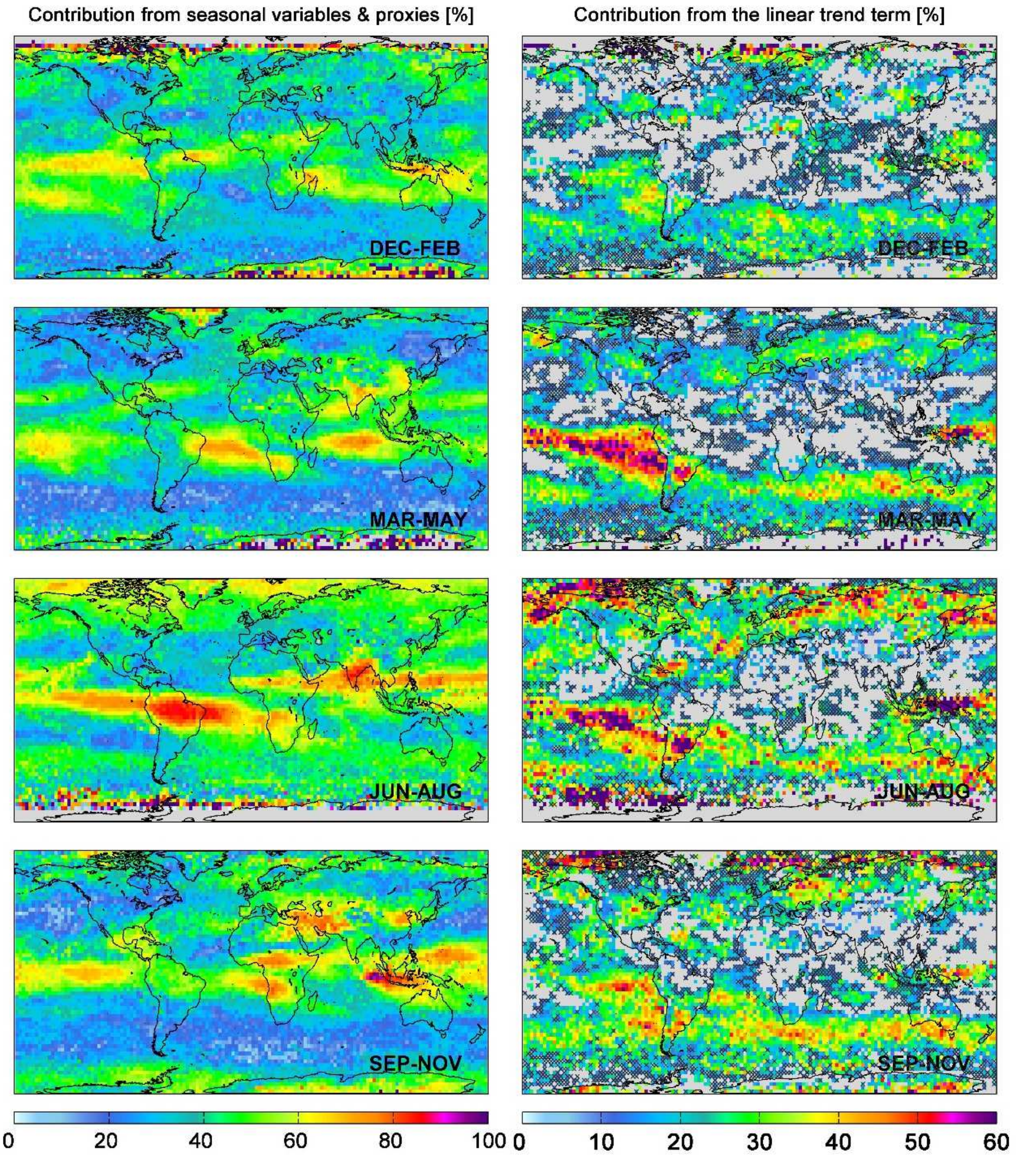
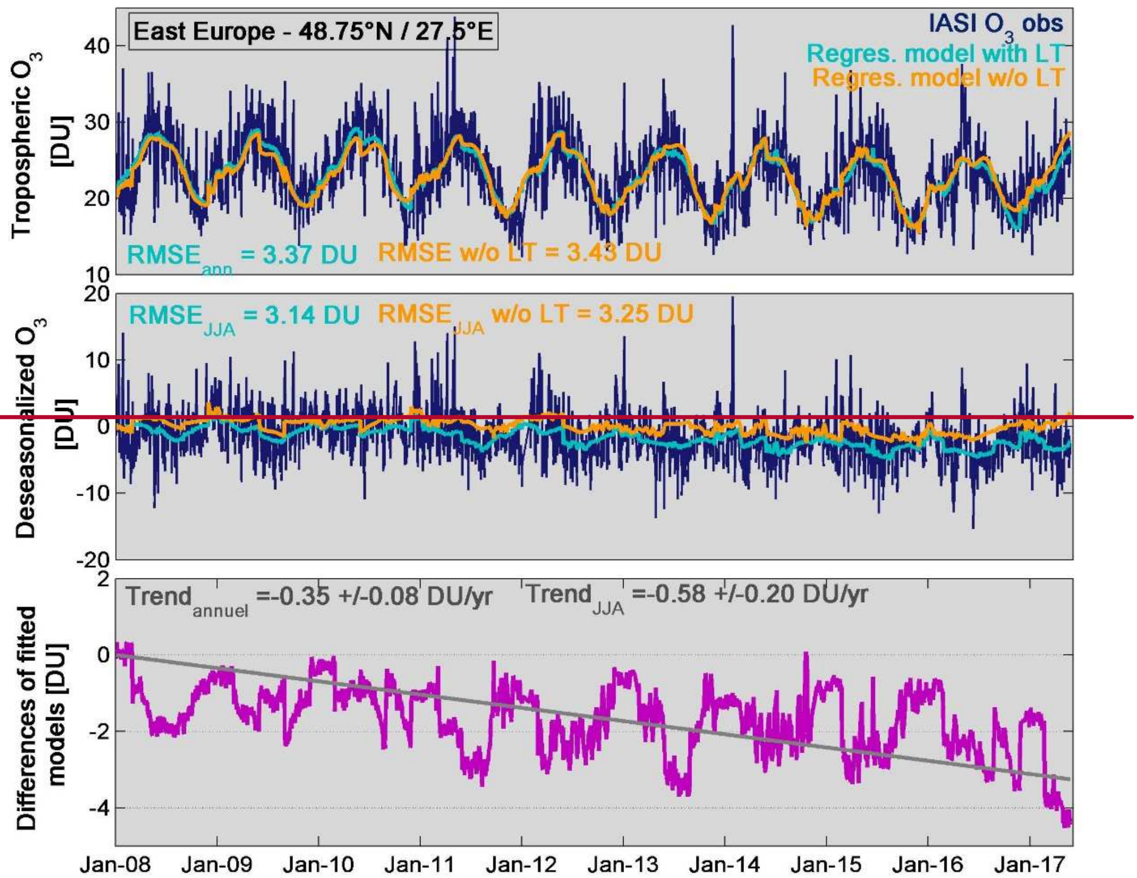


Fig.2. Seasonal distributions of the contribution from the seasonal and explanatory variables into the IASI O_3 variations estimated as $\left[100\times\sigma\left(\sum_{n=1;j=2}^{4;m}[a_n;b_n;x_j][\cos(n\omega t);\sin(n\omega t);X_{norm,j}]\right)\Big/\sigma(O_3(t))\right]$ (in %, left panels) and of the contribution from the linear trend calculated as $\lfloor 100\times\sigma(x_{j=1}\cdot trend)/\sigma(O_3(t))\rfloor$ (in %, right panels). The grey areas and crosses refer to the non-significant grid cells in the 95% confidence limits (2σ level). Note that the scales are different.


East Europe - 48.75°N / 27.5°E
IASI O3 obs
Regres. model with LT
Regres. model w/o LT
Tropospheric O3 [DU]
RMSEann = 3.37 DU
RMSE w/o LT = 3.43 DU
Deseasonalized O3 [DU]
RMSEJJA = 3.14 DU
RMSEJJA w/o LT = 3.25 DU
Differences of fitted models [DU]
Trendannuel =-0.35 +/-0.08 DU/yr
TrendJJA =-0.58 +/-0.20 DU/yr
Jan-08
Jan-09
Jan-10
Jan-11
Jan-12
Jan-13
Jan-14
Jan-15
Jan-16
Jan-17

1273

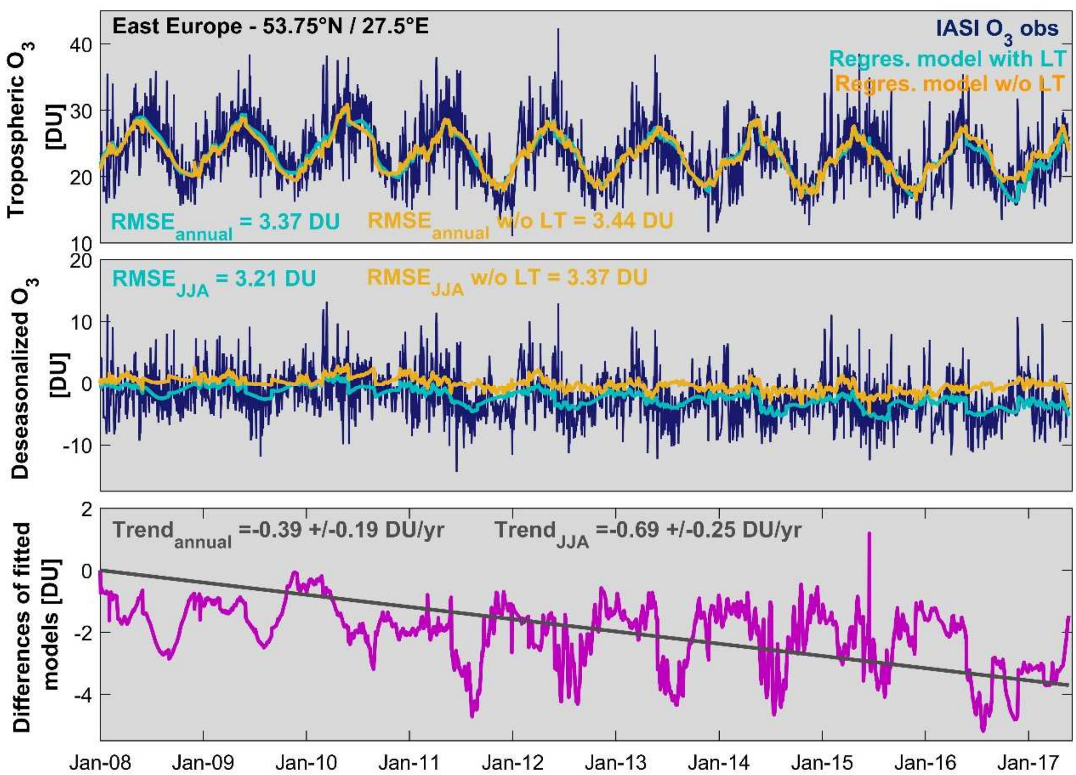
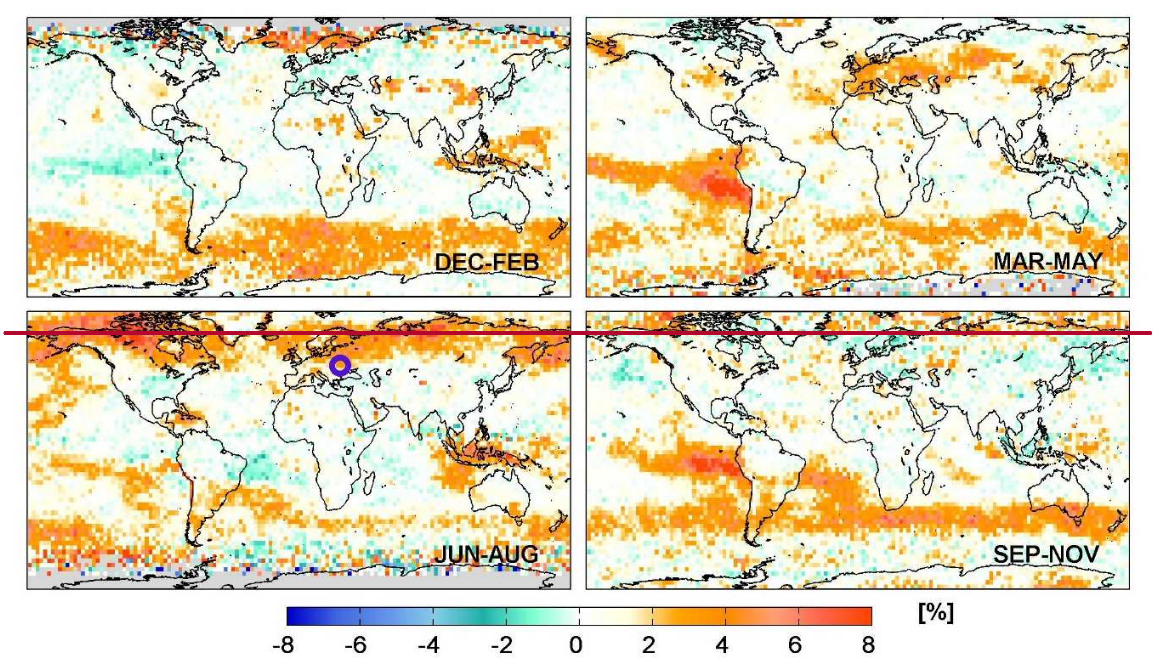


Fig.3. Examples of daily time series of IASI O_3 measurements (dark blue) and of the fitted seasonal regression models with (light blue) and without (orange) the linear term in the troposphere (1^{st} row). Daily time series of the deseasonalised O_3 (observations and regression models; 2^{d} row) and of the difference of the fitted models with and without the linear trend term as well as the adjusted annual trend (pink and grey lines, respectively; 3^{d} row) (given in DU). The *RMSE* (annual and for the JJA period in DU) and the trend values (annual and for the JJA period in DU/yr) are also indicated.



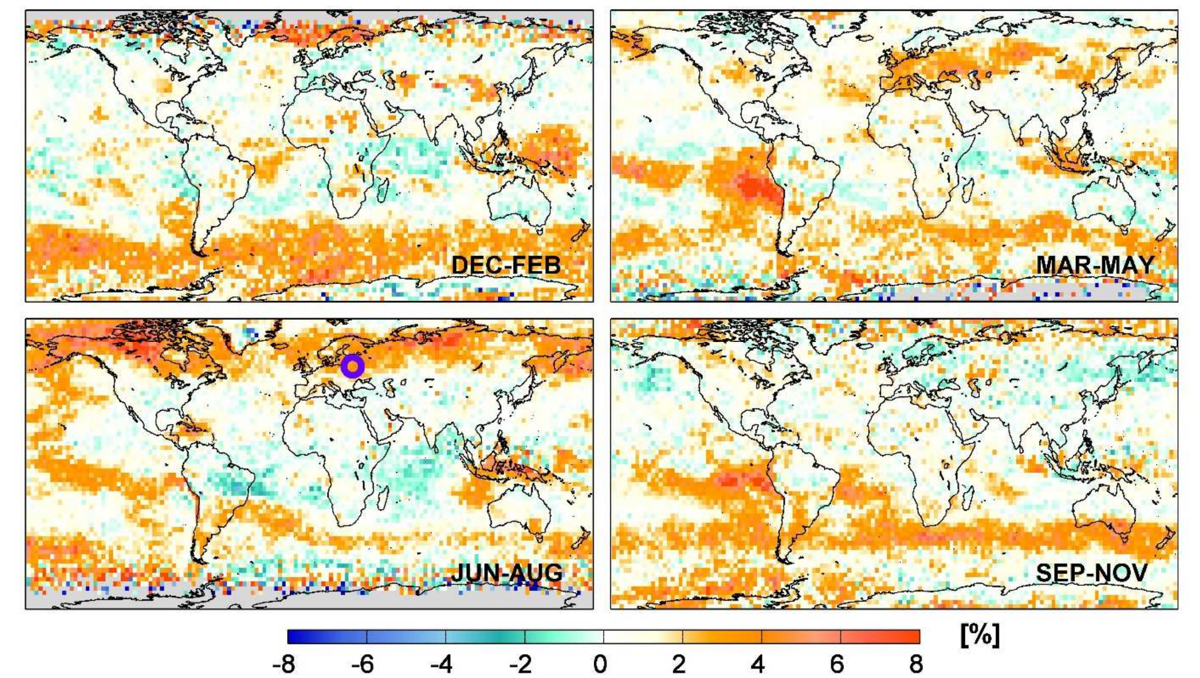
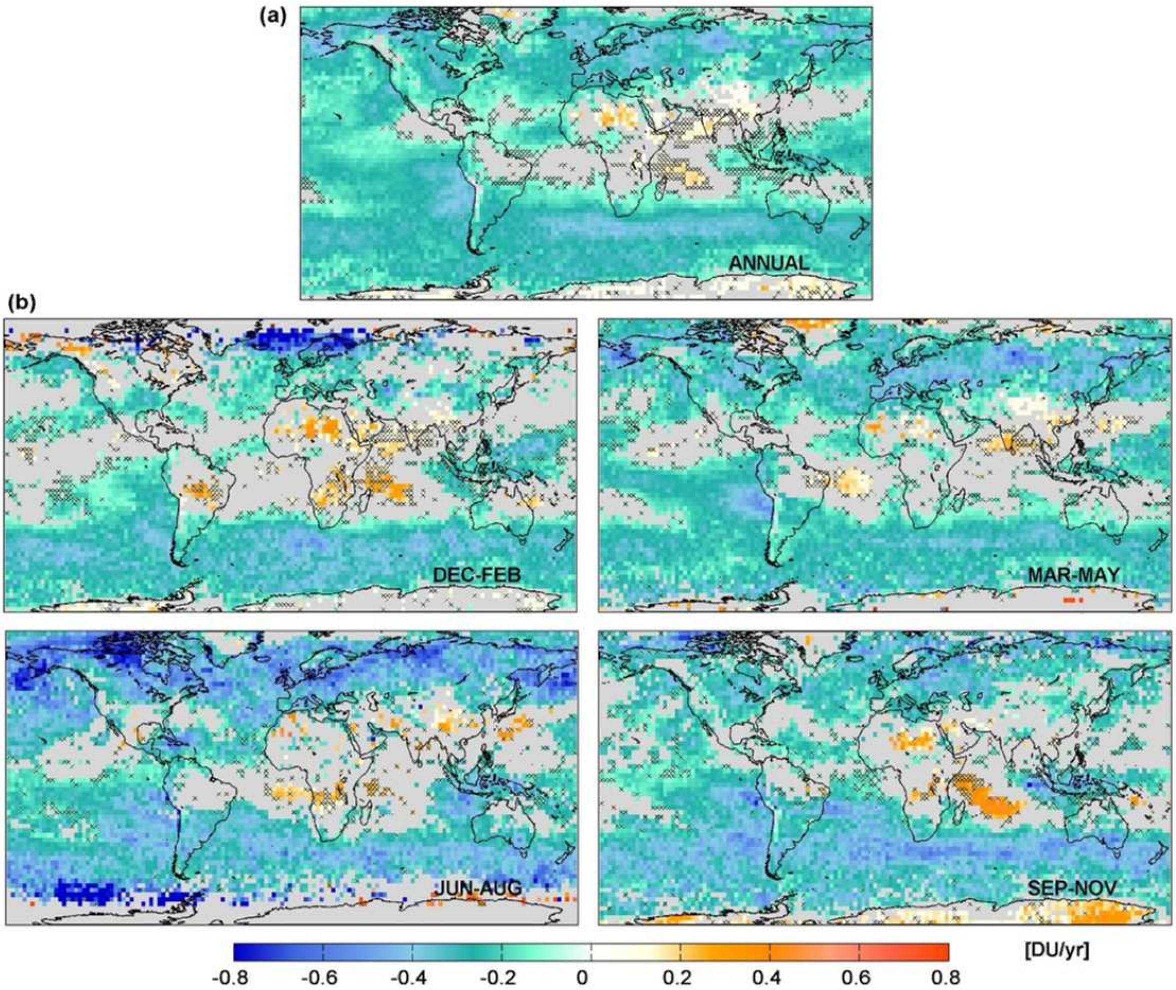


Fig.4. Seasonal distribution of the differences between the *RMSE* of the regression fits with and without linear trend term [$(RMSE_{\_w/o\_LT} - RMSE_{\_with\_LT})/RMSE_{\_with\_LT} \times 100$] (in %). The blue circles in the JJA panel refer to the case presented in Fig.3.


(a)
ANNUAL
(b)
DEC-FEB
MAR-MAY
JUN-AUG
SEP-NOV
-0.8
-0.6
-0.4
-0.2
0
0.2
0.4
0.6
0.8
[DU/yr]

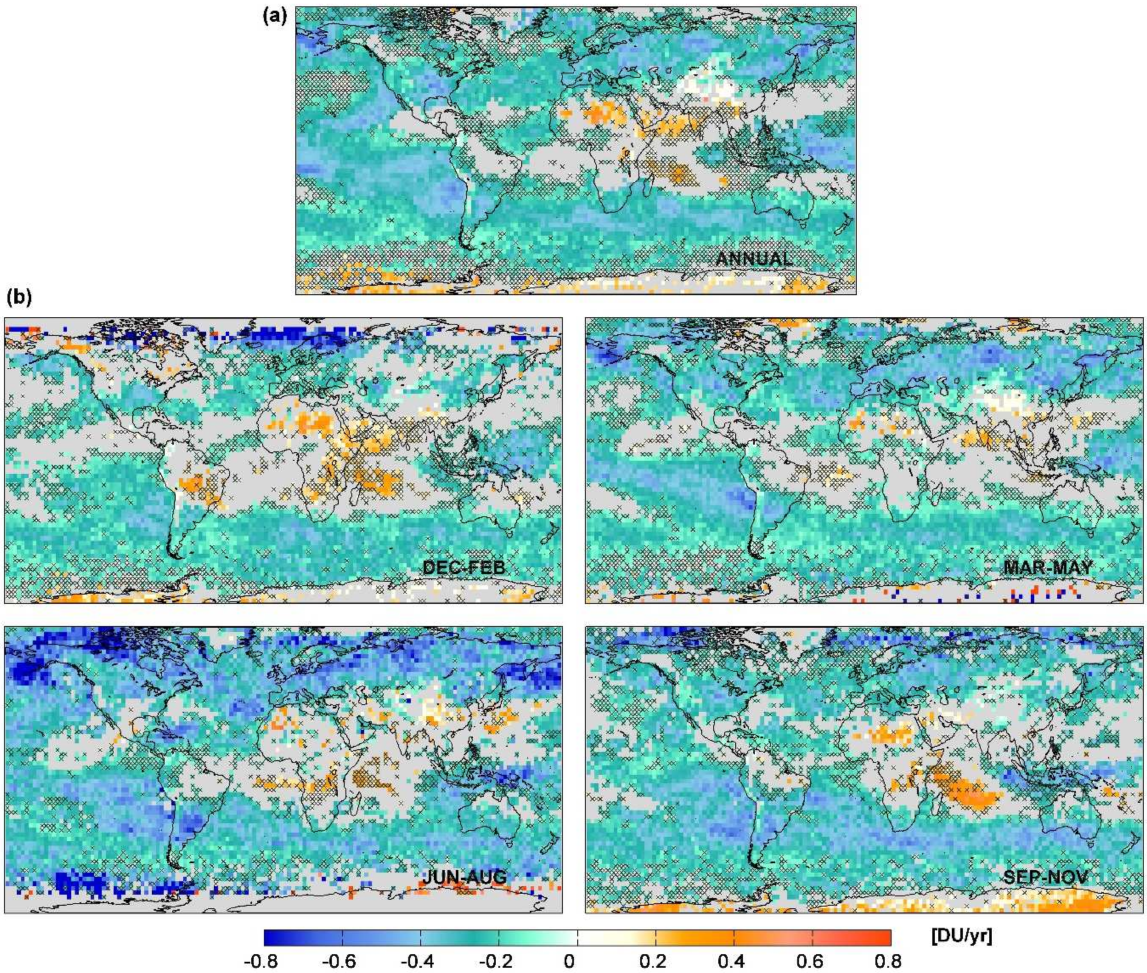
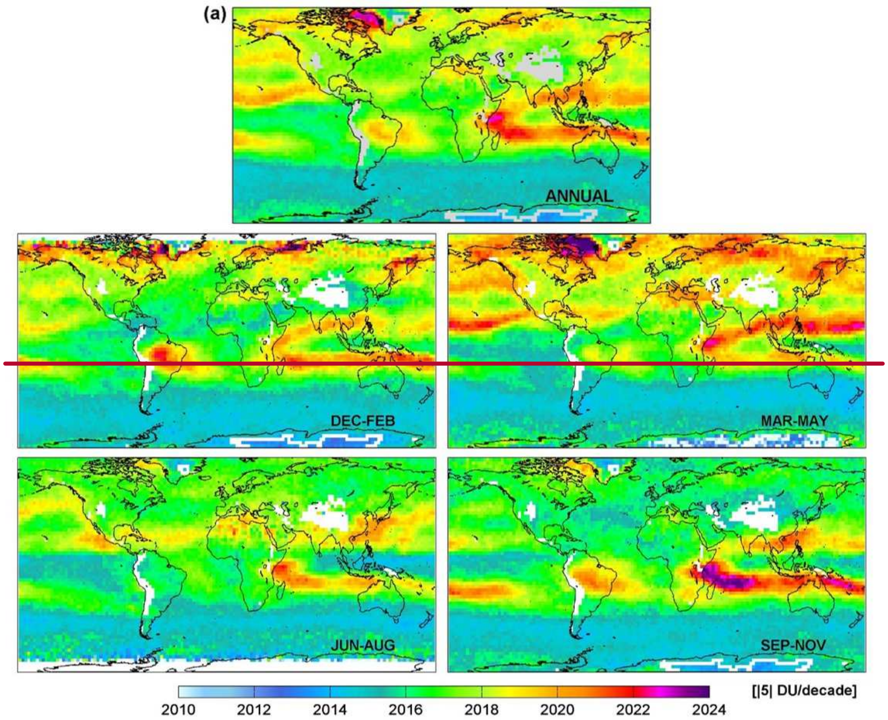
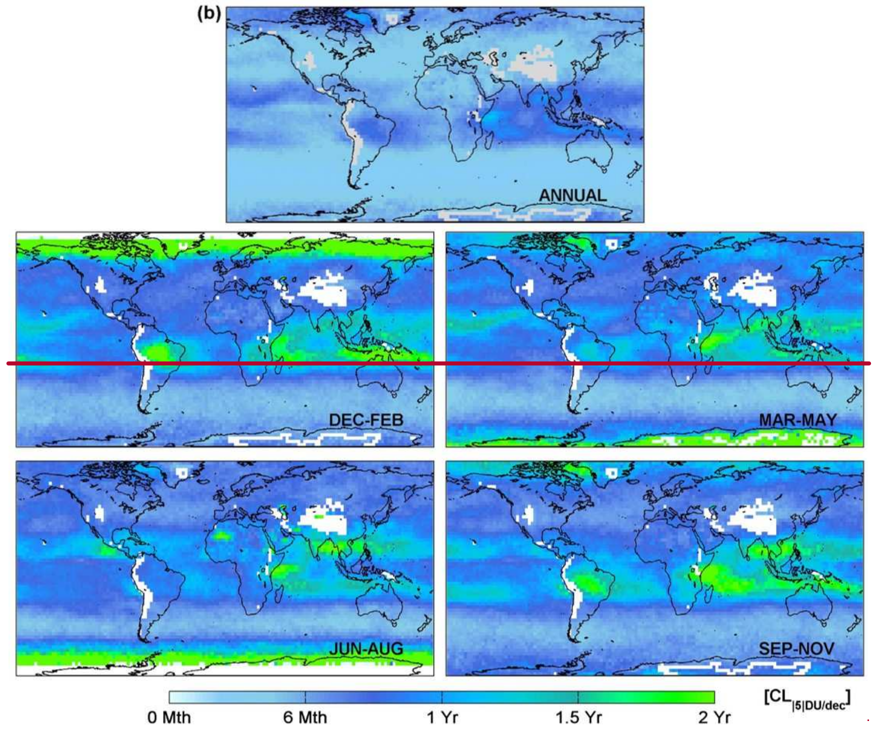
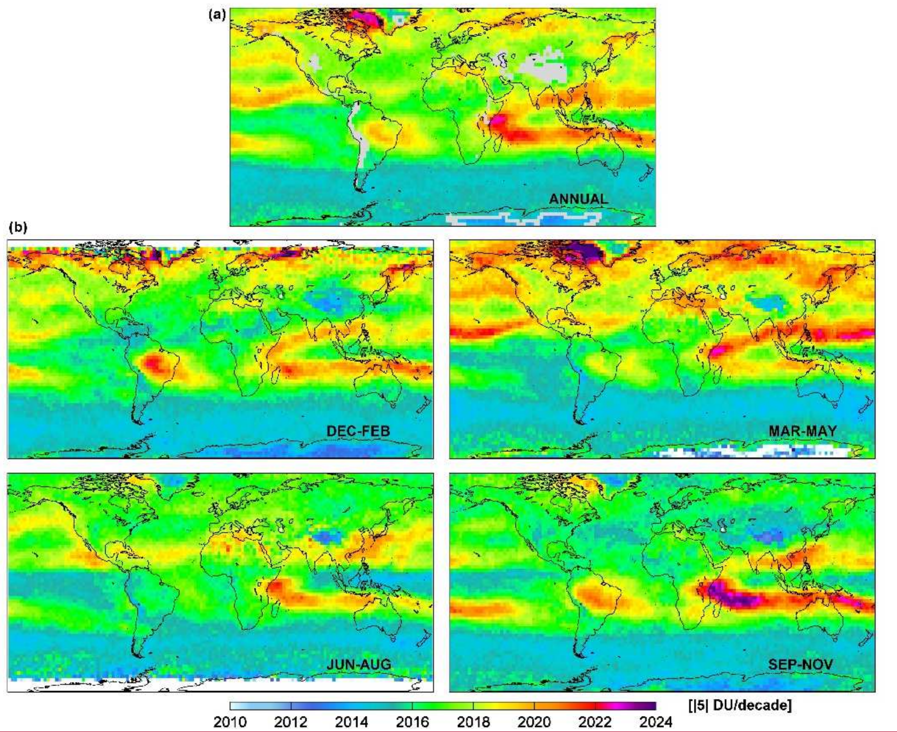


Fig.5. (a) Annual and (b) seasonal distributions of the adjusted trends in DU/yr from the multi-linear regression models. The grey areas and crosses refer to the non-significant grid cells in the 95% confidence limits (2σ level).


(a)
ANNUAL
DEC-FEB
MAR-MAY
JUN-AUG
SEP-NOV
2010
2012
2014
2016
2018
2020
2022
2024
[|5| DU/decade]


(b)
ANNUAL
DEC-FEB
MAR-MAY
JUN-AUG
SEP-NOV
0 Mth
6 Mth
1 Yr
1.5 Yr
2 Yr
[CL|5|DU/dec]



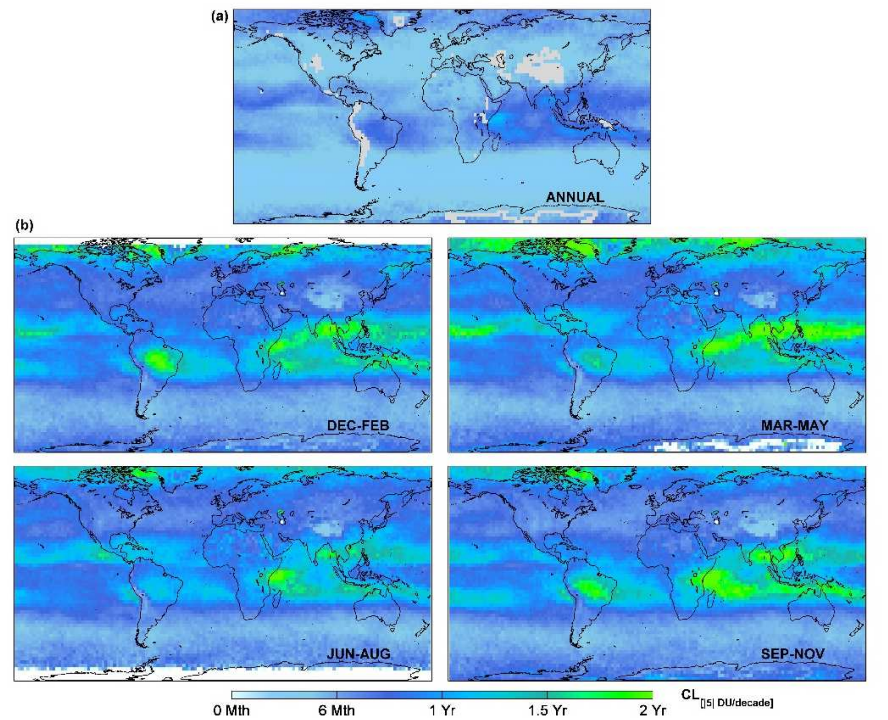
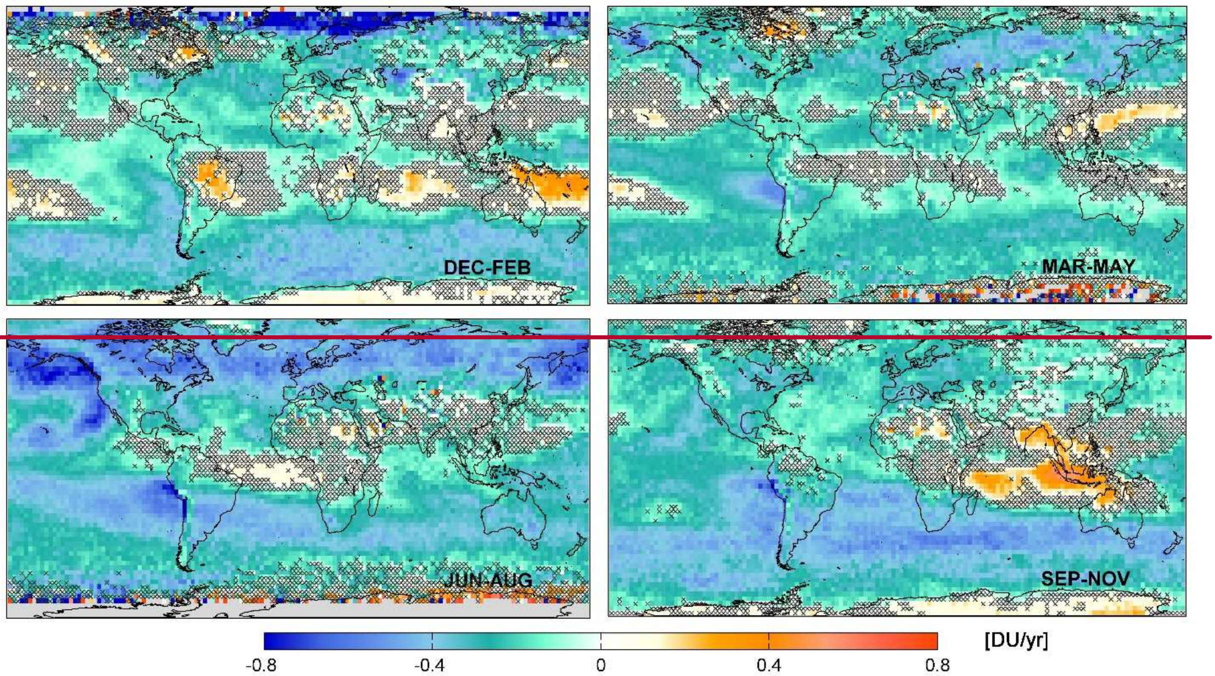


Fig.6 (a) Estimated year of tropospheric IASI O_3 trend detection (with a probability of 90%) for a given trend of |5| DU per decade starting at the beginning of the studied period (20080101) and (b) associated maximal confidence limits from the annual (top panel) and the seasonal (bottom panels) regression models.



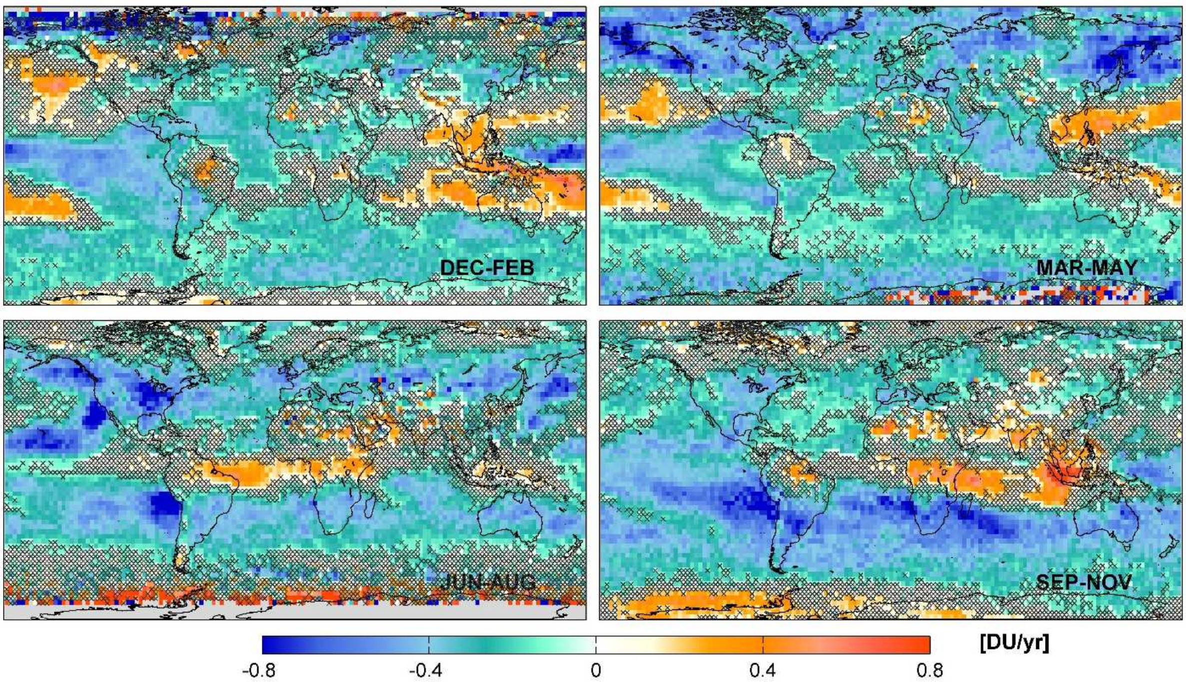
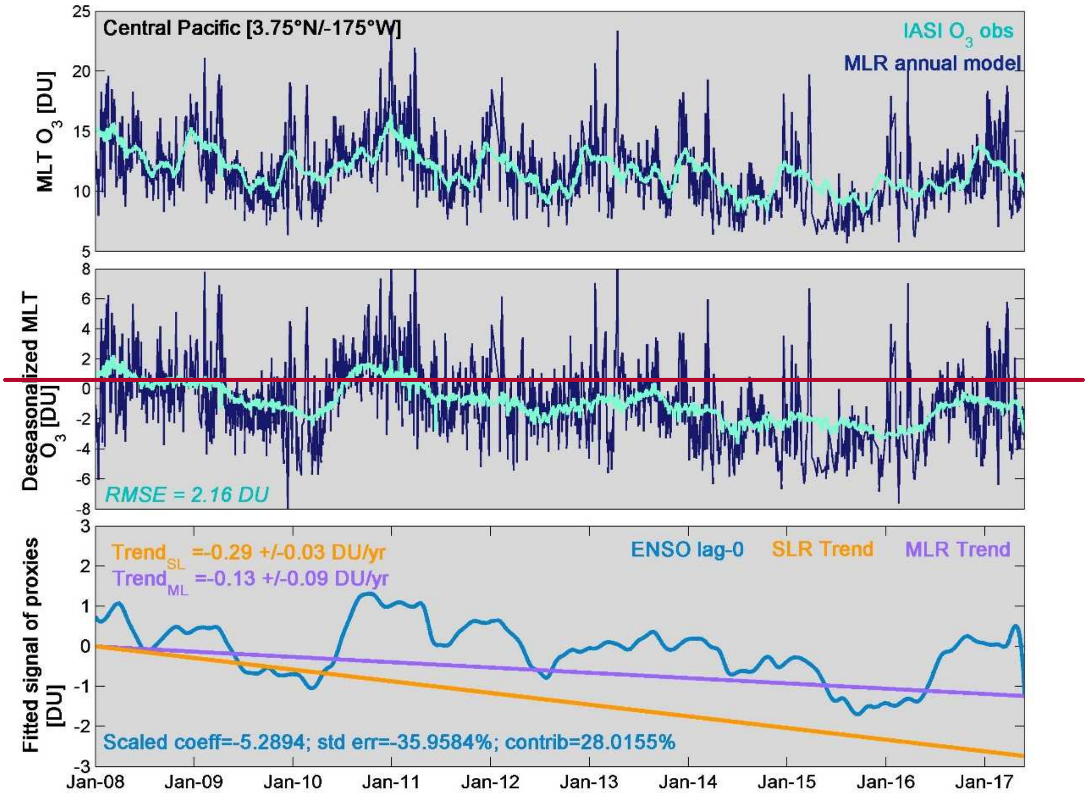


Fig.7. Seasonal distributions of the fitted linear term trends (given in DU/yr) derived from a single linear regression model. The crosses refer to the non-significant grid cells in the 95% confidence limits (2σ level).


Central Pacific [3.75°N/-175°W]
IASI O3 obs
MLR annual model
MLT O3 [DU]
Deseasonalized MLT O3 [DU]
RMSE = 2.16 DU
Trend_SL =-0.29 +/-0.03 DU/yr
Trend_ML =-0.13 +/-0.09 DU/yr
ENSO lag-0
SLR Trend
MLR Trend
Fitted signal of proxies [DU]
Scaled coeff=-5.2894; std err=-35.9584%; contrib=28.0155%
Jan-08
Jan-09
Jan-10
Jan-11
Jan-12
Jan-13
Jan-14
Jan-15
Jan-16
Jan-17

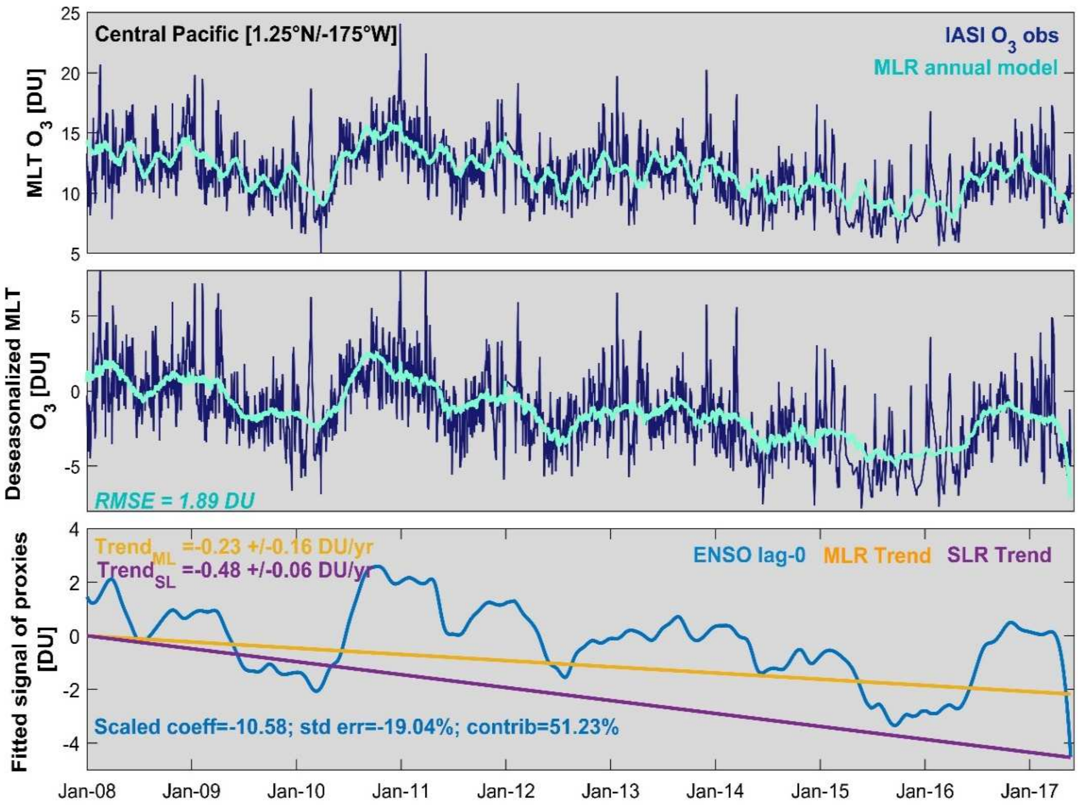


Fig.8. Daily time series of O_3 measured by IASI and adjusted by the multivariate annual regression model (top row and middle row for the deseasonalized O_3), along with the adjusted trends derived from the single and the multivariate linear regressions (SLR and MLR) and of the fitted signal of ENSO proxy (one of the main retained proxies in the multivariate regression model) calculated as $[x_j X_{norm,j}]$ (bottom row) over the equatorial central Pacific (negative ENSO "dynamical" effect) (given in DU). The *RMSE* of the multivariate regression fit and the fitted SLR and MLR trend values are also indicated.

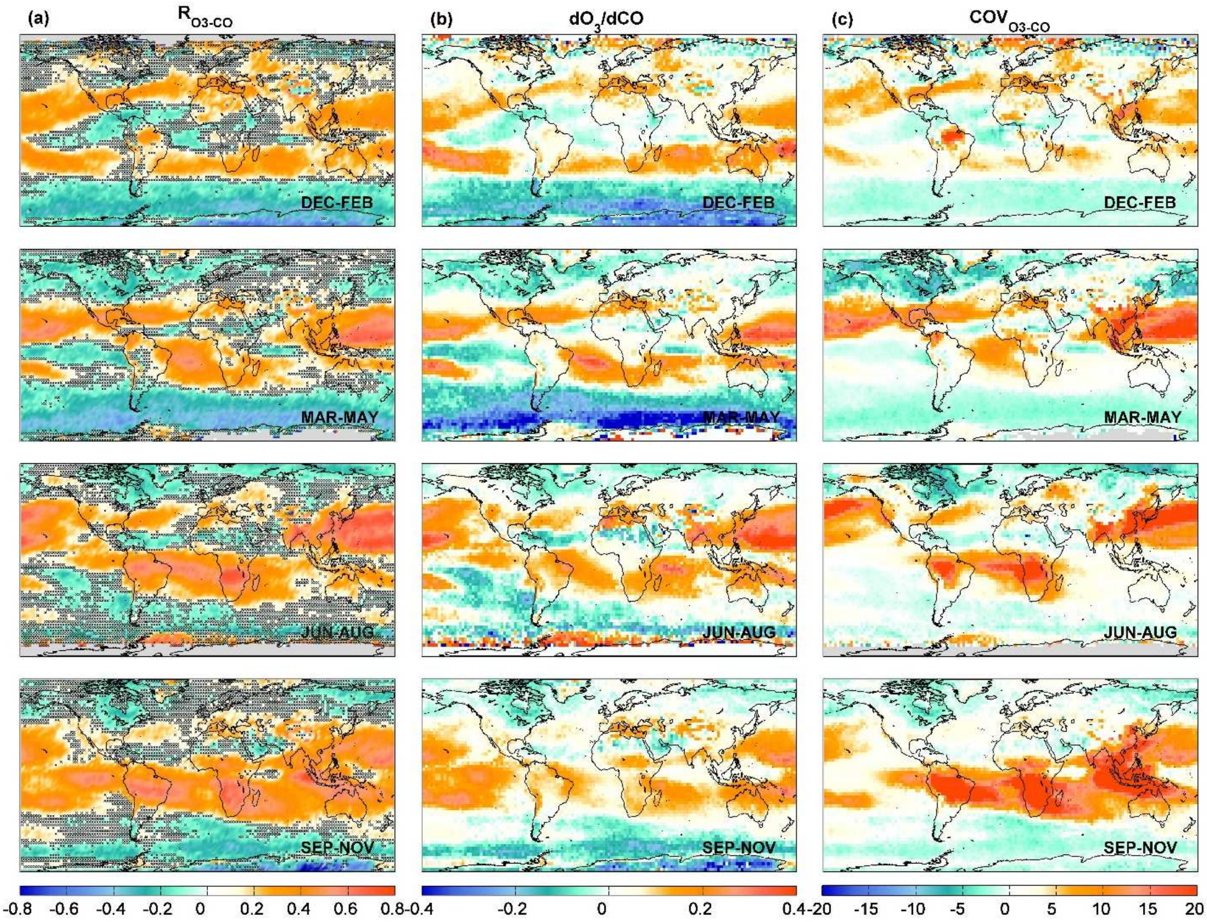


Fig.9. Global distributions of (a) the correlation coefficients ($R_{O3\text{-}CO}$), (b) the regression slope (dO_3/dCO in mol.cm^{-2}/mol.cm^{-2}) and (c) the covariances ($COV_{O3\text{-}CO}$ in 10^{33} mol.cm^{-2}×mol.cm^{-2}) of daily median IASI tropospheric O_3 and CO over January 2008 – May 2017. Data are averaged over a 2.5°x2.5° grid box. Crosses in $R_{O3\text{-}CO}$ panels (a) refer to the non-significant grid cells in the 95% confidence intervals (2σ level).

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