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Ozone variability in the troposphere and the stratosphere from the first six years of IASI observations (2008–2013)

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Introduction

This supporting information gives details on model-measurement comparisons. First, we evaluate the variations in O_3 simulated with MOZART-4 (Emmons et al., 2010a) against IASI by using the regression model described in the manuscript (Section 3). This statistical model is used as a tool for understanding possible biases between MOZART-4 and IASI. Then, the stratospheric influence as seen by IASI in the O_3 tropospheric column (Section 4 of the manuscript) is estimated.

S.1 MOZART-4 simulation set up

The simulations are performed after a 6 months spin-up and driven by offline meteorological fields from the NASA Global Modeling and Assimilation Office (GMAO) Goddard Earth Observing System (GEOS-5) assimilation products (http://gmao.gsfc.nasa.gov/products/). MOZART-4 was run with a horizontal resolution of 2.5°×1.9°, with 56 levels in the vertical and with its standard chemical mechanism. In the stratosphere, MOZART-4 does not have a detailed chemistry and O_3 is constrained to observations from satellite and ozonesondes (Horowitz et al., 2003). The emissions are the same as used in Wespes et al. (2012), with the constant in time anthropogenic emissions from D. Streets' ARCTAS inventory (see http://www.cgrer.uiowa.edu/arctas/emission.html) and the fire emissions from the daily Fire Inventory from NCAR (FINN, Wiedinmyer et al., 2011). Details on chemical mechanisms, parameterizations and emission sources can be found in Emmons et al. (2010a; 2012). MOZART-4 simulations of numerous species (including O_3 and related tracers) have been previously compared to in situ and satellite observations and used to track the intercontinental transport of pollution (e.g., Emmons et al., 2010b; 2013; Pfister et al., 2006; 2008; Wespes et al., 2012).

S.2 O_3 time series from MOZART-4 *vs* IASI

In Fig. S1, the seasonal cycles of ozone from MOZART-4 fitted regression model are compared against IASI fit by taking into account its associated averaging kernels (see Section 2 of the manuscript). In the stratosphere (US and MLS), despite the non-explicit representation of the chemistry and the coarse vertical resolution in this layer, MOZART-4 reproduces the observations in terms of ozone concentrations, amplitude of the seasonal cycle and timing of the maximum. Differences between the fitted cycles associated with the simulations and the observations are lower than 10%, except over the Southern polar region where they reach 30%. In the UTLS region, while the amplitudes of the seasonal cycles and the timing of the maxima are well captured in the model, we observe a systematic bias with an underestimation of O_3 concentrations in the model of around 30% over the high latitudes (north of 50°N and south of 50°S), possibly resulting from a misrepresentation of the STE processes.

In the troposphere, for each 20-degree latitude band, the model shows, contrary to the upper layers, an overestimation of the ozone concentrations (reaching 25% in the equatorial belt) as well as a mismatch in the timing of the maximum which occurs one to two months before the observed spring peak. Especially in the N.H., the shift of the maximum from high to mid-latitudes observed by IASI (see Section 4.1 and Fig. 7 of the manuscript) is not reproduced by MOZART-4 which shows a latitudinal independent maximum in April. This is explained by the constant in time anthropogenic emissions used in MOZART-4. This finding gives further confidence to the ability of IASI to detect anthropogenic production of O_3. The mismatch in the timing of the maximum in the troposphere is characterized by different regression coefficients for the annual term from MOZART-4 and IASI. The annual component (Constant scaled a_1+b_1) decreases from Northern latitudes (from 5% to 10%) to high Southern latitudes (from -30% to 0%) with negative amplitudes south of 10°N and a maximum positive amplitude at 20°N (10%) for MOZART-4, while IASI shows negative values both south of the equator (-20-0%) and north of 30°N (-10-0%) and a similar maximum at 20°N (see Fig.8b of the manuscript). Note that this mis-representation of MOZART-4 in the UTLS and in the troposphere is unlikely due to errors in climatology values used in the stratosphere since the concentrations and the timing of the maximum are well reproduced in that layer.

To better evaluate the sources of the discrepancies between model and measurement, we compare the constant terms from MOZART-4 time series with IASI (Fig.S2, also Fig.8a of the manuscript) using the regression procedure (Section 3 of the manuscript). The comparison indicates that MOZART-4 has a good climatology in the US and MLS (differences < 10%). The biases of MOZART-4 in the UTLS and in the troposphere reported above are highlighted in the fitted constant with, in UTLS, underestimations of ~35% and ~15% over the high Southern and Northern latitudes, respectively, and, in the troposphere, an overestimation of ~25% in the tropics. The latter could possibly point out issues with horizontal transport in the model or overestimated ozone production efficiency at these latitudes.

S.3 Stratospheric influences as seen by IASI

After verifying above the agreement between the O_3 time series from IASI and from MOZART-4, we can investigate to what extent the stratosphere could influence the O_3 variations seen by IASI in the troposphere. To this end, we focus hereafter on variations in the MLT, using a “tagging” method to track all tropospheric odd nitrogen sources (the “tagged” nitrogen species) producing ozone ($O_3^{tagged\_NOx}$) through the tropospheric photochemical reactions (Emmons et al., 2012). This method allows isolating the portion of the stratosphere to the tropospheric O_3. Since the method is fully linear, this contribution is simply calculated as the difference between the total simulated O_3 and the $O_3^{tagged\_NOx}$ (Emmons et al., 2012; Wespes et al., 2012). Fig.S3 (a) presents, for each 20-degree latitude band, the averaged seasonal cycles in the MLT for total O_3 (line) and $O_3^{tagged\_NOx}$ (dashed lines) from fitted MOZART-4 time series accounting for the IASI averaging kernels. The difference between total O_3 and $O_3^{tagged\_NOx}$ represents the stratospheric part as seen by IASI in the troposphere. It is expressed in Fig.S3 (b) as a percentage of the total O_3. The stratospheric contribution ranges between 30 and 65% depending on latitude and season. The largest contributions are calculated for the highest latitudes in winter-spring, and they are attributed to descent of stratospheric air mass into the polar vortex and to less IASI sensitivity. Exception is found over the South polar region which shows a minimum (~25%) due to ozone hole depletion. The smallest stratospheric contributions are calculated in the lower latitudes. As expected from the IASI vertical sensitivity (see Section 2 of the manuscript), the *a priori* contribution is anti-correlated with the stratospheric contribution to some extent and it ranges between ~10% and ~35%. These results suggest that the variability of ozone in the troposphere observed by IASI (Section 4.3.2 of the manuscript) may partly be masked by the *a priori* information and significantly be driven by the variability in the stratosphere.

To further characterize the stratospheric influence, the constant factors associated with the $O_3^{tagged\_NOx}$ fitting time series in the troposphere are superimposed in Fig.S2 (dashed grey line). They represent between 40 and 60% of the constant factors derived from the total O_3 fitting. The north-south gradient for the $O_3^{tagged\_NOx}$ is smaller than for the total O_3, with maximum over the low latitudes of the N.H. instead of over the high latitudes. From Fig.S3 (a), we see in the N.H. that the differences between the variability of total O_3 and that of $O_3^{tagged\_NOx}$ mainly rely on the timing of the maximum with a shift of 2-3 months (maximum in spring for the total O_3 *vs* maximum in summer for the $O_3^{tagged\_NOx}$). That shift is characterized by a positive annual component (Constant scaled *a1+b1*) for the total O_3 (~10%) and a negative one for the $O_3^{tagged\_NOx}$ (~ -20%). In the S.H., we observe a same timing of the maximum between the two runs.

4 Conclusions

Two important results can be derived from MOZART-4 *vs* IASI time series:

1- By comparing the fitted O_3 variations and regression coefficients using the same regression model, systematic biases are found in the troposphere and can be attributed to specific model limitations (no-interannual variability in the anthropogenic emissions, errors in the transport, coarse spatial and vertical resolution of the model and overestimation of ozone production efficiency). In particular, the fact that the MOZART-4 model settings used constant anthropogenic emissions tends to strengthen the ability of IASI to detect anthropogenic production of O_3 and to highlight the need for developing long term continuous anthropogenic emissions inventories (including seasonal and inter-annual variations) for better estimating the impact of anthropogenic pollution changes on tropospheric ozone levels.

2- Our results suggest that the apparent negative trend in the troposphere observed by IASI in the N.H. summer (see Tables 2 and 3 in Section 4.3.2 of the manuscript) is largely attenuated by the influence of the stratosphere (through STE processes and medium vertical sensitivity of IASI in the troposphere). In other words, the decrease of tropospheric O_3, which could be attributed to decline of O_3 precursor emissions, is probably much more important than what we estimate from IASI. This opens perspectives to further comprehensive studies for investigating the influence of stratospheric O_3 recovery on the apparent decrease of O_3 in the troposphere.

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Figure captions

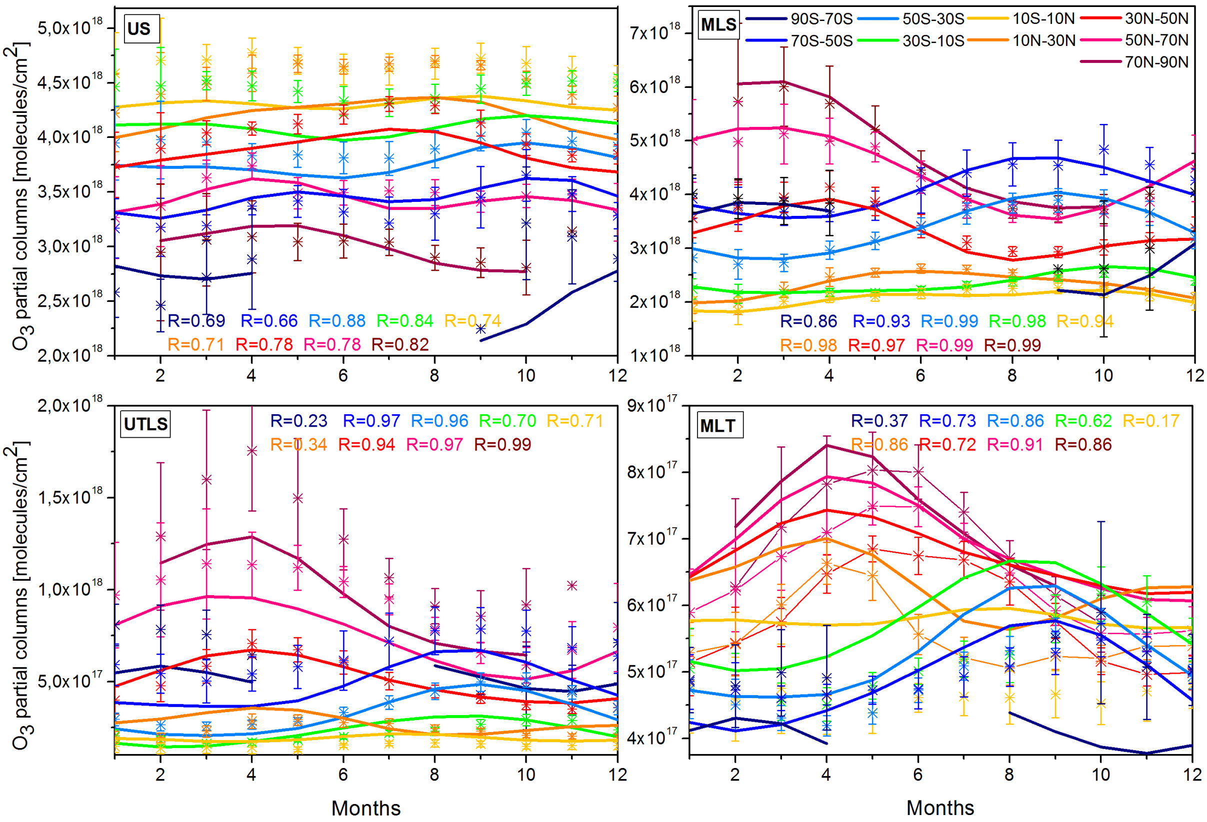

Figure S1: Same as Figure 7 of the manuscript, but for the fit of MOZART-4 simulations (line) smoothed according to the averaging kernels of the IASI observations. The IASI O_3 columns observations (stars) are indicated for the sake of comparison. In the N.H. for the MLT, they are plotted with lines and symbols for clarity. Correlation coefficients (R) between the daily median fitting of IASI and of the smoothed MOZART-4 are also indicated. Note that the scales are different.

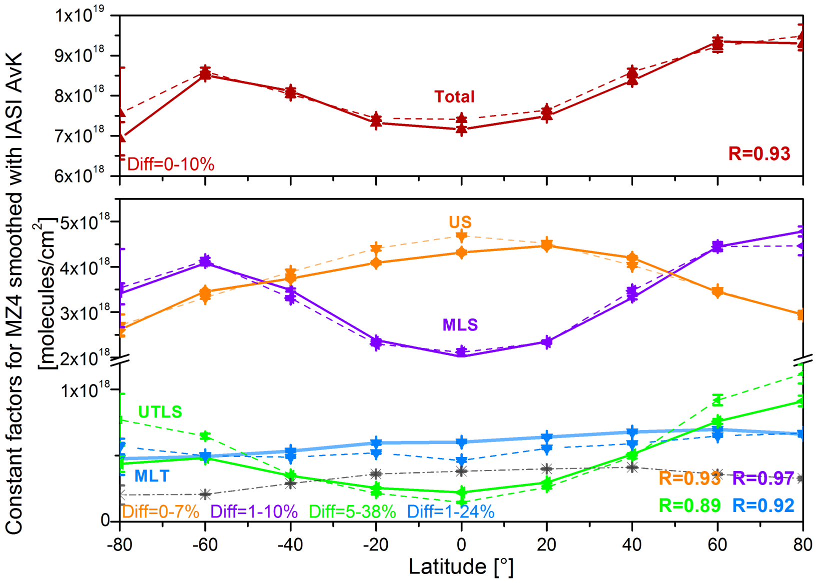

Figure S2. Same as Fig.8 (a) of the manuscript but for the MOZART-4 O_3 time series, smoothed according to the averaging kernels of IASI. Correlation coefficient (R) and relative differences between the Constant factors in the IASI fitting time series (dashed line) and in the MOZART-4 fitting time series (full line) are also indicated. For the troposphere, the Constant factors in the MOZART-4 $O_3^{tagged\_NOx}$ fitting time series are also represented (dashed grey).

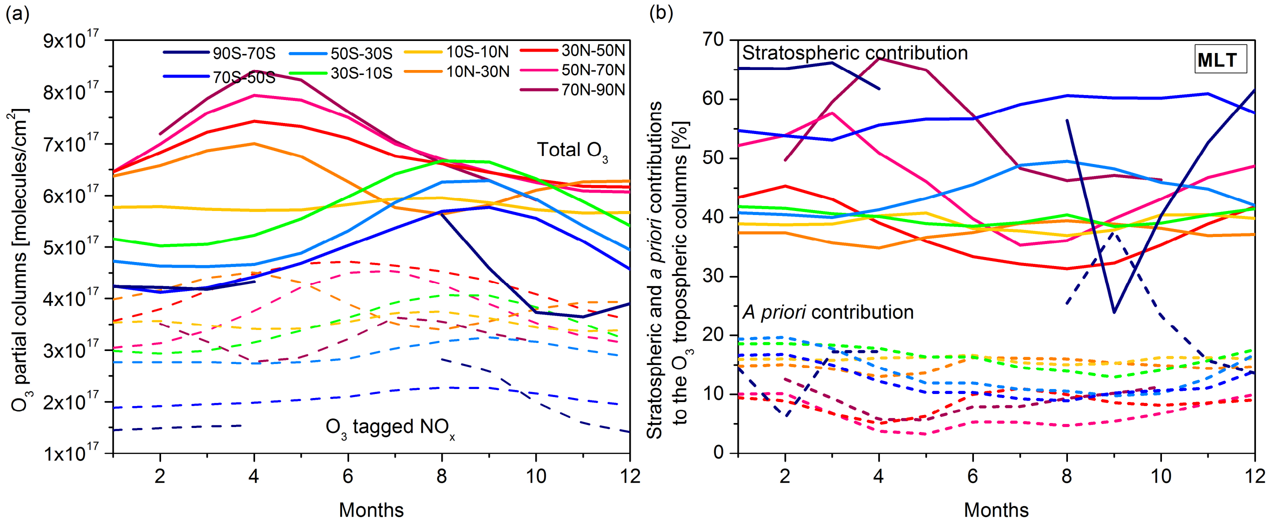

Figure S3: (a) Same as Figure 7 of the manuscript in the MLT layer, but for the fit of MOZART-4 O_3 (full line) and of $O_3^{tagged\_NOx}$ time series (dashed line) accounting for the IASI sensitivity. (b) Stratospheric (full line) and *a priori* contributions (dashed line) to the MLT columns simulated by MOZART-4 accounting for the IASI sensitivity (%).