

# **Tropospheric and total ozone columns over Paris (France) measured using medium-resolution ground-based solar- absorption Fourier-transform infrared spectroscopy**

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## **Responses for report referee # 1:**

First, we wish to thank the Referee for his positive and insightful comments. We have discussed his comments and modified the paper accordingly. See below our answers to his specific comments.

### **Specific comments**

P. 3340, L. 10-15: this part of the introduction is a simple repetition of the content of the abstract. The introduction should focus on explaining the context of the study and on describing the state-of-the-art of the science topic under investigation, rather than summarizing results.

We would propose to delete this part and replace it by (p 3340 line 10-15):

“Assessment of long-term trends in tropospheric ozone is difficult due to the scarcity of representative observing sites with long records (IPCC, 2007). For instance, limited spatial and temporal coverage of ozone sondes, and *in situ* surface measurements, do not account for sufficient tropospheric ozone data set. However, few remote sensing techniques are now able to monitor tropospheric ozone, such as ground based high-resolution FTIR (Pougatchev et al., 1996; Schneider et al., 2005), LIDAR (Beekmann et al, 1995), and FTIR onboard satellite (Keim, 2009 ; Dufour et al., 2010). ”

P. 3342, section 2.2: since this paper reports for the first time on ozone profile retrieval using mid-resolution FTIR, more should be said on the details of the inversion scheme used. In particular, what are the differences with ozone retrievals using high resolution FTIR? How good is the information content compared to high resolution FTIR? Why are retrievals performed on a logarithmic scale? How sensitive are the results to the choice of the *a-priori*? Etc

The inversion scheme used for mid-resolution measurements is quite the same as that for high resolution one, except for the size of the micro-windows. Since the line-widths in mid-resolution spectra are larger than in high-resolution spectra, the micro-windows need to be widened to take into account more lines, in order to achieve the same information content.

The logarithmic scale retrieval is useful for ozone since it assures that the correct *a-priori* knowledge is applied by the inversion algorithm. At the tropopause region the *a-priori* probability density function can be well described by a log-normal pdf (a log-scale retrieval considers this detail). At other altitudes with relatively low ozone variability log-normal and

normal pdfs are equally valid. For more details please refer to Hase et al., 2004 and Schneider et al., 2006.

In addition, retrieved ozone profiles do not depend on the choice of the *a-priori* profile since a change of *a-priori* profile produced a negligible differences on the results.

So we propose to add more details in this section 2.2:

P 3342 from line 10: "This code is widely used by the NDACC community to retrieve trace gases from high-resolution FTIR measurements. However it can be adapted and properly used for mid-resolution retrievals. Note that to achieve sufficient information content in the retrievals, the micro-windows have to be widened compared to high-resolution ozone retrievals."

P 3342 from line 18: "It is worth noting that all retrievals are performed with the same *a-priori* data set to ensure that all variability seen on retrieved profiles comes from the measurements. Furthermore, it was observed that the retrieved ozone profiles do not depend on the choice of the *a-priori* profile since a change of *a-priori* profiles produced negligible differences on the results."

P 3342 from line 21: "The probability distribution function (pdf) of highly variable trace gases is asymmetric. In general the asymmetry is well captured by a log-normal pdf. This is the case for ozone at the tropopause region. Consequently we perform the retrieval on a logarithmic scale in order to assure that the correct a priori information is employed by the inversion algorithm. In case the variability is small a log-scale retrievals is no disadvantage if compared to a linear retrieval since then the log-normal distribution is very similar to the normal distribution. For more detailed discussion please refer to Hase et al. (2004) and Schneider et al. (2006)."

P. 3343, L. 8-25: in this section, the authors show that the retrieved stratospheric and tropospheric columns do not correlate significantly with each other. Although this certainly adds confidence to the results, one should be clear that an absence of correlation, per se, does not validate the tropospheric retrievals. In this sense, it is premature to state that "we have demonstrated that ground-based FTIR measurements are indeed capable of monitoring tropospheric ozone". Such a demonstration can only be achieved through careful comparison with correlative data sets.

We propose to add (p 3343 from line 23): "In addition, the resolving capability as documented by the avks is affirmed by the fact that we see no significant correlation between the ozone values retrieved for the troposphere and the stratosphere respectively"

We propose to change this statement by:

p 3343 line 23-25:"We have demonstrated that ground-based FTIR measurements are indeed capable of monitoring separately tropospheric ozone from stratospheric ozone with little interferences due to the vertical sensitivity of the retrievals; however, comparison of our results with other independent data sets would be highly desirable in the future."

P. 3343, L. 18: in the same paragraph, a regression plot is used to demonstrate the poor correlation between the stratospheric and tropospheric ozone series. If results are uncorrelated, what do represent the slope of the linear regression line with zero intercept? Do

we or do we not expect some level of correlation between tropospheric and stratospheric ozone columns? If yes, what are the physical or chemical mechanisms by which these two quantities can be linked? Note by the way that the slope of the regression shown here should be unit less (not in DU as indicated in the text).

Concerning the slope of the linear regression, it means that stratospheric ozone amount is, on average of 75 daily-measurements, 12.5 times higher than tropospheric ozone amount. But because it has no real impact on the paper's subject, I propose to remove the sentence concerning the slope and delete the value in the caption.

Concerning the level of correlation, we think we can accept it, considering that there are two explanations for this correlation value:

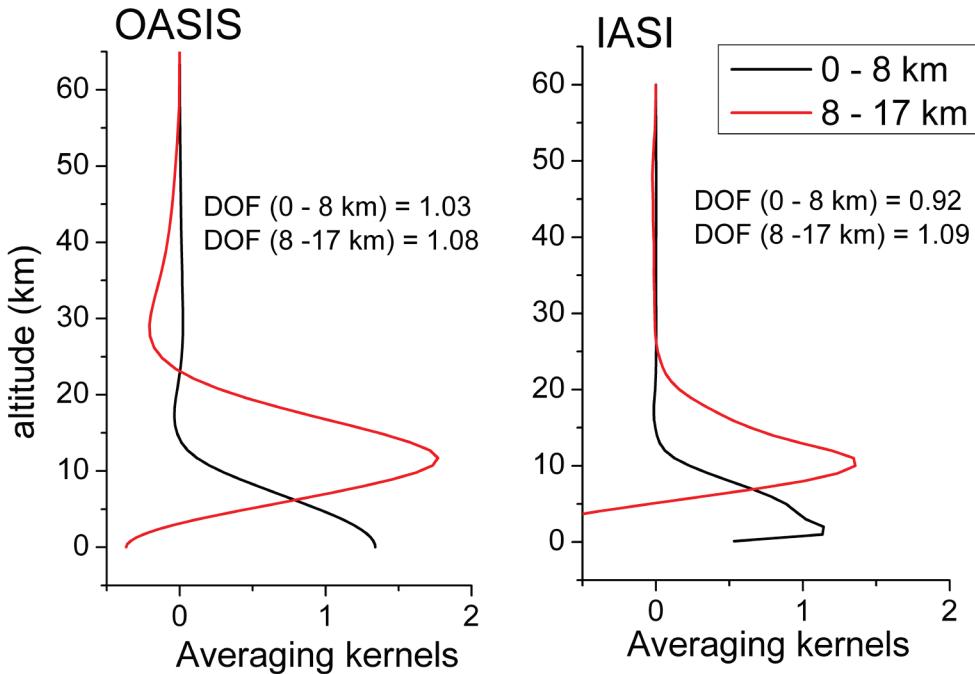
- First, the retrievals' nature implies a certain dependence between all atmospheric layers. That is why the term, "semi-independent" partial columns, is widely used (Eremenko, 2008). The averaging kernel (Fig 2), that reflects the vertical sensibility of the retrievals, clearly shows that the retrieved tropospheric ozone have a small stratospheric contribution.
- Second, atmospheric dynamical processes occurring around the tropopause region can also partly explain this correlation (Holton, 1995). Extreme events, which are common at mid-latitudes (e.g. Reider et al, Atmos. Chem. Phys., 10, 10021–10031, 2010), are caused by meteorological conditions at the synoptic scale, and have been observed during the measurements period presented in the paper.

So we propose to add in the text (p 3343 from line 16): "This correlation can be attributed both to the retrieval and to atmospheric dynamical processes occurring around the tropopause region (Holton, 1995). Indeed, the averaging kernels, which reflect the vertical sensitivity of the retrievals, show that the retrieved tropospheric amounts have a small stratospheric contribution, and vice-versa (Figure 2). Furthermore, one can note that, two episodes of extreme ozone events, due to meteorological conditions, were observed during this measurement period (days 77 and 392), for which stratospheric and troposphere ozone concentrations are correlated".

P. 3345, L. 5: differences are "probably" due to the fact that IASI and OASIS do not have the same vertical sensitivities. This comment is important and definitely deserves more than one line in the discussion. What are these differences in vertical sensitivity? Here averaging kernels should be compared and the implications on the comparison results should be discussed in more details.

To answer this comment, we add IASI and OASIS averaging kernels plots in figure 2 (lower panel) and comment the differences between IASI and OASIS AVK in section 2.2 (p 3343 from line 8): "Figure 2 (lower plot, right) shows typical averaging kernels of IASI retrievals which apply to the tropospheric and stratospheric partial columns where the DOF reach 0.92 and 1.09 respectively. One can note that, compared to OASIS, IASI vertical sensitivity is stronger around 20 km region because it measures thermal infrared radiation emitted by the Earth's surface and the atmosphere in Nadir geometry, whereas OASIS measurements are performed in solar absorption geometry. Furthermore, IASI retrievals are slightly less

sensitive to tropospheric ozone compared to OASIS given values of DOF in the troposphere (0.92 compared to 1.03).”



Concerning the IASI and OASIS comparisons conclusion (section 3.1, p 3345 from line 3), we propose to add: “However it must be acknowledged that the different IASI and OASIS averaging kernels (Figure 2), which reflect differing physics, vertical sensitivities, geometry, and analysis methods, just allow us to draw qualitative conclusions from the comparison, before employing further quantitative analysis, for example with the techniques described in Rodgers and Connor, 2003.”

P. 3345, section 3.2: why not converting partial columns in the lowest layers into surface VMR to allow for more quantitative comparison with in-situ data? Discuss how sensitive to surface ozone OASIS is (cf. AK plot in figure 2). At this stage in the study, why not using ozone sonde data (some are certainly available in the region of Paris) to compare with OASIS tropospheric columns? This could provide a more quantitative insight into the validation process.

In the ozone retrievals,  $DOF = 1.03$  between 0 and 8 km so there is one « semi-independent layer » but there is no sufficient information at the surface level.

In addition, converting partial columns into surface VMR implies to simplify atmospheric meteorological conditions and trace gas profiles from surface to 8 km.

We have already discussed OASIS vertical sensitivity in section 2.2 (p 3343 from line 8). So here we propose to add this sentence: p 3345 line 10 “Indeed, Figure 2 shows that OASIS ozone retrievals are strongly sensitive to variability of the lower atmospheric layers.”

To my knowledge, there were no ozone sonde launching during this comparison period over Paris.

P. 3346, section 3.3: The regression plot in Figure 6 looks pretty good. Are the CHIMERE run performed within MACC constrained by assimilation of in-situ data? In addition to the regression plot, please also include a figure comparing the time-series of tropospheric ozone columns by OASIS and CHIMERE. It would be nice to see how good the variability in tropospheric ozone is captured by both data sets.

There is no data assimilation in the CHIMERE run, but this is effectively the MACC reanalysis set-up (within boundary condition, emissions inventory and meteorological data set).

Furthermore, we add one figure (new Figure 6) of OASIS measurements and CHIMERE simulation tropospheric partial columns (DU) time series for the two pollution events by introducing it in the text as (p 3347 line 13): “In the Figure 6, which presents OASIS and CHIMERE tropospheric ozone time series for the first (upper panel) and the second pollution episode (lower panel), one can see that the variability in tropospheric ozone is well captured by both data sets.”

