

## RESPONSE TO ANONYMOUS REVIEWER #2

We would like to thank the reviewer for his/her comments. We have done our best to address each of the points as detailed below.

Note: All reviewer comments in *italics*; all responses by the authors in normal font.

**1) Even if the findings about the effects of aerosols on OMI AMFs and of the marine biogenic source are valuable and innovative, the main objective of the paper is not attained. Other possible reasons for the enhanced formaldehyde columns over the Mediterranean Sea (like transport from continental sources) should be more discussed (on the base of GEOS-Chem).**

Elevated levels of formaldehyde over the Mediterranean Sea were noticed by many investigators. Curci et al. (2010) suggested that several combined factors, including the life of marine organisms (and their related emissions) and large amounts of Saharan dust (potential nutrients for plants) may play a role for such an enhancement. Therefore, the two main objectives of the current paper were: i) to show the consistency of spatiotemporal variation of enhanced HCHO vertical columns over the Mediterranean Sea, and ii) to explore two potential causes of the problem - satellite retrieval error and/or ocean biological sources of VOCs. As both of these objectives are examined in details, we believe that the main goal of the paper is achieved. Moreover, results of our study help reducing the number of possible missing HCHO source in marine boundary layer (Ayers et al., 1997; Weller et al., 2000; Elrod et al., 2001). We agree with the reviewer that the other sources (such as the transported primary and/or the secondary formed HCHO, formaldehyde emitted from biomass burning, as well as intense ship traffic) are likely contributors to increased HCHO levels over the Mediterranean and therefore deserve further investigation; however it is outside the scope of the current study.

**2) The comparison between OMI and SCIAMACHY observations is not convincing. The use of SCIAMACHY data do not bring anything more to the discussion and we advice either to improve this section (taking into account differences between OMI and SCIAMACHY products such as the treatment of aerosols in the AMF calculations or differences in the sampling frequency of the two sensors), either to skip this part of the paper.**

Comparison of the OMI and SCIAMACHY satellite sensors has been removed in the updated manuscript.

### *Specific comments*

**3) Intro, p17915, line 26: Reference to Stavrakou et al., 2009b would be more appropriate than 2009a.**

This correction has been included in the updated manuscript.

**4) Intro, p17917, line 1:** Remove the word “polluted”. Continental air masses can transport VOC from pollution, from biogenic sources or from fires, especially in summer.

This correction has been included in the updated manuscript.

**5) Section 2.1:** The references of the two satellite products used in the study should be mentioned earlier in section, before detailing the retrievals (after line 21, p17917).

The reference of the OMI product is mentioned earlier in the updated manuscript.

**6) Section 2.1, p17917, line 24:** Both retrieval algorithms (OMI and SCIAMACHY) are based on a non-linear least-squares fitting of the recorded spectra (the DOAS technique). Please give a reference for the OMI HCHO retrieval procedure.

Reference for the OMI HCHO retrieval procedure is included in the updated manuscript.

**7) Section 2.1, p17918, line 16:** There is a mistake here. SCIAMACHY retrieval is using the IMAGES chemistry transport model and the LIDORT radiative transfer code for the AMF calculation (and no correction for aerosols is applied) [De Smedt et al., 2008].

Comparison of the SCIAMACHY data with that of OMI has been removed in the updated manuscript.

**8) Section 2.1, p17918, line 29:** Remove the words “daily swath data: “while for SCIAMACHY, the TEMIS level 2 HCHO product (version 1.2) is used (<http://www.temis.nl/airpollution/ch2o.html>). ”

Comparison of the SCIAMACHY data with that of OMI has been removed in the updated manuscript.

**9) Section 2.1, p17918, line 18:** The description of the GEOS-Chem version used in the present study should come after the description of the satellite products, at the end of section 2.1.

The description of GEOS-Chem has been moved to the end of section 2.1 in the updated manuscript.

**10) Table 1 of the supplement:** Why the formaldehyde yields are they taken from Dufour et al. (2009), while the authors used the GEOS-Chem model? It would be more logical to provide the yields calculated with the GEOS-Chem chemistry.

Table 1 of the supplement has been removed in the updated manuscript.

**11) Section 2.1, p17919, line 4:** I don't understand how the detection limits are evaluated. For which spatial and temporal resolution are they given? a single observation, an averaged column in a grid cell, other? Do they correspond to the random errors? The given numbers seem small for a single slant column. Please clarify and give an estimation of the errors corresponding to

*the satellite gridded data at 0.25° resolution and averaged over 8-days or one month, plus an approximation of the number of OMI and SCIAMACHY observations in a grid cell.*

The “detection limit” used in the text actually referred to the estimated average error on a single observation. We acknowledge the term is misleading and in the updated manuscript changed it by the “uncertainty”. The uncertainty associated to a single observation contains the random error due to fitting uncertainty and to other sources (e.g., AMF). Assuming a negligible systematic bias in OMI observations, the uncertainty may be reduced with the square root of the number of observations. In this study OMI daily swath data was averaged onto a  $0.25^\circ \times 0.25^\circ$  grid and the contribution of each pixel to a grid cell was weighted according to the intersecting area. Each grid cell was therefore assigned the average column and the average column uncertainty of the pixels falling in it each day. The number of OMI pixels that contributes to a cell is 2-4 per day. The uncertainty of the gridded daily HCHO columns may then be reduced applying temporal average. The reduction scales with  $1/\sqrt{N}$ , where  $N$  is the number of days with valid data over a given period. The uncertainty may thus be reduced by  $\sim 3$  times with 8-days average and by  $\sim 5$  times with 1-month average. This issue has been clarified in the updated manuscript.

**12) Section 3.1, p17922, last paragraph:** *- Can you conclude that the OMI HCHO columns are currently too elevated over the Mediterranean Sea? - When the new AMFs are used, the east-west difference in the formaldehyde distribution over the Mediterranean Sea disappears (Figure 3). Please elaborate on this.*

Discussion regarding the east-west difference in the formaldehyde distribution over the Mediterranean Sea has been included in the updated manuscript.

**13) Section 3.2:** *Please precise which OMI HCHO columns have been used for the estimation of isoprene emissions (the original level 2 columns or the recalculated columns with reduced Saharan dust sources). Does the reduction of the vertical columns using the newly calculated AMFs bring the isoprene emissions to more realistic values?*

The use of the OMI HCHO vertical columns derived from recalculated AMFs for the isoprene emission estimate is described in the updated manuscript. The updated manuscript now states that the isoprene emission flux is derived from corrected OMI HCHO vertical column densities.

**14) Section 3.3:** *We suggest showing a plot with the simulated GEOS-Chem HCHO columns.*

The monthly means of GEOS-Chem HCHO columns are now added to the online supplement.

**15) Section 3.4:** *The authors should state clearly if the enhanced summertime concentrations of HCHO over the Mediterranean Sea is also observed in the SCIAMACHY data (possibly by showing maps similar to Figure 1, together with the GEOS-CHEM columns).*

In the updated manuscript, the use of SCIAMACHY data to compare with that of OMI has been removed.

**16)** *Section 3.4, Figure 5: - Considering the sampling time of SCIAMACHY (global coverage in 6 days, at best), and considering the noise inherent to formaldehyde retrieval, 8-days averaged columns are not a fair representation of the SCIAMACHY formaldehyde observations. Monthly averaged columns are much more suitable in order to catch the seasonal variations. It is also recommended not to use the SCIAMACHY results for solar zenith angles larger than 60°. This could explain the abnormally high value in winter time over the Mediterranean Sea. Please, modify the figure accordingly.*

In the updated manuscript, the use of SCIAMACHY data to compare with that of OMI has been removed.

**17)** *Furthermore, the SCIAMACHY AMFs are not corrected for aerosols. An additional test with OMI AMFs calculated for aerosol optical thicknesses switched to zero would be more appropriate for the comparison with SCIAMACHY.*

In the updated manuscript, the use of SCIAMACHY data to compare with that of OMI has been removed.

**18)** *Figure 1S: SCIAMACHY HCHO slant columns are normalized with the reference sector correction above the Pacific Ocean. As can be seen on the figure, the SCIAMACHY normalized slant columns are around zero in regions A, B and H. The final HCHO vertical columns in the Pacific Ocean are taken from the IMAGES model. It is therefore meaningless to show comparison of slant columns and this figure has to be removed. It would be more appropriate to show comparison of AMFs (with and without aerosols for OMI).*

In the updated manuscript, the use of SCIAMACHY data to compare with that of OMI has been removed.

**19)** *Technical corrections p17917, line 24: short lived VOCs with high HCHO yields.*

These correction have been included in the updated manuscript.

**20)** *p17926, line 12: SCIMACHY -> SCIAMACHY*

In the updated manuscript, the use of SCIAMACHY data to compare with that of OMI has been removed.