Reply to anonymous Referee#1

We would like to thank the reviewer for his/her positive evaluation of the manuscript and for the useful comments and suggestions. Below we address the raised concerns. The reviewer's comments are *italicized*.

In my opinion, this is a great paper, and surely a benchmark in the field. Although it is long! It presents a thorough discussion of the inferred top-down fire and isoprene emissions, and compares them both to independent emissions inventories and flux measurements (for isoprene).

Figures are excellent (though units should really be attached to color bars in figs 1, 2, 5, 6, 16)

Units are added in Fig. 1, 2, 5, 6, 16, as well as in supplementary figures S2, S3 and S4.

If I had one issue, and this is maybe really for a future paper, it would be to compare the simulated IMAGES tracers based on the a-priori and optimized emissions against observations. That it is actually compare the simulated concentrations of isoprene, HCHO + other key VOCs and tracers against in-situ ground and aircraft observations to really see how the model improves. Comparing with other emission estimates is good, but you really want to see if the model does better in simulating atmospheric chemistry. There are plenty of observational datasets during the studied time period to do this.

As the paper is already long, we limited the validation to comparison with available isoprene flux measurements over the Amazon (Section 4). The modelled formaldehyde concentrations based on a priori and optimized emissions were evaluated against aircraft observations over North America in our previous inverse modelling study based on OMI (and GOME-2) HCHO columns (Stavrakou et al., 2015). Nevertheless, we agree with the referee that in a future study the satellite-based emissions and model concentrations for other compounds should be validated against available observations.

Minor comments

Abstract: first sentence does sound right when you read it. Maybe: "As HCHO is a high yield…"

Corrected.

OMI row anomaly - did you check how the number of observations changes per grid cell, and how that correlates with inferred emission trends.

As explained in our answer to the next comment, we omitted from the calculation of the monthly OMI column averages the rows that disappear in 2013 throughout the entire OMI time series (2005-2013), in order to minimize the effect of the OMI row anomaly on the estimated trends. Nevertheless, it is a good suggestion by the referee to check whether the changes in the number of observations correlate with the inferred emission trends. The following figure shows that the

annually averaged HCHO columns (VCD, in red) are indeed positively correlated with the number of observations (NBP, in blue). Furthermore, despite our removal of anomalous OMI rows, the number of observations shows a decreasing trend in many regions, likely reflecting a slow deterioration of the OMI retrievals unrelated to the row anomaly. As already pointed out in the manuscript, it cannot be ruled out that time-dependent instrumental effects might impact the inferred emission trends.

The strong correlation between the number of observations and the OMI columns displayed in the figure would be very worrisome if it could not be easily explained by the important role of clouds in both the HCHO retrievals and the formation of HCHO in the atmosphere. Because of our cloud filter excluding scenes with more than 40% cloud fraction, cloudy conditions are associated to fewer observations, as shown by the good correlation of NBP with the PAR (Photosynthetically Active Radiation) irradiance at the surface, obtained from the ECMWF ERA-Interim analysis. The HCHO columns are also usually depressed under cloudy conditions, primarily due to the influence of light on biogenic VOC emissions. PAR is also correlated with temperature (which also drives BVOC emissions) and with the abundance of the OH radicals which is the primary VOC oxidizing agent. As a result, PAR is found to be well-correlated with the HCHO columns over most regions, e.g. over the Southeastern U.S. (0.73) and over Amazonia (0.83). Although there appears to be a clear declining PAR trend over Western Europe between 2005 and 2013, there is no discernible PAR trend over Amazonia and the S-E U.S. A trend in cloudiness is therefore not responsible for the negative emission trends over these regions.
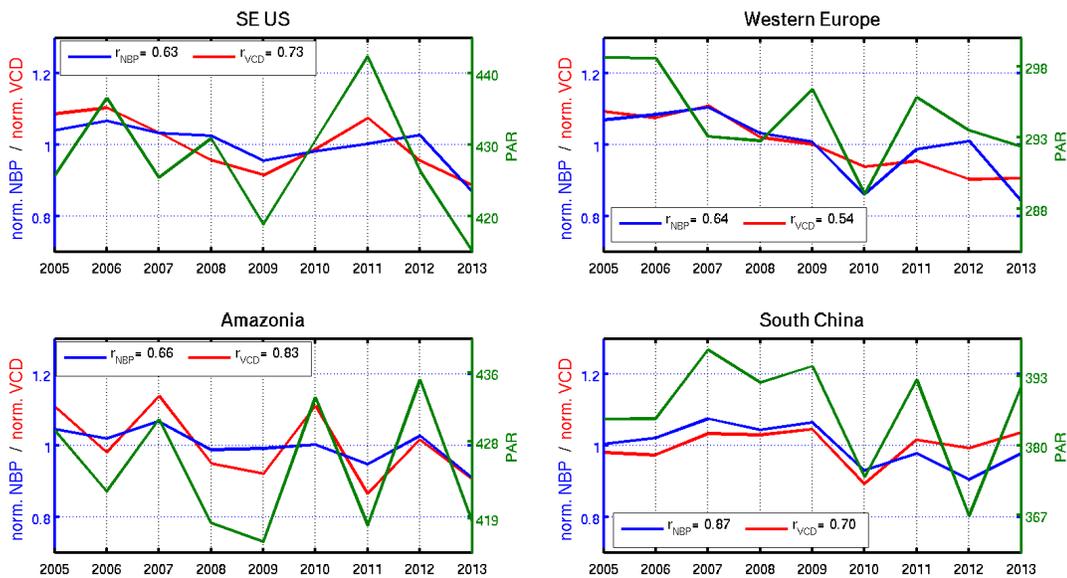

Figure. Normalized mean annual number of observations (NBP, blue) and OMI HCHO columns (VCD, red) over Southeastern US (26-36 N, 75-100 W), Western Europe (37-71 N, 10W-20 E), Amazonia (5-20 S, 40-75 W) and South China (18-32 N, 109-122 E) between 2005 and 2013. Annual mean photosynthetically active radiation (PAR) in the same regions is shown in green. The correlations between PAR and NBP (r_{NBP}) and between PAR and VCD (r_{VCD}) are shown inset.

Did you also try only the OMI rows which are unaffected throughout the mission?

As mentioned in the manuscript, the filtering of the row anomaly leads to a loss of coverage throughout the years. To ensure meaningful determination of trends, we omitted from the calculation of the monthly OMI HCHO column averages the rows that disappear in 2013 (rows 28-46, 54 and 55) throughout the entire OMI time series (2005-2013). The choice results in a reduction in spatial coverage of the OMI data since the beginning of the mission, but ensures a constant spatial coverage in 2005-2013 and pixels of the same size. The effect of this filtering is a slight decrease of the column means in the beginning of the time series.

page 5, line 24: "Inversions are performed separately for each year" - in the framework of a continuous adjoint simulation? i.e was it a start-stop inversion?

We performed 9 independent inversions for each year of the study period, each constrained by satellite observations of the corresponding year. The inversions start in January after a 4-month spin-up. In each inversion, the model is confronted with 12 months of OMI observations, after a 6 month spin-up.

page 6, line 23: there is a '(s)' -is this a typo?

The (s) denotes the GFED4s inventory which includes small fires. To avoid confusion we removed it.