Reply to Anonymous Referee #2

We thank the Referee for his/her careful reading of the manuscript and for his/her constructive and detailed comments. We have followed his/her suggestions to improve the content as recommended. A detailed point-by-point reply (in bold) is provided hereafter.

> The manuscript title "The formaldehyde budget as seen by a global-scale multiconstraint and multi-species inversion system" aims to bring a lot of data and computational effort to constraining formaldehyde. The inversion nicely involves multiple species and data sets, which is an improvement on previous studies. I really appreciated the thorough set up of the inversion and the use of extensive independent data.
> It's a little disappointing though that with all the data and computational machinery, it doesn't seem like we are learning much. Is that right? It would be great to see more analysis and discussion of the results.

This study already brings a lot of data together to constrain the global and regional budgets of the 3D production of HCHO (that rely on NMVOC emission), of the CO emissions and of the CH_4 emissions. To further develop the analysis, we have added:

- **a paragraph on the possible causes of discrepancies between the OMI concentrations and our prior model concentrations (Section 5.1).**
- **a paragraph on the seasonality of the posterior 3D HCHO production by NMVOC compared to the prior one (Section 5.2.1).**
- **An analysis of the year 2010 by extending the inversion period to 2005-2010. This year is especially interesting because of the Amazon drought, co-occuring with peaks of fire activity in South America, that are now analyzed in the paper (Section 5.4.2).**
- **a paragraph on the CO emissions interannual variability (Section 5.4.2).**

> Other than that, I have a several specific comments that I list below.
> p 6911, l14. that seems like an outdated reference

We think that it is worth comparing newer studies to the established reference of IPCC (not updated since 2001). We have kept it.

> p6911, l23-24: the reference list is representative of previous work, but not exhaustive. I'd suggest adding "eg" in front of it

This has been done.

> p 6915, l3: "may be biased"? or just not validated?

According to Crawford et al. [2004], MOPITT's thermal band radiances are more sensitive to surface emissivity at night than during the day, and consequently less sensitive to the CO distribution at night. Therefore the CO retrieval errors are larger at night than during the day. We have added this information in the text.

> section 4: It was very disappointing to see that errors were set arbitrarily. Is this really the best we can do at this point? While many other studies devote a lot of time thinking about this issue, it seems like a step back to set arbitrary errors. I found that unacceptable. At the least there should be some discussion as to why the authors had to resort to setting errors to arbitrary numbers.

The errors were not set at random, still we honestly acknowledge some degree of arbitrariness. We have rephrased this part more precisely: "This choice of a relatively large value accounts for uncertainties in the seasonal cycle of particular emissions, such as fires [Chevallier et al., 2009]. Given the large discrepancies associated with the biogenic NMVOC estimates (e.g, between IPCC [2001] and Guenther et al. [2006]), errors assigned to the scaling factors of the 3D-chemical production of HCHO are set at 400%. For MCF emissions, the

EDGAR-v3.2 inventory by Olivier et al. [2001] has been adapted to give estimates of MCF emissions over our time period (2005-2010) by applying an exponential decrease (update of Bousquet et al., [2005]). As MCF emissions are well known, errors are set to 1% of the flux for MCF. The errors assigned to the scaling factor of OH are set to 10%, based on the differences seen between various estimates of OH concentrations [Prinn et al., 2001; Krol and Lelieveld, 2003; Bousquet et al., 2005]. Finally, errors are set to 100% of the flux for CH_4. Several sensitivity tests associated with these prior settings are presented in Section 6."

section 5.1.1., p6920 What does that 1 month lag mean in the results? why are the emissions wrong? It'd be great to read more insight

As now written in Section 4, the norm of the gradient of the cost function has been reduced by 98% (against 95% previously) and there is no 1-month lag anymore between the prior and the posterior concentrations.
However, we agree with your remark: we have added information to explain the difference between OMI observed concentrations and the prior concentrations. Section 5.1 is now:
"The HCHO prior columns simulated by the model weakly agree with OMI observations, both in magnitude and in their seasonal cycle. Except over the boreal and European regions, OMI measurements are smaller than the prior columns, all over the year. The largest discrepancies are found over the tropical regions, and particularly over South America and Indonesia. This has already been pointed out by Barkley et al. [2008], who found higher model HCHO columns (using the GEOS-Chem chemistry-transport model and the MEGAN inventory for NMVOC emissions) than GOME HCHO measurements. HCHO concentrations are mainly driven by NMVOC emissions and the uncertainties associated with these NMVOC emissions (e.g., in the GEIA inventory, or in Model of Emissions of Gases and Aerosols MEGAN) are large [Barkley et al.,2008]. These uncertainties result from errors in emission factors, and from incorrect or incomplete parameterizations of activity factors. For example, tropical rainforest emission in the GEIA inventory are based on ambient isoprene concentration measurements from a single study [Zimmerman et al., 1988; Barkley et al., 2008], which could explain the notable differences in term of magnitude over tropical regions.
In tropical regions, except Indonesia, our prior model does not reproduce the observed seasonal cycle, particularly over South America and Africa. It should be noted that the entire growing season of isoprene emission is represented by a single basal emission factor in GEIA inventory. Kuhn et al. [2004] found it inadequate for certain representative tropical plant species. This could explain the differences in terms of seasonality over tropical regions.
After optimization of the 3D HCHO production, the model succeeds in capturing both the seasonal cycle and the magnitude of the concentrations. Indeed, Fig. 4 shows a better fit than between the posterior simulated columns and the observations compared to the prior ones, over regions USA, South Asia, South East Asia, Australia, South American Temperate, South American Tropical and Indonesia. However, some discrepancies remain: for example, the model fails to reproduce the observed seasonal decrease from July to October over North Africa. This could be explained by the relatively large OMI data uncertainties over this region (particularly over Sahara), reaching more than 250%, which implies less deviation from prior fluxes as compared to regions with less uncertain data. The agreement for boreal regions is not as good as for other regions because OMI data north of 65°N are not used in the inversion."

section 5.1.2, last paragraph: Again, what does the "significant modification" imply? Do we learn something about HCHO production or precursor emissions?

We have added some text on the posterior HCHO production by NMVOC (linked to precursor emissions) over Northern Africa in Section 5.2.1: "The African continent has a posterior 3D HCHO production by NMVOC of 54 TgHCHO (34 TgHCHO for Northern Africa and 20 TgHCHO for Southern Africa), 36% lower than the prior one. It is in

agreement with the study of Marais et al. [2012], who infer isoprene emissions from HCHO OMI satellite data and applied it to the African continent: they found total OMI-derived isoprene emissions 22% lower than MEGAN (60 vs 77 TgC/yr), and concluded that isoprene emissions are overestimated over the central African rainforest in the MEGAN inventory."

We have also added a paragraph on the posterior seasonal cycle, which is dramatically modified compared to the prior one:
"Over tropical regions (except Indonesia), the optimization shows dramatic changes in the seasonal cycles. For example, the posterior cycles over Northern Africa present 2 peaks (in September and in April), in both the wet and dry season, with highest values during the dry season. Over the region South American Tropical, instead of peaking at 4.2 TgHCHO in August like the prior, the posterior estimates peak at 2 TgHCHO in April (wet season) and at 2.1 TgHCHO in September (dry season). Interestingly, this posterior seasonal cycle agrees well with the in-situ tower measurements of isoprene, also showing two peaks, made for year 2002 at Tapajos National Forest in Brazil ([Barkley et al, 2008], their Fig. 8). "

In Section "Evaluation with Independent Data" (now Section 5.3), we have rephrased the sentence: "With significant modifications in terms of magnitude (monthly mean value of about 1.5 TgHCHO for the posterior, against about 3 TgHCHO for the prior) and in terms of seasonal variations (with peaks both in the wet and dry season) over Northern Africa,..."

> section 5.2.2, do we conclude that these regions are well constrained and accurate in a priori information or just not well constrained given the data that are available

A quantitative answer to this question would require performing the calculation of the error reduction for each region which is a huge computational task . However, the fact that satellite data used in this study cover tropical and temperate regions well makes it reasonable to assume that these regions are constrained by the inversion, although their magnitude change very little.
In section 5.2.2, we have rephrased the sentence: "Because of the small uncertainties prescribed on prior MCF emissions and MCF observations constraining the OH concentrations (and consequently on the loss of methane), the posterior global HCHO production by methane is only 2% lower than the corresponding prior (946 TgHCHO/yr)."

> p6927, l10-15. I don't quite follow here, especially the "amplify difference"

The difference found between Kopacz et al. [2010] and Fortems-Cheiney et al. [2011] on the North American mass budget is increased in this study. They found posterior emissions of 46.5 TgCO/yr, almost three times smaller than the budget of 127 TgCO/yr found with the MOPITT-only inversion [Fortems-Cheiney et al., 2011], and more than 3 times smaller the budget of 147 TgCO/yr found with this inversion.
We have re-phrased the paragraph and have added some information: "In our previous work with "MOPITT-only" inversion, we found CO emissions of 127 TgCO/yr, much higher than Kopacz et al. [2010] results (46.5 TgCO/yr). Here, the differences between our model and Kopacz et al. [2010] are increased as posterior emissions of 147.5 TgCO/yr are found. The cause of such a difference is still unclear. However, our value of 206 TgCO/yr for North America (6-year average) is in agreement with Hooghiemstra et al. [2012] who found 208 TgCO/yr and 202 TgCO/yr (inverted respectively with NOAA stations and with MOPITT for year 2004, their Table 1)."

> p6928, l14-19: I can't help wondering if CO emission and production offset each other here with respect to other studies

Indeed, the same CO fields can be modeled with CO production and loss spread differently between emissions and chemistry. Our system allows discriminating between the two, to some

extent.