**Reply to Anonymous Referee #1**

**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.**

Fortems-Cheiney and co-authors apply a multi-species inversion analysis with a 3D model to simultaneously constrain a 1) surface emissions of CH4, CO, and methyl chloroform (MCF) at 3.75x2.5 degree resolution; 2) CO, CH4 and MCF initial conditions (8-day, 3.75x2.5 degree resolution); 3) scale factors for 3-D production of HCHO from VOC oxidation (8-day, 3.75x2.5 degree resolution); and 4) scale factors for OH concentrations at 8-day resolution in 4 latitude bands. The analysis employs satellite observations from MOPITT and OMI (CO and HCHO) and surface observations (CH4, MCF) as the observational constraints in the inversion. They infer various changes to above 4 sets of variables on the basis of their analysis, and interpret those changes (briefly) in terms of the underlying drivers.

The analysis is novel in optimizing for these variables simultaneously, and in the way that it incorporates a range of space-based and in situ observational constraints to get at the questions of interest. It is also smart in the way that it exploits the interrelationship of the CH4-HCHO-CO system. The topic is certainly appropriate to ACP. The main weakness that I see is that it doesn't do a sufficient job of exploring and demonstrating the extent and resolution of the constraints that this observation and analysis system can really provide.

**The extent and the resolution of the constraints are rigorously quantified by computing the Bayesian posterior error covariance matrix. The high computational cost of this computation made us focus on some elements of this matrix only: the posterior variances for yearly CO and for yearly HCHO production by NMVOC in the 14 land regions of the globe and in the whole globe (Tables 1 and 2). This information from the theory is complemented with comparisons with independent measurements (Fig. 5) and with sensitivity tests involving critical variables of the inversion system (Fig. 10, Fig. 11 and Fig. 12).**

I have some specific comments below that I feel should be addressed before this is published.
——————— Scientific comments ———————
- I wonder how well we can expect the inversion to independently resolve all these different variables that are being optimized simultaneously. Clearly many of them are strongly coupled. For instance, how well can a bias in model OH be resolved from a bias in the surface fluxes of CO/CH4/MCF? I feel the authors need to demonstrate the robustness of their approach for us to give the findings full credence. For example, one could do this by generating pseudo-observations (with errors applied) with different aspects of the state vector perturbed, and test how well the inversion can get back to the true solution. And without messing up some other aspect of the state vector! Perhaps I'm wrong, but it seems likely there is some degree of conflation between those sets of variables being optimized.

**Contrarily to previous studies that prescribed the OH fields or the CO atmospheric production, we propose a chemically more consistent framework with species coupled and optimized simultaneously. We show that taking into account this complexity is important to estimate CO emissions, in order to avoid aliasing between atmospheric chemistry (production and loss) signals and the fluxes at the surface.**

**Making Observation System Simulation Experiments under various scenarios is certainly very interesting but constitute a full paper in itself, and is beyond the scope of this paper. We**

**have already performed a thorough evaluation of our results (by comparing our posterior modelled concentrations with independent surface and aircraft measurements, and by comparing our results with other recent studies).We have also performed several sensitivity tests, described in Section 6 (and also in Fig. 10, Fig. 11 and Fig. 12), to demonstrate the robustness of our approach.**

- I guess much of the constraint on OH is coming from MCF, especially since the assumed error on MCF emissions is only 1%. This 1% is not a physically reasonable assumption. Current MCF emissions are fairly small, but they are probably mostly from banks, stockpiles, landfills and so forth at this point, and so quite uncertain in a relative sense. For example, US emissions during 2003-2006 were shown by Hurst et al. (JGR, 2006) and others to be ∼60x larger than EPA estimates. How does your solution change if a more realistic uncertainty on MCF emissions is used? I'd be hard-pressed to think of any compound whose emissions are known to better than 1%! Especially on a grid-basis, as you're doing. We likely have a reasonable constraint on the global flux (though certainly not close to 1%), but much less so on a grid-cell by grid-cell basis.

**We do not use the EPA estimates for MCF prior emissions, but the inventory EDGAR-v3.2 given by Olivier et al. [2001]. EDGAR-v3.2 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]). This decrease represents the abatement of MCF emissions since the Montreal Protocol.**

**We have added this information in Section 4: "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]). This decrease represents the abatement of MCF emissions since the Montreal Protocol. Moreover, we made several sensitivity tests with respect to the prior MCF emissions errors settings (errors set at 5%, at 10% and at 100%). As seen in the following figure, and as an example for year 2005, these different error assignations do not change our solution on posterior MCF emissions, and consequently do not change our solution on OH concentrations.**
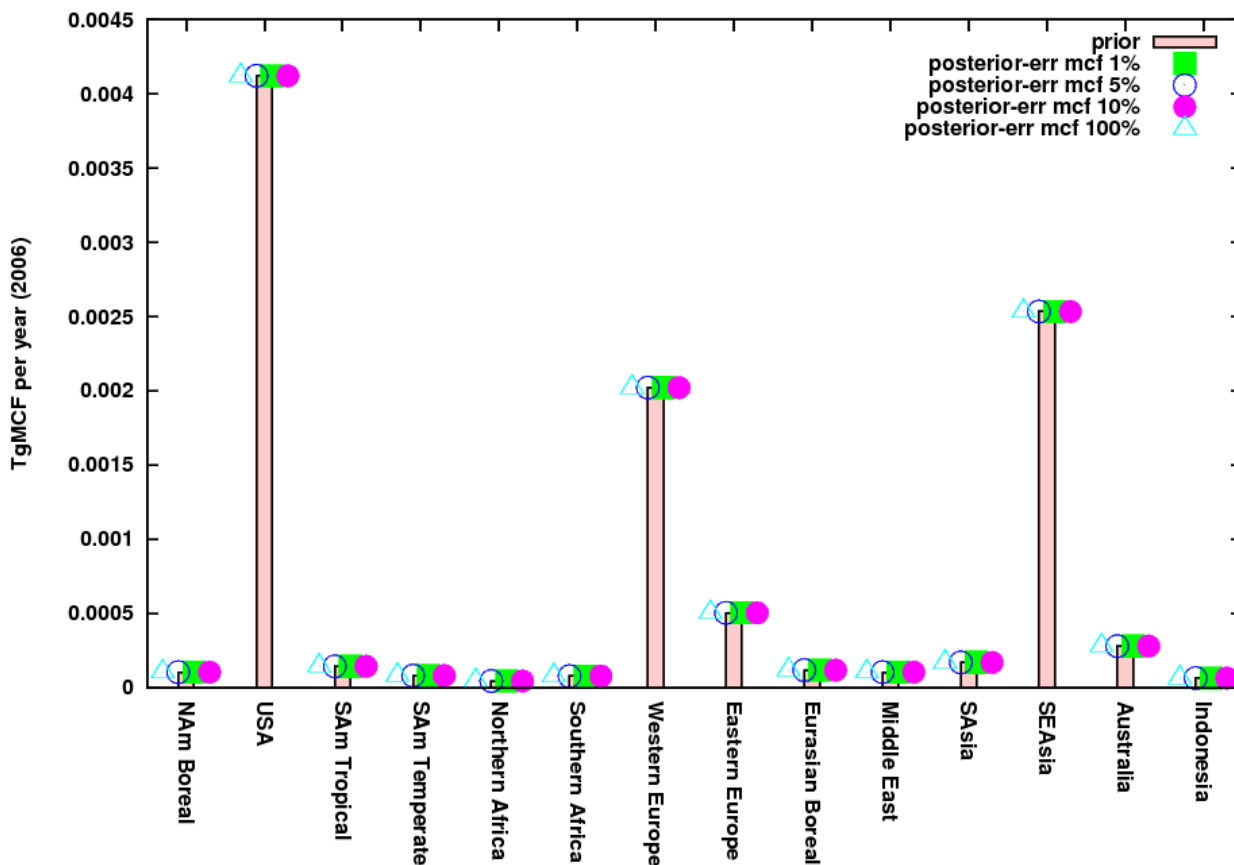
This can be understood because the magnitude of MCF emissions is very small for the 2005-2010 period: whatever uncertainties on prior MCF emissions are (even large changes of 100%), this would only have a very small impact on atmospheric concentrations. We have added one of these sensitivity tests (MCF prior emissions errors set to 10%) in Section 6.

- please discuss how uncertainty in model OH in high-isoprene, low-NOx environments might affect your solution. MCF provides a constraint on OH, but only in a mean sense as it has a 5-year lifetime. You could well have an OH bias that, in the tropics correlates spatially to a degree with one of the variables you are trying to solve for P(HCHO). What does it mean to be optimizing OH separately from and independently of P(HCHO), which in reality is driven by OH?
- Since your model includes the effect of OH on CO and CH4, but not vice-versa, is there potential for a result that is not self-consistent? In other words, CO and CH4 are the main global sinks of OH, but to my understanding this dynamic isn't captured in your model framework, only the effect of OH on CO and CH4.

We are aware of the simplification made in this work but it represents already an improvement compared to single-gas inversions usually performed so far, which do not account for interactions between gases at all, and in which OH is not optimized simultaneously with all other gases. We do not represent such refined processes yet, because of the high computing cost it would induce.

- to their credit, the authors carry out a bootstrap analysis to put some error estimates on some aspects of their solution. But they do not assess how their solution might change if different errors are assumed for the prior and for the observation system.

Sensitivity tests demonstrating the robustness of our system are now described in Section 6. We have also added Fig.10, Fig.11 and Fig.12.

"In this section, we discuss the robustness of our system from the spread of the regional HCHO production by NMVOC and of the regional CO and CH₄ emissions, in four sensitivity

**tests (cases A to D, described in Table 6) with respect to prior settings:**
**1. In case A, the OH field is replaced by OH-v2 field. The alternative field has also been derived from a simulation of the full chemistry model LMDz-INCA, but using another realistic emission scenario (the combination of anthropogenic emissions from IIASA, QUANTIFY for ship and GFEDv2 for biomass burning). OH-v2 field is within 5% of the reference OH field. All other settings are the same as in the reference.**
**2. In case B, HCHO prior 3D production by NMVOC error is set to 200%, instead of 400%.**
**3. In case C, CO prior emissions error is set to 150%, instead of 100%.**
**4. In case D, MCF prior emissions error is set to 10%, instead of 1%.**

**The HCHO production by NMVOC and CO and CH$_4$ emissions for the whole year 2006, found from each test, are given for the 14 regions in Fig.10a, Fig.11a and Fig.12a respectively. These posterior regional results are further compared with the reference inversion in terms of anomalies in Fig.10b, Fig. 11b and in Fig.12b. For the three fields (HCHO production, CO and CH$_4$ emissions), all sensitivity tests present the same departure from the prior as the reference inversion. The different sensitivity tests show very robust regional and global budgets.**
**For CH$_4$ (Fig. 12a), the differences in yearly methane emission remain below 2% at the global scale and range between less than 1% (Middle East, Southern Africa) to 12% (Indonesia).**
**The regional estimates of 3D HCHO production by NMVOC estimates obtained by the different sensitivity tests are also in a strong agreement. The largest range is observed over the regions Indonesia and South American Temperate (17% and 15% respectively), because of the smaller increments (compared to the reference) of case B and C. Nevertheless, these regional results are largely within the error bounds of the reference inversion (calculated with a robust Monte-Carlo approach, see Section 4).**
**The largest scatterings between the sensitivity tests are seen for CO emissions at regional scale. While on the global scale, the sensitivity tests show results within 2% of the reference inversion, the differences reach up to 39% at the regional scale. However, they are limited to 15% in all regions emitting about or more than 100 TgCO/yr (e.g, range of 14 TgCO over Southern Africa, for an annual reference budget of 133 TgCO), except for South East Asia (range of 51 TgCO for an annual reference budget of 208 TgCO).**
**The range of the inverted fluxes is well within the 1σ posterior uncertainty for all regions, except for the regions Middle East, South East Asia and Indonesia. Finally, it is worthnoting that the difference between the regional inverted emissions in cases B, C and D relative to the reference inversion is small, indicating that the inversion is not very sensitive to the prior errors statistics, at the scales of interest here."**

- model transport is another important source of potential error which is not addressed or discussed.

**We have added the information in Section 4: "The diagonal R matrix representing observation errors is filled with variances which combine representation errors (e.g., the mismatch between the observation and model resolutions), errors of the observation operator (including transport and chemical-scheme errors in LMDZ-SACS) and measurements errors. The errors on the observation operator and the representation error are difficult to estimate precisely. LMDz-SACS accumulates the errors of the reference model LMDz-INCA, complemented by those due to the simplifications made in the chemical scheme. As a consequence, we chose to define the variance of the individual observations errors in R as the quadratic sum of the measurement error reported in the MOPITT and the OMI data sets, and of the CTM errors set to 50% of the retrieval values following to Pison et al. [2009]."**

- The GEIA isoprene emissions are now more than 17 years old, and the following version,

MEGAN, has been available for more than 6 years. I wouldn't insist that the authors switch to MEGAN, but this does seem odd and in my view somewhat reduces the impact that their findings could otherwise have.

**The GEIA inventory is not the most recent estimate indeed, but we choose it for consistency with the study of Fortems-Cheiney et al. [2011]. Further, the annual global isoprene emissions estimated by MEGAN are similar to those of GEIA (e.g, 600 Tg against 570 Tg respectively for year 2003). The main conclusion (the overestimation of the NMVOC emissions in the GEIA inventory) would not be changed with MEGAN.**

- For the simplified (5-reaction) chemistry being employed, which assumes methane to formaldehyde in a single step, one thing that is neglected is the NOx dependence. For example at low NOx you're more liable to form methyl hydroperoxide, which would delay the HCHO production by a couple of days, and slightly reduce it to the extent that CH3OOH is deposited. I guess this would be a minor effect but worth mentioning.

**We agree. We have added the following sentence: "We assume that methane is oxidized into HCHO in a single step, thereby neglecting the formation of methyl hydroperoxide under low NOx conditions, that can delay or slightly reduce the HCHO atmospheric production."**

6921 and Fig 4. Why does the agreement get worse over W. and E. Europe?

**The agreement between the posterior and the OMI HCHO concentrations was worse only in terms of magnitude, but better in terms of seasonality (compared to the prior concentrations). Nevertheless, as now written in Section 4, the norm of the gradient of the cost function has been reduced by 98% (against 95% previously). The posterior HCHO concentrations now agrees as well as the prior ones (both in terms of magnitude and seasonality, see Fig. 4) with the OMI observations over Western and Eastern Europe.**

6923, when attributing changes in P(HCHO) as a result of the inversion to VOC emissions, how do you know it's not the HCHO yield that's wrong? Can you separate the two? Particularly in tropical or low-NOx areas, how do you know the timescale and/or yield for HCHO production in the model is accurate?

**We have assumed that the yield is perfectly known and we have not optimized it. It would be an interesting development, but also at a large computational expense as there are more than 40 chemical reactions to describe the HCHO atmospheric production by NMVOC. Moreover, we do not have adequate constraints to infer the yield of these reactions.**

6927, how do you reconcile this CO emission difference with the Kopacz and Hudman papers? Hudman et al. showed that their source estimate was consistent with aircraft observations. Given that, I don't see how a factor of 3 increase compared to their value is physically reasonable. "it can be due to an underestimate of the CO traffic emissions in the EDGAR-v3.2 inventory"? That could explain the change from your prior to your posteriori, but not the discrepancy between your posteriori and the other studies.

**The same CO fields (e.g, compared with aircraft observations) can be modeled with CO production and loss spread differently between emissions and chemistry. Large differences in terms of production and loss compared to Hudman et al. [2008] can explain large differences in terms of emissions. Hooghiemstra et al [2011, 2012] also pointed out discrepancies between their emission estimates for USA and those of Kopacz et al. [2010].**

**We have rephrased the paragraph: «"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)."**


> 6930, 10-11 "we demonstrated the robustness of our multi-species inversion system", not sufficiently, see comments above.

**In the submitted version of the manuscript, the robustness of our multi-species inversion system has been demonstrated through the following items:**
- **by comparing our results with other recent studies (for CO and for the HCHO production by NMVOC),**
- **by evaluating our posterior HCHO production by NMVOC and CO emission estimates with independent data,**
- **by calculating the uncertainty reduction associated with the posterior HCHO production by NMVOC and CO emission estimates.**

**We have now added sensitivity tests, described in Section 6, to show the reliability of our approach. We have re-phrased the sentence: «By comparing our posterior-modeled concentrations with independent surface and aircraft measurements, and by computing sensitivity tests, we demonstrated the robustness of our multi-species inversion system..."**

6917, is Fung et al. (1991) really the most current estimate for this?
**We know this is not the most current estimate, but we choose it for consistency with the study of Fortems-Cheiney et al. [2011].**


———————- Technical comments ——————-


> 6910, opening paragraph of introduction. By talking about the direct HCHO sources first, you give the impression that these are most important when in fact they are quite small.

**We now describe HCHO atmospheric production first.**


> 6911, 4-5 and 17: "HCHO modulates the budget of CH4, CO, and NMVOCs", and "HCHO and its interactions with CH4 and NMVOCs". Ok for CO, but for CH4 and NMVOCs this is odd phrasing, since HCHO is produced from them rather than the other way around. HCHO doesn't really modulate the budget of CH4 and NMVOCs, or interact with them, at least in any direct way. Did you mean indirectly as a source of Hox?

**We have re-phrased the sentence: "HCHO is a key species in the oxidation chain of methane and of NMVOC, and can modulate the budget of carbon monoxide (CO)." We have removed the part "HCHO and its interactions with $CH_4$ and NMVOC".**


> 6911, 8 (and also a similar comment at the beginning of 5.2.1): "NMVOCs and their short lifetimes". In fact lifetimes vary from hours or less to weeks or longer.

**We have re-phrased the sentence: "This is mainly explained by the diversity of NMVOC, their lifetimes varying from hours to weeks, and by the large spatio temporal variability of their emissions, leading to large uncertainties of bottom-up estimates (based on emission factors or biogeochemical models)."**


> 6912, 8: not sure I'd agree that there exists a "consistent global measurement network" for NOx. Unless you mean NO2 from space, but that isn't Nox.

**We have re-phrased the sentence: … "as it exists for greenhouse gases or other air pollutants such as CO, $CH_4$ and ozone."**


> 6912, 10: "these limitations can now be addressed by using HCHO total columns retrieved

by satellite". Careful not to make it sound like measurement errors are now a thing of the past – the satellite data of course have their own errors and uncertainty!

**We have re-phrased the sentence: "These limitations on the spatial coverage of the HCHO measurements can now be addressed by using HCHO total columns retrieved by satellite, which offer the unique possibility of sensing atmospheric HCHO at a global scale. Even though uncertainties remain large for HCHO satellite retrievals, past studies have demonstrated the usefulness of HCHO column data to constrain NMVOC emissions".**

**Please note that we also mentioned the measurements error in the conclusion: "While OMI provides a daily coverage dataset at a high spatial resolution, uncertainties remain large for OMI".**

6912, 18-19: "However, none of these studies combine retrievals for various species together". That's not a true statement. What about Stavrakou et al. (ACP, 2009) who inverted simultaneously for HCHO and CHOCHO? Also, Barkley et al. (JGR, 2008) and Marais et al. (ACPD, 2012) both combined space-borne HCHO and NO2 measurements, though NO2 was used as a diagnostic for fires and chemical regime rather than as something to be optimized.

**We have removed the sentence "However, none of these studies combine retrievals for various species together" and we have added the three cited references.**

6914, 4: On the ACPD page for the Kim et al. paper, it says "publication in ACP not foreseen". Nothing against that paper in particular, but what is the protocol for citing papers that did not make it through peer review? Do we want to be doing that?

**We have removed this reference.**

6914, 7: since TPK is a co-author it seems unnecessary to cite a personal communication in this way.

**We have removed it.**

6915, 23: Also a minor ocean sink for MCF, cf Wennberg et al. (GRL, 2004).

**We have re-phrased the sentence: "MCF only reacts with OH and its sources and sinks (emissions, photolysis, ocean sink) are assumed to be quantified with a rather good accuracy."**

6925, 5-8, unclear, awkward.
**We have removed it.**

- Section 5.2.3 – Section on changes to the global HCHO sink. Doesn't the change in the sink just come about as an inevitable consequence of the change in the source? Doesn't seem to need its own section.

**For the sake of completeness we think it is useful to include a section on changes to the global HCHO sink. We have kept it.**

section 5.3.1, "copares", "fulle"
**We have changed these words.**