Review of New Constraints on Biogenic Emissions using Satellite-Based Estimates of Carbon Monoxide Fluxes by Worden et al.

Anonymous Referee #1

This paper deals with the top down estimate of biogenic CO emissions based on the GEOS-Chem model constrained with MOPITT observations. The paper brings interesting results about biogenic CO sources and their seasonal variability. The method provides improved estimation of these emissions. The paper is well structured, clear and well written. It should therefore be published in ACP. Nevertheless, the methodology and results that looks solid are often described too briefly. Some more detailed explanations should be given for some specific points that are detailed below.

We thank the reviewer for their effort and useful comments and questions. We think that addressing these concerns will improve the manuscript. Our responses are embedded below in blue italics with modified or new text.

1) P3: it is mentioned that 3 different MOPITT products are used (columns, full profiles

and tropospheric profiles) to empirically evaluate errors due to transport. How is this error estimate integrated in the total error of the posterior fuxes? What are the error values?

Please see response to #4 below

- 2) P4: could you provide details about prior BB uncertainties? Some values? *Please see response to #4 below*
- 3) P4: why 50% is assumed for BIO and FF prior flux estimates Is this value coming from sensitivity tests with varying uncertainties? Is this the value that provides the best fit between model and observations? This choice should be discussed as well as the metrics and methodology used to evaluate the improvement of the modeled CO distributions relative to the MOPITT observations. And the criteria used to decide that convergence is reached.

These uncertainties were chosen based on previous experience with error constraints and the objective of allowing the sector emissions to vary sufficiently to test new probability distributions within each grid cell. While a more complete sensitivity test would be desirable for future top-down inventory partitioning, our main goal for this manuscript was to demonstrate that this technique has skill in terms of reproducing the seasonal and spatial variability as found independently in top-down isoprene estimates using OMI HCHO observations. We will add the following to the text (section 3) where we state the use of  $\pm$  50%.

"This choice of uncertainty for the BIO and FF sectors is based on previous experience with error constraints and allows sufficient variability in the sector emissions for testing new probability distributions within each grid cell".

4) P5: the average posterior errors ar given. The different contributions to the error have been mentioned previously (such as the empirical transport error) but we do not have a clear idea about the complete budget. An equation indicating the different contributions to the posterior error and the contribution of each error source to the total error given here would be of interest.

P.4 Eq. 1 shows how the probability distribution is re-partitioned based on the errors assumed in each sector, and we have added equations 3-5 to show how the probabilities and cost function are computed with the associated apriori uncertainties. The posterior errors (1-sigma) are obtained from the sampling the MCMC probability distributions that are unique for each grid cell and month. We now include the following new table and text in section 4 to help the reader understand the error sources and average outcomes for the tropical regions of interest in this study.

Table 1. Uncertainties applied in the Bayesian source attribution (Eq. 1). Values are monthly averages for single grid boxes (5° x 4° longitude x latitude) in the tropical study regions.

CO sector distribution	A priori source	A priori uncertainty	Average Posterior Uncertainty (tropics grid boxes)
Total flux top-down estimate	GEOS-Chem Inversion based on MOPITT V7J CO data (Jiang et al., 2017)	$\sigma_F$ ± 50% (assumed)	± 12% average constraint <sup>a</sup> , with 11% 1- sigma standard deviation for tropical grid cells <sup>b,c</sup>
BIO direct + biogenic NMVOC oxidation	MEGAN v2.0 (Guenther et al., 2006)	$\sigma_{BIO_{ap}}$ ± 50% (assumed)	± 24%
BB biomass burning	GFED4s (van der Werf et al., 2017)	$\sigma_{BB_{ap}}$ ± 24% (Akagi et al., 2011)	± 22%
FF fossil fuels	EDGAR 3.2 (Olivier and Berdowski, 2001)	$\sigma_{FF_{ap}}$ ± 50% (assumed)	± 45%

<sup>&</sup>lt;sup>a</sup> The total flux posterior error is estimated from 3 flux inversion types (see text for description) to approximately account for model transport errors.

<sup>&</sup>lt;sup>b</sup> Average and standard deviation are computed for tropics (20°S to 20°N) using grid boxes with with emissions > 0.1 gCO/m2/month.

<sup>&</sup>lt;sup>c</sup> The variance in tropical grid cell flux errors includes both spatial and temporal variability, however, these errors have not been weighted to account for sampling effects, such as inflated errors due to fewer MOPITT observations during rainy seasons.

"Uncertainties are available by 5° x 4° grid cell, month and source sector (BB, FF or BIO) and represent the 1-sigma width of the posterior distributions; these distributions are critically dependent on the a priori uncertainties and therefore subject to change when different a priori distributions and covariances are assumed in the Bayesian attribution approach. Table 1 lists the sources of a priori data and uncertainties and gives average monthly values representative of the individual grid cells used in this study. For the remote tropical regions considered here, FF contributions to total CO fluxes are small and we find the most improvement over prior errors in BIO CO posterior flux uncertainties, especially in months with little or no BB emissions. This can be seen in Fig. 2, where monthly grid box posterior errors were averaged spatially for the region of interest and over years 2005-2012. One of the assumptions in this study is the prior uncertainty in BB, which only considers emission factor uncertainties (Akagi et al., 2011) and does not explicitly account for other factors in BB CO fluxes such as combustion completeness and biomass (fuel) amount (e.g. Bloom et al. 2015). Future work will examine the effects of using a wider range of prior uncertainties that reflect multiple inventories."

1) P5: it is unclear to me why posterior error for FF is twice larger than for BIO and BB. I would have expected that this source is better constrained in the prior inventory. And why MOPITT constrain this source much less than the 2 others? Could the authors elaborate on this point?

The FF component is very small in the tropical regions we consider so there is little information to improve on the FF error compared to the prior. This was already stated in the text, but revisions to address the comment above (e.g., error table) make this more explicit.

2) P6: the present study finds BB emissions (290 Tg/yr) of about 1/3 of those from Folberth et al. 2006 (811 Tg/yr). It is a large difference that is briefly justified by the fact that tropical fires have declined during the 2005-2012 period relative to the one used in Folberth et al. 2006 according to Andela et al. (2017). Could you give more details to convince the reader?

This could also be an overestimation in the BB CO emissions considered by Folberth et al (2006). Recent estimates using GFED4 (van der Werf et al., 2017) report annual mean emissions for the 1997-2016 period for CO as 357 Tg/yr, while Granier et al., 2011 reported a range of 414 to 509 Tg/yr for 6 inventories in the 1997-2000 period, a period with significant interannual variability due to the strong 1997-1998 ENSO episode. We will modify the text to state:

This contribution from BIO CO represents a larger percentage (~41%) of the sum of BB, FF and BIO CO sources than expected (~27%) based on Folberth et al. (2006) which has 811 Tg(CO)/yr for BB and 672 Tg (CO)/yr for FF). However, there is a wide range in reported biomass burning emission estimates, with large interannual variability. Stavrakou et al., (2006) used 467 Tg(CO) for the year 2000 as the BB CO a priori from GFEDv1; van der Werf et al., (2017) reported 357 Tg/yr mean emissions for BB CO over 1997-2016 while Granier et al., (2011) reported a range of 414 to 509 Tg/yr for 6 emission inventories in the 1997-2000 period. Because our 2005-2012

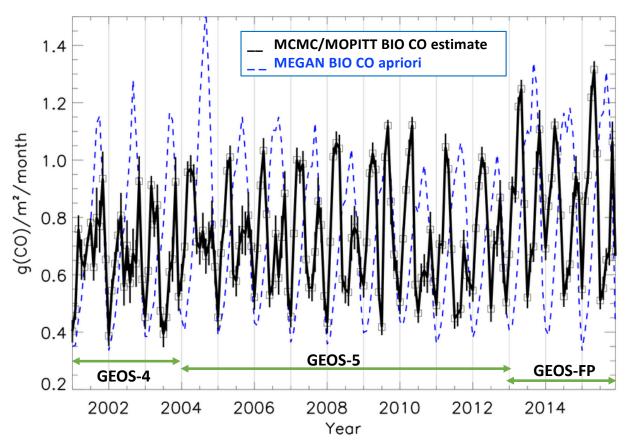
study period did not include the significant ENSO episodes in 1997 and 2015, we would expect lower average values for BB CO emissions than these other annual averages. Furthermore, in recent decades, there is a decreasing contribution of BB CO associated with a decline in tropical fires (e.g., Andela et al., 2017), as well as declining FF CO emissions (Yin et al., 2015; Strode et al., 2016; Jiang et al., 2017; Zheng et al., 2018).

3) P7: how is the posterior estimate affected by the change in forcing fields (GEOS FP versus GEOS-5? Is the top down method more robust to such changes than MEGAN?

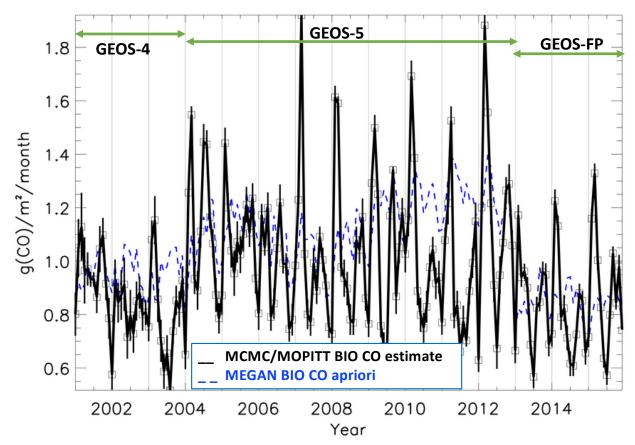
Although there does appear to be less dependence on the version of the meteorological fields in the posterior results compared to the MEGAN apriori (see Response Figure 1 below for the N. African savannas region), we did not want to draw conclusions, especially about trends in biogenic fluxes, without more consistent meteorological fields. Also, the dependence on the prior can still vary from region to region depending on the errors in the other emission terms (BB and FF). This is more obvious in Response Figure 2 for the Equatorial Africa region where there is more interference from BB emissions, and the time dependence of the prior is more clearly affecting the posterior result. Therefore, we chose the 2005-2012 period for the analysis in this paper.

4) P7: the results concerning the seasonality of the biogenic emissions are very interesting. The coincidence of isoprene and CO bimodal variability gives confidence in these results. Nevertheles, it is a bit desappointing not to have more explanations about the discrepancy between biogenic emissions and LAI variabilities! Are there some possible explanations? Why temperature plays a controling role in this N African Savannahs?

Marais et al., (2014) originally found that surface layer temperature dominates over LAI for controlling isoprene emissions in the N. African savannas region. Cooler temperatures during the monsoon (July-August) due to cloudy conditions induce a minimum in isoprene emissions that coincides with peak LAI, so LAI and isoprene emissions appear decoupled. At the same time, MODIS observations could underestimate LAI during the rainy season due to cloud contamination, but since LAI is not the main driving factor for isoprene emissions, especially in tropical regions, we do not expect this to explain the discrepancies of top-down observations with MEGAN. The Marais et al., (2014) reference is already cited, so we have not modified the text in response to this comment. Furthermore, the study presented here is meant to demonstrate the methods that we will build on in future work to test the processes and potential changes needed in MEGAN to reproduce top-down estimates of biogenic emissions.



Response figure 1. Timeseries of apriori (dashed blue) and estimated CO flux (solid black, with error bars) for the N. African savannas region. Green arrows indicate the different time periods for the GEOS-4, GEOS-5 and GEOS-FP meteorological fields used to calculate the apriori with the MEGAN model and for the inverse analysis for total CO flux.



Response figure 2. Timeseries of apriori (dashed blue) and estimated CO flux (solid black, with error bars) for the <u>Equatorial Africa region</u>. Green arrows indicate the different time periods for the GEOS-4, GEOS-5 and GEOS-FP meteorological fields used to calculate the apriori with the MEGAN model and for the inverse analysis for total CO flux.

## References to be added:

van der Werf, G. R., Randerson, J. T., Giglio, L., van Leeuwen, T. T., Chen, Y., Rogers, B. M., Mu, M., van Marle, M. J. E., Morton, D. C., Collatz, G. J., Yokelson, R. J., and Kasibhatla, P. S.: Global fire emissions estimates during 1997–2016, Earth Syst. Sci. Data, 9, 697-720, https://doi.org/10.5194/essd-9-697-2017, 2017.

Granier, C., Bessagnet, B., Bond, T. et al.: Evolution of anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980–2010 period, Climatic Change (2011) 109: 163. https://doi.org/10.1007/s10584-011-0154-1