Interactive comment on “Decadal trends in global CO emissions as seen by MOPITT” by Y. Yin et al.

Reply to Referee #1

We thank the reviewer for her/his thoughtful comments and valuable suggestions. We have addressed all the issues raised in the review. The full review is copied hereafter in blue and our responses are inserted where appropriate.

> ***Anonymous Referee #1***
> *This paper uses as data-assimilation framework to infer CO emissions from satellite observations. The CO inversion system is coupled to CH2O, CH4, and MCF, which provide constraints on the sources and sinks of CO. The target period of the study is a full decade (2002-2011). The study has the aim to provide a consistent analysis of the drivers of the observed decline in CO total columns from MOPITT (figure 3). Although the paper is relative convincing in some aspects, some inconsistencies are also apparent, which need to be better explained or analyzed. Below, these are listed under major issues.*

We thank the reviewer for pointing out some elements that we did not articulate well enough in the previous version of the manuscript. We hope that this reply clarifies these issues.

> *Major issues*
>
> *1. Is the system well balanced?*
> *The focus of this paper is on CO, but also CH4 and MCF measurements are assimilated. One of the burning questions around today is the role of OH, and a possible trend in OH, in the observed CH4 growth rate changes, exactly in the analyzed period. I was therefore a bit disappointed to find only one sentence: “Similarly for CH4 and MCF, the inversion fits the assimilated data fairly well, but these results are not shown, as they are not the main focus in this study”. This casts doubts on the added value of the CH4 and MCF assimilation.*

In the revised paper, we will add the following table (Table 2) that presents the mean model-data bias and the residual root mean squares (RMS) before and after optimization for CH4 and MCF.

Table 2. Fitness of CH_4 and MCF observations assimilated in the inversion.

Region	OH-type	CH_4 (ppb)				MCF (ppt)			
		Mean bias		RMS		Mean bias		RMS	
		prior	*posterior*	*prior*	*posterior*	*prior*	*posterior*	*prior*	*posterior*
NH(30-90)	*TransCom*	20.6	2.4	749.6	22.2	1.02	-0.02	1.14	0.05
	INCA	-21.0	2.5	658.1	20.1	0.44	-0.09	0.28	0.07
NH(0-30)	*TransCom*	15.8	1.5	452.6	19.1	0.90	-0.20	0.91	0.16
	INCA	-21.1	-0.1	564.9	13.9	0.40	-0.20	0.21	0.11
SH(0-30)	*TransCom*	10.9	-2.4	222.1	20.1	1.19	0.09	1.61	0.13
	INCA	-14.3	-4.0	264.9	29.5	0.88	0.23	0.94	0.15
SH(30-90)	*TransCom*	14.4	0.2	308.3	4.7	0.88	-0.24	0.90	0.17
	INCA	-7.9	-0.4	96.6	5.7	0.60	-0.06	0.46	0.07

The substantial decrease of both the mean bias and RMS reveals the system ability to draw information from the CH4 and MCF observations.

> *I presume that the cost function consist of a term related to model-data mismatch of MOPITT observations, a background term (emissions of CO, MCF, and CH4 that deviate significant from the prior), and terms related to CH4 and MCF misfits at the stations. If such a system is not well balanced, it might be that little of no information is drawn from the MCF and CH4 misfits. Since the authors claim that they infer no trend in OH they have to verify if their system is adequately set up to detect a possible OH trend. Stating that the inversion fits the MCF observations fairly well is certainly not enough.*

Estimating whether the system (or the cost function) is balanced among species is far from trivial. For the prior error covariance matrix **B** and observation error covariance matrix **R**, we assign the error terms separately based on our best knowledge of their uncertainty in the "physical" world based on bottom-up inventories and previous model studies. This is consistent with the Bayesian approach where each information piece brings its own probability density function independent from the other ones: we do not tune the error statistics to artificially balance the weight of each species.

In terms of observation numbers of different species included in the assimilation, which could influence their relative shares in the cost function, MOPITT CO retrievals have higher spatial coverage over the globe so they have implicitly more observations than CH_4 and MCF. Furthermore, model-data biases could also contribute a potentially large portion of the initial cost function (in our case the model bias is larger for CO than for other species) and we do not think that there is a necessity to "balance among species" in terms of a misfit correction.

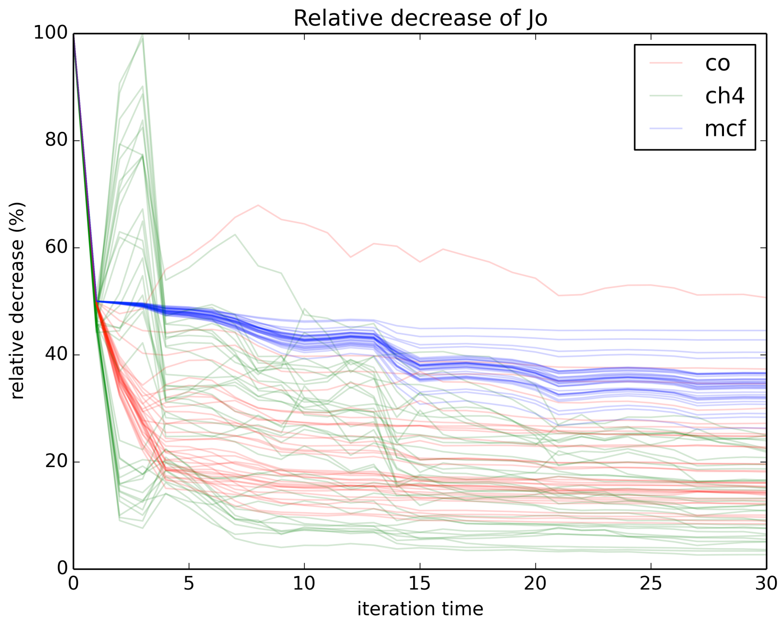
> *Moreover, it would be interesting to present some analysis of the cost function, showing*

how the optimization changes the cost function, and how CH4 and MCF observations are used to inform the CO budget in terms of sources and sinks (e.g. by neglecting couplings).

For the reasons stated above, we think that balancing the absolute share of the cost function from each species is not appropriate for the study. Instead, we have analyzed the relative contribution of the three species in the observation term of the cost function during the iterative minimization.

$$J_o = \frac{1}{2}(H(\mathbf{x}) - \mathbf{y})^{\mathrm{T}}\mathbf{R}^{-1}(H(\mathbf{x}) - \mathbf{y})$$

Fig. S2. Relative decrease of Jo for each species for each year during iteration. CO are shown in red, methane in green and MCF in blue.



In this figure, each line represents the relative decrease in *Jo* for one species for a year during each iteration. It shows an efficient decrease compared to the initial cost for all species. For methane (in green) and MCF (in blue) observations at surface station, which are more difficult to represent by the model than satellite CO column retrievals averaged at model resolution (in red), the relative decreases are smaller in some occasions. Nevertheless, the inversion system appears robust at minimizing the cost function.

2. Are the results realistic?
A large fraction of the atmospheric CO comes from the oxidation of NMHCs. Yet, figure 6 shows that large seasonal biases exist with independent satellite observations of CH2O. This implies that the atmospheric CO sources are also seasonally biased, and these biases will be projected on CO emissions.

We agree that assimilating CH_2O observations could further improve the estimation of CO emissions, in particular to improve the seasonal cycle. It is actually one of our perspectives in this paper. However, the observational data of CH_2O is not fully available for the decade that we study; thus, for the sake of consistency we do not assimilate CH_2O. Instead, the OMI CH_2O retrievals are used as independent evaluation. Fig. 6 shows, on the one hand, the overall agreement of satellite CH_2O column retrievals with CH_2O production from methane and NMVOC oxidation for latitudinal means. On the other hand, it shows that no significant large-scale trends are found in the CH_2O column to contribute to the decrease of CO concentrations that are observed over many regions.

> *Even more worrying are the large regional emission increments that are presented in figure 9. For instance, in the region SHSA the emissions are calculated to increase from roughly 50 Tg/yr in 2002 to almost 200 Tg/yr.*

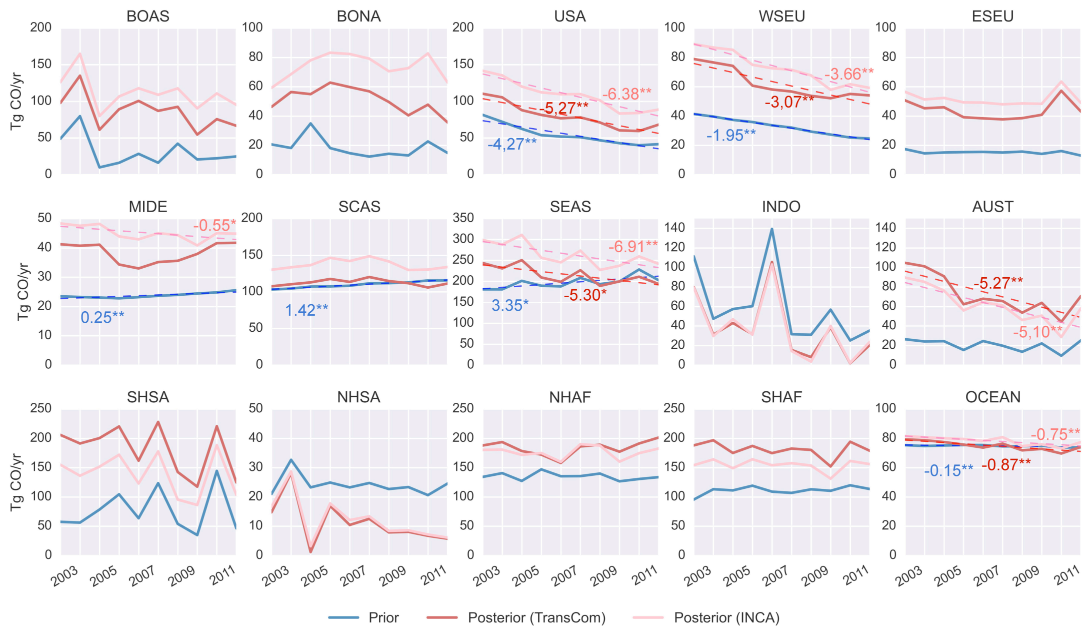
We agree that the relatively large increments in South Hemisphere South America (SHSA) and Australia have large uncertainties due to CH2O production, OH sink and other factors concerning transport and vertical mixing. However, our study here focuses on the trends of CO emissions, and the regional uncertainties are more systematic than time dependent. Therefore, regional increments from the prior to posterior as well as their relative uncertainties are not specifically discussed. Further investigation of this point for a different study will be very interesting.

> *In later years calculated increments are smaller but the recently described biomass burning year 2007 (Bloom et al., GRL, 2015) visible in the prior seems to disappear in the posterior. Over Australia and Africa also some large increments are calculated. Likely, the two issues are related since CH2O from isoprene is a major source of CO over SHSA (Stravakou et al., ACPD, 2015).*

We thank the reviewer for pointing this out. We traced the problem back to an artefact caused by the way we processed the whole decade in the initial paper. Indeed, for practical reasons, we did split the period into segments of 14 months with a 2-month overlap between adjacent years. We thought that this approach would not influence the inverted flux trends under the assumption that the initial conditions for each segment are properly optimized by the system with just 2 months of overlap, a spin-up time that is about the lifetime of CO in the atmosphere. Under closer scrutiny, it appears that it actually generates a small discontinuity in the concentration time series that affects some regional signals. We have re-run the inversion with only three segments and with an overlap of 4 months between adjacent segments with initial conditions optimized upstream. The main conclusions are not affected, but all figures will be updated.

The updated Fig. 10 is inserted below. High emissions in regions south hemisphere South America (SHSA) and Indonesia (INDO) in the year 2007 are very visible now in the posterior, consistent with the prior and other bottom-up studies.

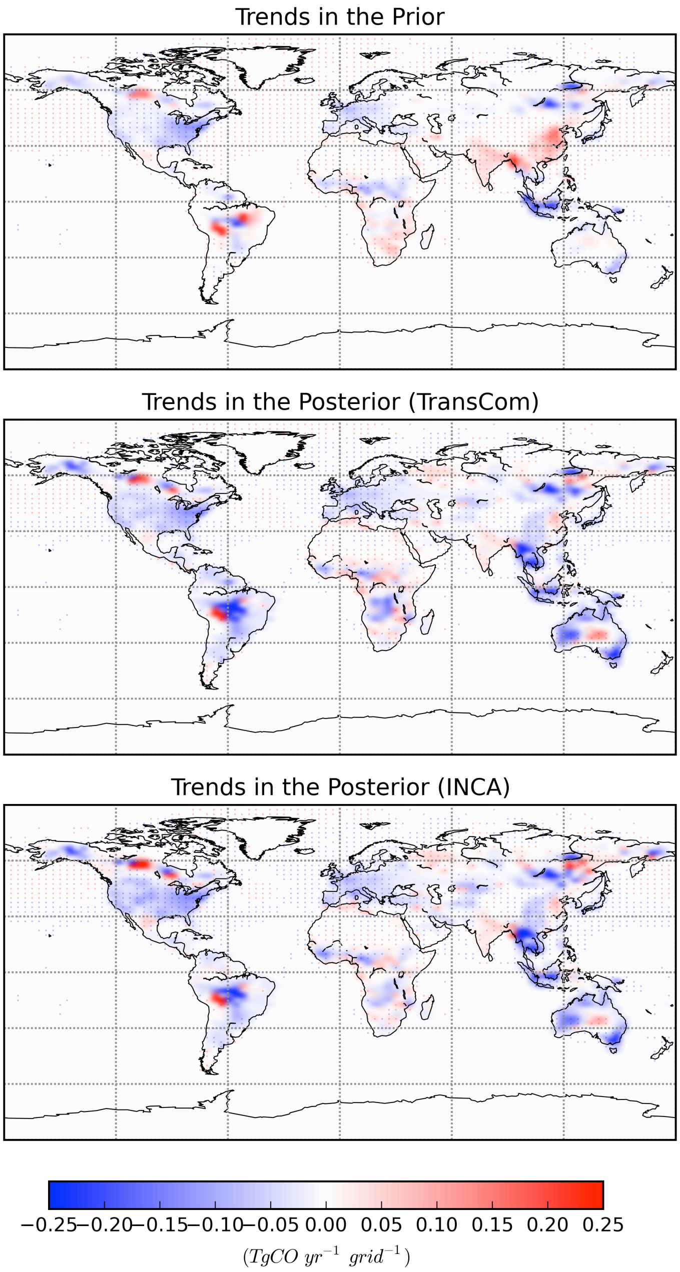
Fig. 10. Regional annual CO emissions from 2002 to 2011. The prior emissions are shown in blue and the posteriors are shown in red. Dark-red represents posterior with TransCom-OH and pink-red represents posterior with INCA-OH. Linear fit is shown when statistically significant, and slopes are denoted with corresponding colour. * denotes significant at 95% confidence level, and ** denotes significant at 99% confidence level. The notations for the sub-regions are listed below, and the extent of each region is shown in Fig. S1. BOAS - Boreal Asia, BONA - Boreal North America, USA - USA, WSEU – West Europe, ESEU – East Europe, MIDE – Middle East, SCAS – South Central Asia, SEAS – South East Asia, INDO- Indonesia, AUST – Australia, NHSA- North Hemisphere South America, SHSA - South Hemisphere South America, NHAF - North Hemisphere Africa, SHAF – South Hemisphere Africa, OCEAN – all ocean emissions.



Finally, only the results for the TRANSCOM-OH are shown. These fields have a NH/SH OH ratio closer to 1 and this will surely influence the NH/SH CO emissions (Patra et al., Science, 2014). On the global scale the CO budget might not be influenced too much by the OH field, but given the importance of OH as CO sink, some analysis and discussion about this issue is also needed.

As shown in the updated Fig. 10 above, for the time series of regional annual emissions we now present both the posterior using TransCom and posterior using INCA OH field. Indeed, the distribution of OH field has a considerable impact on the regional budgets. TransCom OH has lower concentration in the NH compared to INCA, thus the inverted emissions using TransCom OH in the NH are generally lower compared to the one using INCA. On the contrary, in the SH where TransCom OH concentration is higher than INCA, the inverted emissions using TransCom OH generates higher CO emissions than INCA. The differences of the two posterior using contrasting OH fields are of a smaller magnitude than the differences between the posterior and the prior. It is noted that the trends in the posterior emissions are very consistent, irrespective of the OH fields used as shown in the figure below (updated Fig. 9).

Fig. 9. Trend distributions of CO surface emissions from 2002 to 2011 (a) in the prior, (b) in the posterior with TransCom-OH and (c) in the posterior with INCA-OH.



Minor issues:

14507, 23: TES is probably not referring to the "Technology Experiment Satellite", but to the Tropospheric Emission Spectrometer.

Thanks for point it out. We have corrected it.

14508, 1: "The interpretation : : :": sentence reads awkward, rewrite.

Done. We have revised it as "Comparing modelled and observed trends is complicated by the common biases of atmospheric chemistry-transport models (CTMs) forced with current emission inventories."

14508, 8: "Understanding this model-data misfit is all the more so challenging that surface emissions and chemical production each account for about a half of the total CO sources" ! "Understanding this model-data misfit is challenging because surface emissions and chemical production each account for about half of the total CO source".

Thanks for the edits. It has been implemented as suggested.

14508, 11: can contribute ! could have contributed

We have made the change.

14508, 20: information piece ! piece of information

We have made the change.

14508, 27: to infer the origin of the observed CO concentration decrease in the past decade ! to infer the most likely origin of the observed CO concentration decrease over the past decade

Thanks. We agree it is more accurate stating it in the way as suggested.

14509, 2: remove "at once"

We have made the change.

14509, 4: "chemically connected to hydrocarbons"? unclear

We have replaced this part of the sentence by "a species that only reacts with OH and therefore informs about it".

14509, 13: "The algorithm has undergone continuous improvements and several reprocessings of the archive have been made (Streets et al., 2013)." The algorithm has undergone continuous improvements and the archive has been reprocessed several times (Streets et al., 2013)" By the way: is the Streets et al reference correct? It is not in the list, like the Cressot (2014) reference. Please check all references!

We have made the change in the text as suggested. Thanks for spotting this mistake in the reference. The corresponding reference should be Deeter *et al.,* 2013. In addtion, we have double-checked all the references.

14511, 6: this requires some explanation. If you use a "fixed OH field", it is important in know how this has been obtained, and possibly what was the role of NOx in obtaining these fields.

The description was a bit confusing in the old text and we have edited the sentence. The prior OH field is modeled with full chemistry models with NOx and many other processes considered. The corresponding references for INCA OH is Folberth *et al.*, 2006; and the one for TransCom is Patra *et al.*, 2011 and references therein. In the optimization process, we optimize OH scale over big regions assimilating MCF, CH4 and CO as shown in Fig. 1, during which the effect of NOx is not relevant.

> *14511, 14: pressure weighted concentration? Is that not simply a mixing ratio?*

Yes. The text has been simplified following the suggestion.

> *14511, 17: CH2O has also direct source from biomass burning (Stravakou et al., ACPD, 2015). How do you account for surface emissions in this procedure?*

There are direct CH2O emissions (as shown in Figure 1) in our model both from biogenic and biomass burning emissions in addition to the CH2O chemical productions from VOC oxidation. The surface emissions are optimized together with the total quantity VOC emissions that will turn into CH2O (there is a single scaling factor for both).

> *14511, 27: as described below*

We have added the preposition "as" as suggested.

> *14512, 18: the Cressot reference is not in the list.*

It has been added.

> *14513, 12: Leeuwen ! van Leeuwen*

Thanks for pointing it out. It has been added.

> *14513, 13/16: m2 ! m-2*

It has been changed.

> *14513, 23: month? The period is 8 days, right?*

For our optimization, the period is 8 days; for the prior emission, we use 8-day resolution when the datasets are available. However, the prior emission datasets do not necessarily have such high temporal resolution, in particular for anthropogenic (fuel combustion) emissions. For biomass burning emissions whose daily emissions are available from GFEDv3.1, we calculate weekly mean emissions from the daily emission data. We assume the temporal variability is larger for biomass burning emissions for both CO and CH_4. But for other emission sectors, monthly mean emissions are used as the prior.

> *14515, 25: what about observational errors? Probably smaller than model errors, bur still good to mention.*

In the revised text, we have added the global mean measurement error for all the species. "The global mean measurement error for Xco is around 6.4±2.9 ppb, which is approximately 8.2±1.9 % of corresponding Xco observations. The measurement errors are set as 3 ppb for CH4 and 1.2 ppt for MCF if not explicitly provided by the surface observation datasets."

> *14515, 26: how is the yearly mean of the synoptical variability defined? DO you apply the same filter as for the 3-sigma filtering?*

No. The variability of synoptic variability was calculated upstream.

> *14517, 8: whatever ! irrespective*

This has been changed as suggested.

14518, 8: I suggest to add something like: "when for instance the vertical mixing in the model is too conservative, this could lead to a positive bias at the surface, because the sources are adjusted to fit the satellite data."

Thanks for the suggestion. This example has been added in the text.

14518, 16: Negative ! A negative

We have made the change.

14519, 1: A logical discussion here would be: what are the trends in the direct prior CO emissions from anthropogenic activities and from biomass burning? I see this discussion later...so please point forward to that discussion.

Section 3 focuses on the concentrations, so the underlying attribution from anthropogenic or biomass burning is not yet discussed.

14519, 2: To compare the trend in columns to trends at the surface, please convert the column in a mean mixing ratio.

It is done in the new version. We have converted all the column data from molec cm^{-2} into volume mixing ratio (ppb).

14520, 7: I find this not very convincing. To my eye, for at least some stations, it seems the prior simulation reproduces observed trends better than the posterior simulation. So, why not provide the information in a table? (e.g. average improvement of trend).

The trends are now summarized in the following table (Table 3) with information on both the mean bias and the trends.

Table 3. Summary of CO model-data comparision and trend analysis for MOPITT satellite retrievals, surface station observations and corresponding prior/posterior modelling. Trends for each region (in the unit of ppb yr^{-1}) are the mean values for all the grids whose trends are significant at 95% confidence level. The percentages of significant trends are also given per model grid for positive (+) and negative (-) respectively.

REGIONS	MOPITT		Surface		MOPITT column trends										Surface station trends									
	Prior Bias	Post Bias	Prior Bias	Post Bias		Observation			Prior mod			Posterior mod				Observation			Prior mod			Posterior mod		
	(ppb)				*N*	Trend	+ %	- %	Trend	+ %	- %	Trend	+ %	- %	*N*	Trend	+ %	- %	Trend	+ %	- %	Trend	+ %	- %
BONA	-14.1	0.7	-20.8	20.5	*210*	-0.84		*99*	-0.46		*95*	-0.88		*99*	*2*	-2.51		*100*	-1.46		*100*	-2.96		*100*
USA	-16.7	-3.1	-20.4	20.1	*108*	-0.82		*97*	-0.41		*96*	-1.08		*100*	*3*	-1.67		*67*	-1.13		*100*	-3.30		*100*
NHSA	-10.0	-3.0			*74*	-0.61		*84*	-0.19	***8***	*53*	-0.68		*100*	*0*									
SHSA	-14.2	0.2			*160*	-0.59		*58*	0.38	***41***		-0.56		*67*	*0*									
NHAF	-15.0	-2.0	-20.0	6.8	*211*	-0.45	***7***	*55*	-0.38		*85*	-0.73		*95*	*1*	-0.90		*100*	-0.73		*100*	-1.04		*100*
SHAF	-16.5	0.5	-1.5	13.6	*96*	-0.57		*75*	0.07	***27***	*11*	-0.64		*96*	*1*	-0.78		*100*	0.42	***100***		-0.48		*100*
WSEU	-16.1	0.3	-36.7	18.7	*106*	-1.00		*100*	-0.49		*100*	-1.12		*100*	*6*	-2.73		*100*	-2.05		*100*	-3.51		*100*
ESEU	-16.8	0.4			*108*	-0.77		*100*	-0.40		*100*	-0.93		*100*	*0*									
BOAS	-17.3	1.0			*227*	-0.92		*99*	-0.51		*92*	-1.02		*99*	*0*									
MIDE	-16.5	-2.9			*64*	-0.57		*100*	-0.30		*100*	-0.88		*100*	*0*									
SCAS	-12.0	-0.3			*80*	-0.65		*63*	-0.30	***4***	*38*	-0.92		*100*	*0*									
SEAS	-20.1	-3.6	-30.6	22.6	*129*	-1.23		*97*	0.13	***19***	*24*	-1.35		*99*	*3*	-1.76		*100*	-1.83		*100*	-3.78		*100*
AUST	-15.5	-1.7	-6.4	15.4	*105*	-0.62		*100*	0.17	***28***		-0.78		*100*	*3*	-0.34		*67*	--			-1.18		*67*
INDO	-3.8	0.2			*64*	-1.20		*98*	-0.84		*28*	-1.03		*98*	*0*									
OCEAN	-11.9	-0.2	-15.1	9.6	*3092*	-0.72		*96*	-0.07	***22***	*25*	-0.67		*97*	*27*	-1.23	***4***	*89*	-1.00	***7***	*59*	-1.46		*89*

14520, 17: fairly agree ! agree fairly well

We have made the change.

14520, 20: trend ! a trend

We have made the change.

14521, 7: "INCA-OH has higher than TransCom OH concentrations in the NH during summer OH maximum, but lower than TransCOm OH concentrations in the SH Tropic" ! INCA has higher OH concentrations than Transcom in the NH during summer, but lower OH concentrations in the SH Tropics"

We have made the change.

14521, 19: A trend of roughly half a percent per year should have an influence on the CO trend (which are typically 0-2.5 %/year). The sink term read -k.CO.OH and trends in CO and OH should be equally important. In figure 8 the posterior trend in the "sink" (k.OH.CO) is also steeper than the posterior trend in the "source", which indicates some role of OH trends (but indeed rather small).

We have rephrased this part as "Such small trends are considered of very small effect on the CO trends."

14521, 20: considered of minor effect on the CO trends ! to be of minor importance for the CO trends

We have made the change.

14522, 5: Please check all units in the paper. For instance, emission maps now have the label "Tg/year", which misses a unit area. In figure 8, the unit should be Tg CO/months, and the trend should also have a unit.

All units for the emission maps are noted as Tg/year, which indeed will be clarified as Tg/year/grid (2.5 degree×3.75 degree). This is the case for both Fig. 2 and Fig. 9. In Fig. 8, the unit should be specified as Tg CO/year (instead of Tg CO), because we have calculated the yearly sum.

14522, 7: SD? You have only two realizations.

The figure has been updated showing the posterior with both OH fields.

14523, 1: more negative ! steeper negative

We have made the change.

14523, 9: no significant trend in the OH concentrations IS found by the inversion: : :.but: when the burden of CO decreases, one would expect OH to go up, because one of the most important sinks goes down. In that respect, the absence of an OH-trend is surprising, and I think your results point to a small positive OH trend.

A small positive trend is noticed for the SH (+0.2% yr^{-1} with TransCom-OH and +0.7% yr^{-1} with INCA-OH) and a small negative trend in the SH mid-high latitudes (-0.4% yr-1 in TransCom-OH and -0.3% yr-1 in INCA-OH) (14521, 16–20). However, they are really small. We changed the text slightly to be more conservative.

14523, 14: I think that this argument does not make sense. MCF lives 5 years, so a trend in OH anywhere on the globe would be reflected in the MCF mixing ratios also on remote stations.

The signal could be really small in the remote stations if it is not a change through large scales. Meanwhile, different regions could have contrasting changes, but we optimized over big regions. Moreover, the concentration of MCF has been decreasing and is progressively less informative.

14523, 21: This is also incorrect. The Montzka (2011) study only addresses variability, and not trends, since all data were de-trended.

Thanks for pointing this out. We misunderstood the "interannual variability (IAV)" stated in this paper, taking it as normalized values (not de-trended). This was corrected in the revised paper.

14523, 26: unclear why the positive dots appear over oceans. Legend does not explain this.

The reason is that the values over ocean are really small, while the trends are siginificant. The information will be added in the figure caption.

14523, 28: "estimated by the prior"?? Do you mean: "in the prior emissions"?

Yes. It has been changed.

14524, 28: changing rate ! growth rate?

We have made the change.

14526, 11: "Such decreasing: : :observations". This is not a conclusion of this paper.

We have deleted this point from the conclusion.

14527, 15: OH: like above, invalid argument.

We have modified this argument as "Assimilating observations of CH_2O and other chemically related species could inform more about regional CO budgets, in particular the chemical sources and sinks, and therefore could further improve the top-down estimation of CO budgets for each region."

Figure 2: Units Tg per year per unit area (gridbox?)

Yes, it is now specified as TgCO/grid.