

Interactive comment on “On the use of satellite derived CH₄ / CO₂ columns in CH₄ flux inversions” by S. Pandey et al.

S. Pandey et al.

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We thank the referee for his/her useful comments. We have included the referee's comments and comment specific replies (AC) in [blue](#) below. The corresponding changes made in the manuscript are written in *italics*.

1 Summary of review:

This paper presents an implementation of a method for assimilating the ratio between satellite observed total column methane and carbon dioxide, which is in some ways

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more robust than the standard proxy method, which is plagued by the uncertainty of the model-derived XCO₂, while maintaining the larger number of measurements associated than are left with a full physics retrieval. Overall the paper is well written and the results are well-presented, and the method seems to hold some promise. It would be a more interesting study had they chosen to assess real measurements in this study rather than simply testing the mathematical framework using pseudo-data, especially as the approach is not entirely new (see Fraser et al., 2014, who did use real measurements), and the fact that there is now a long record of GOSAT measurements available. I'm sure this was a decision guided by publication strategy rather than scientific merit. but it does detract from the potential impact of the study. The experimental design seems to overstate the capabilities of the satellite measurements due to a variety of choices (not perturbing the pseudo-measurements, using "true" fluxes derived using the same transport model, and possibly using a truth derived from satellite measurements, although this last point is not clear). These need to be addressed and potentially rectified. Despite these misgivings, the study is appropriate for publication in ACP once the following points have been addressed.

2 Substantive points:

As mentioned by a previous reviewer, it seems that overall the newness of the method is overstated, given that Fraser et al. have very recently published a similar approach in the same journal. Given the similarities, the relative newness of the present study should be better framed in context to this already published work, and, if possible, the approaches and results should be compared. Of course this would be easier if this study had used actual measurements in addition to testing the concept with pseudo-measurements.

[AC: Paul Palmer has also raised this issue in his review. We have included a more](#)

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elaborate comparison between our method and the Fraser et al. (2014) approach in the revised manuscript. Please refer to our replies to his first specific comments.

The performance of the inversion under these conditions is almost certainly overly optimistic. Adding a purely Gaussian noise to the "true" fluxes which were derived by the same model is almost too easy a problem: The truth is clearly statistically compatible with the prior assumptions, and the difference is very well-behaved, with no systematic differences.

AC: We define our pseudo truth fluxes on a 4 x 6 (latitude x longitude) grid and then add a Gaussian noise, which is correlated in space and time with parameters defined in Table 1 of the manuscript. This method allows a significant deviation in the CO₂ prior fluxes from the truth in regions with large uncertainties. Using a purely Gaussian case has the advantage that we know how the inversion is supposed to behave, which helped to verify the implementation of the method. Also, we don't make ourselves dependent on a particular choice of biased priors or measurements, as there is no choice that could be considered general. To address the valid point raised by the reviewer, we added the following statement to the paper:

"The performance of inversions assimilating satellite data in this study is optimistic compared to inversions using real observations as we have not introduced any systematic biases in our measurements."

I'm not entirely convinced by the argument that the pseudo-measurements do not need to be perturbed. Yes, if this perturbation is entirely Gaussian then many realizations would result in a convergence to the true result, but isn't the experiment meant to show what information can be gleaned from the measurements in only one year (i.e. not for many repeated years with identical fluxes but varying random measurement noise)? This does not seem valid, and also overstates the information content of the satellite measurements over those of the surface network, the latter having comparatively few measurements, but notably better measurement precision (and accuracy). Or have I misunderstood the purpose of the experiment? Either this explanation needs to be

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fully justified, or the experiment needs to be repeated with properly perturbed pseudo-measurements.

AC: We weigh satellite and surface measurements according to their uncertainty, and therefore the difference in precision does influence their performance. Again, if you would repeat the experiment many times you would arrive at the unperturbed result. The experiments are not meant to demonstrate the performance of satellite inversions, but to compare the performance of the proxy and ratio methods.

Another question related to the "truth" scenarios: the references of Basu et al., 2013 and Houweling et al., 2014 are given, but the specific inversion from each of these studies is not given. I assume that you are using the GOSAT+flask inversion from Basu et al. and one of the SCIAMACHY+flask inversions from Houweling et al., but I can't really tell. This is relevant, as the Basu study in particular (as well as several recent studies, including a just-published GOSAT inversion intercomparison in JGR by Houweling et al.) point to the fundamental inconsistency of the CO₂ fluxes derived from GOSAT and those derived from surface-based measurements. Given this knowledge, if the "truth" is a perturbed version of what is seen by GOSAT, it's hardly surprising that the satellite measurements are better able to reproduce the fluxes than are the surface measurements. This should be further discussed, laying out explicitly which inversions were the basis for the "truth" scenario. Furthermore, the choice of "true" fluxes derived from the same transport model will likely minimize the true problem of transport errors.

AC: The pseudo true fluxes used in our study are not the posterior results of TM5-4DVAR inversions done by Basu et al, 2013 and Houweling et al, 2014. We use the prior fluxes that were also used in those studies (taken from CarbonTraker, EDGAR, GFED, etc.) as our pseudo true fluxes. Transport model uncertainties affect the performance of both inversion methods, whereas our experiments are meant to isolate their differences. We acknowledge that some of the choices we made (e.g. use the same model to generate pseudo data) ignore transport biases, but like to repeat that the main aim of this study is to evaluate the performance of the ratio method and to

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compare its performance to the proxy method. The use of true GOSAT data and need for bias correcting these will be the subject of a future paper.

Granted, the lack of posterior uncertainty estimates makes it difficult to compare, but assuming that the error bars are of a similar magnitude to those of the PROXY method (which may well be an overestimation, although the PROXY method explicitly does not take into account the uncertainty on the modelled XCO₂), I'm not sure about how much can be read into the differences in Figs. 7 and 8. Isn't it likely that these PROXY and RATIO (and for that matter SURFGHG) perform equally well within uncertainty in most cases?

AC: Statistically the posterior of PROXY, RATIO and SURFGHG fall within the uncertainty range of each other for most regions. This has strong connections to our choice of prior fluxes. The truth is well within uncertainty range of the prior.

"Statistically the posteriors of PROXY, RATIO and SURFGHG fall within the uncertainty range of each other for most regions in figure 7."

In section 3.4 it's argued that the surface network performs significantly more poorly over Temperate North America because of the high model representation error in this region. On what is this based? Why is it higher here than anywhere else? The data records seem to be longer and the sampling better than most regions, and because it's a pseudodata experiment there shouldn't be representation problems related to boundary layer height, or other issues that would affect the surface-based inversion but not the satellite inversion. Please explain.

AC: In reality, Temperate North America is well constrained with surface measurements compared to other regions. The high model representation error in this region is the result of our concentration variability dependent model representation error, which makes sense for a model that has a too coarse resolution to represent the CO₂ variability over continental USA. We have added the following to our revised manuscript:

"In Temperate North America, due to coarse resolution of the model in combination with large emission gradients, large representation errors are assigned to the simu-

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lated measurements. Also, we do not take the full advantage of surface measurement coverage of this region as we use only fully processed NOAA/ESRL flask measurements."

Further to the discussion in 3.4: Is the problem with RATIO in Northern Africa its inability to distinguish the biomass burning fluxes? This was a point in Fraser et al. (2014), it might be good to include in the discussion.

AC: We are not optimizing the biomass burning fluxes in our inversions, so that should not be the reason for poor performance of RATIO in Northern Africa.

It might also be relevant to discuss the sparsity of not only surface but also satellite measurements in the tropical land regions.

AC: The reviewer is right that in applications with true GOSAT data the number of data in the tropics will be lesser than used in our experiment. To clarify this point, we added the following to our manuscript:

"As we do not filter-out measurements taken in cloudy scenes and we use medium gain measurements in our inversion, we are optimistic about the satellite coverage in the tropics compared to real-life inversions. However, it is also true that satellite measurements are an important additional source of information about GHGs concentrations in these regions."

Clarification: p8809, lines 15-19: I think I understand what is meant here with the treatment of the prior, but isn't there still a smoothing error that needs to be taken into account due to the different vertical grids of the model and the prior? (See Rodgers and Connor, JGR, 2003, if this isn't clear.) An equation here might help clarify.

AC: The same prior profile is used for generation of pseudo satellite column data and for converting the model profiles to model columns. There will be an interpolation error, but it will be same for the pseudo measurements and the model that is trying to fit the data. Therefore this error does not play a role in our experiments. However, in an application with real data, interpolation errors would play a role, but the ratio and proxy method would be affected similarly.

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3 Very minor points/typos:

p8803, line 8: about methane -> about the methane
p8803, lines 15-20: rework this, the text is awkward and misleading. CSIRO is not a network, nor is NOAA/ESRL, they're organizations that operate networks.

p8803, line 27: onboard Greenhouse -> onboard the Greenhouse (although it might be better to just say GOSAT, and include the full name in the parentheses if you feel it's necessary).

p8803, line 29: constrains -> constraints

p8805, line 2: RemoteC -> RemoTeC

p8806, line 10: setup -> set up (written together it is only a noun, not a verb)

p8807, line 11: method operator -> method the operator

p8807, line 19: assumned -> assumed

p8811, line 3: form -> from

p8811, line 4 line 7: land TransCom -> TransCom land

p8811, line 12: regions -> region

p8811 line 15: postrior -> posterior

p8812, line 16: in-comparioson -> in comparison

p8812, line 22: worse -> worst

p8814, line 15: is -> are

p8814, line 17: the Fig. -> Fig.

p8814, line 19: the Sect. -> Sect.

p8814, line 22: satellites -> satellite

p8816, line 24: regions -> region

p8817, line 7: BEr -> Boreal Eurasia (either use short forms throughout, or spell it out fully)

p8817, line 23: constrain -> constraint

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p8818, line 5: ratio -> the ratio

p8819, line 8: side of problem -> side of the problem

p8819, line 19: remove comma

p8820, line 1: factor 2 -> factor of 2

p8820, line 27-28: in the applications -> in applications

Figure 5 caption: fluxes deviation from the true fluxes at land Transcom regions -> flux departures from the true fluxes for the land TransCom regions

Figures 2 and 10: please change the units on the axis labels to "months" instead of C2076 "m" to avoid confusion In general.

When did XCO_2 and XCH_4 become X_{CO_2} and X_{CH_4} ? I feel like the latter is more widely used.

Also, I agree with a previous reviewer that the current title underplays the discussion of the CO₂ fluxes, which play quite a large role in the discussion.

I assume that the figures relate to only the biogenic (i.e. not fossil fuel, and perhaps not fire) fluxes, but it would be good to clarify this.

AC: All minor comments are addressed in the revised version of the manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 8801, 2015.

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Interactive comment on “On the use of satellite derived CH₄ / CO₂ columns in CH₄ flux inversions” by S. Pandey et al.

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Received and published: 10 July 2015

We thank the referee for his/her useful comments. We have included the referee's comments and comment specific replies (AC) in [blue](#) below. The corresponding changes made in the manuscript are written in *italics*.

1 Summary of review:

Authors develop and test surface CH₄ flux inversion scheme designed to ingest the XCH₄/XCO₂ ratio retrieved from satellite observations. Authors mention that similar

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method was applied earlier by Fraser et al 2014 using a different transport model and inversion method, thus the new results extend the analysis to the case of grid-scale inversion. The pseudo-data experiment is used to quantify the theoretical performance of the method. The advantage of the developed technique is its ability to use soft constrain on CO₂ fluxes instead of hard constrain applied in a traditional approach when only XCH₄ retrieved with proxy method is used. According to the conclusions, the advantage of the technique is limited to regions of large uncertainties in CO₂ fluxes and simulated XCO₂. The manuscript is well written, except for several mistypes, the originality and scientific value of the results justify acceptance for publication. Minor revision addressing the comments below is needed.

2 Comments:

8807 line 5. Authors suggest that CONGRAD is different from M1QN3 in assuming the cost function as multidimensional parabola, and thus less applicable to nonlinear problems. There are two considerations that do not go along with this discussion. Firstly, Meirink et al, (2008) point that the origin of CONGRAD is a code applied by Fisher and Courtier, (1995) to the nonlinear problem of weather forecast. Secondly, M1QN3 makes estimate of Hessian which is equivalent to approximating the cost function as multidimensional parabola, thus this can not be mentioned as disadvantage of CONGRAD. The actual reason for M1QN3 to perform better in nonlinear case could be ability to rebuild Hessian approximation several times on the course of descent to minimum.

AC: *We agree with the referee. We have made the following update in our manuscript:*

“Mathematically, it has the fastest convergence rate for linear inversions, but it may perform poorly for non-linear inversions.” “Our inversion setup for the proxy approach is linear. However, for the new ratio method operator H includes Eq. (2), and hence,

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the inversion becomes non-linear making M1QN3 a more suitable optimizer than CONGRAD. M1QN3 is a quasi-Newton algorithm based optimizer (Gilbert and Lemaréchal, 1989), which is commonly used in non-linear inverse modeling (Cressot et al., 2014; Krol et al., 2013; Muller and Stavrou, 2005). It has the ability to rebuild the second derivative of the cost functions several times during its descent to minimum, and therefore, performs better for non-linear inverse problems.”

8810 line 24. Authors use both CONGRAD and M1QN3, for consistent comparison single method could be better. So, why single method M1QN3 is not used for all inversions? Need to check if the results are stable with respect to the method applied.

AC: CONGRAD is generally our first choice optimizer for proxy inversions using real data, as it is the most efficient optimization method for linear inversions problems. This is an important advantage of proxy inversions, and we did not want to take away this advantage from PROXY. However, we have included results from new proxy inversions using M1QN3 and CONGRAD in Appendix A.

“To compare the difference in convergence between M1QN3 and CONGRAD, we performed additional proxy inversions using both optimization methods (see Appendix A)”

“Appendix A: M1QN3 and CONGRAD

We tested the convergence rate of CONGRAD and M1QN3 using PROXY setup described in Section 2.4. For this purpose, we carried out inversions with both optimizers for 30, 60 and 100 iterations and compared these to the standard inversion using 50 iterations. Figure 1 shows the corresponding posterior CH₄ flux departures from PROXY that are also shown in figure 7. We find that both the optimizers converge within 100 iterations. After 60 iterations, CONGRAD already reaches the solution, whereas M1QN3 shows slower convergence. Significant flux differences are found between the optimizers for inversions with 30 and 60 iterations. For CONGRAD, the difference between inversions with 50 and 60 iterations is negligible.”

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3 Typos

8811 line 4. Sounds better to say “Transcom land regions” instead of “land Transcom regions”

8811 line 8, 10 and below. Should variables cor and bias be written in italics to separate them from the rest of the text?

8812 line 15. Written as “for 100 M1QN3”, it looks incomplete, would be more understandable when text is extended as “for 100 iterations of M1QN3”

8807 line 19. assumed -> assumed

8809 line 21. ‘Transport model’ starts with capital T here, could be mistype?

8812 line 16. in-comparioson -> in comparison

AC: All minor corrections are addressed in the revised manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 8801, 2015.

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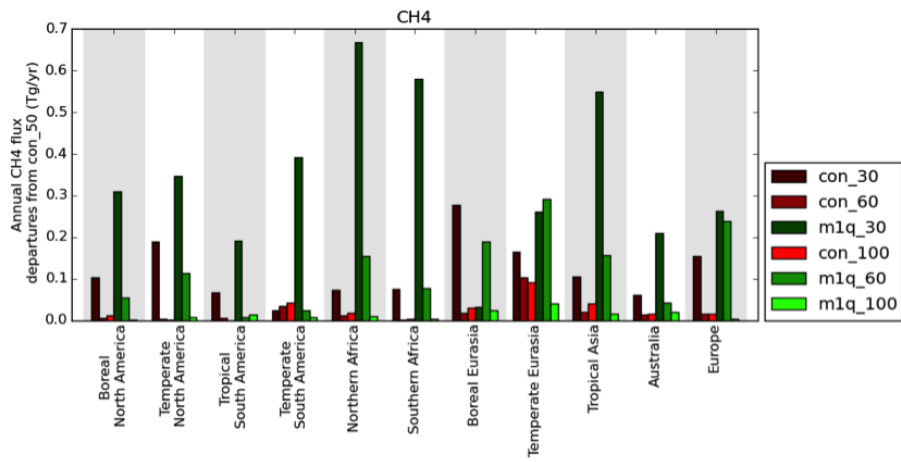


Fig. 1. Annual CH₄ flux departures from PROXY (see figure 7). The first part of a legend's label indicates the optimizer used and the second part indicates number of iterations

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We thank Paul Palmer for his useful comments. We have included the referee's comments and comment specific replies (AC, in [blue](#)) below. The corresponding changes made in the manuscript are written in *italics*.

1 Summary of review:

The authors outline a new method to interpret space-borne atmospheric observations of XCH₄/XCO₂ to infer surface fluxes of CH₄ and CO₂. They concurrently assimilate

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surface observations of these gases to help separate the information embedded in the ratio. The paper is generally good but weak in describing the method in places. Unfortunately, the newness of the method is greatly exaggerated with the technique outlined and demonstrated in a recent paper in this journal. Nevertheless, once this and other comments have been addressed I don't see why it can't be accepted for publication.

2 Specific comments

The authors advertise the newness of the method but this is deceitful. The broad methodology has been reported in Fraser et al, 2014. I'm sure details of the authors' new methodology are indeed new but they cannot claim the method is new. Their one mention of Fraser et al as being noteworthy is disingenuous at best. On a more positive note, it is encouraging that this method works well using a different transport model and inversion method (4D-Var vs MAP for Fraser et al, 2014). At the very least, these authors should discuss the similarities in their method and results with those previously reported by Fraser et al, 2014.

[AC: We agree with the referee that Fraser et al. \(2014\) already used a joint inversion approach. However, we are the first to apply the method to the variational inverse modeling approach. We have clarified this as follows:](#)

“We present a method for assimilating total column CH₄:CO₂ measurements from satellites for inverse modeling of CH₄ and CO₂ fluxes using the variational approach.”

[We have now given appropriate references of Fraser et al. \(2014\) by adding:](#)

“Fraser et al. (2014) developed a method for assimilating Xratio in the MAP inversion setup coupled to the GEOS-Chem global 3-D atmospheric chemistry transport model. Similar to our findings, their OSSEs show that the assimilations of Xratio along with

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surface measurements of CH₄ and CO₂ can reproduce the true fluxes. However, there are some important differences with our study:

1. We focus on a comparison between the proxy and ratio approach and also perform a CO₂ inversion using surface measurements for calculating the model derived CO₂ fields used in the proxy approach. This way the propagation of errors from modeled CO₂ fields into proxy CH₄ measurements is also simulated. Instead, Fraser et al. (2014) add a constant or random bias to the Xratio measurements.

2. Fraser et al. (2014) report posterior uncertainties of CH₄ and CO₂ fluxes derived from their Xratio inversions. Although posterior flux uncertainties can in principle be derived from our method also, they are not reported here for computational reasons.

3. The ratio inversion system is weakly non-linear. The Fraser et al. (2014) ratio inversions assume linearity. We do a non-linear inversion using a suitable optimizer."

Section 2.1: Do the authors assume that R and B are diagonal?

AC: R is assumed diagonal and B is not. The correlation lengths used for calculating B is given in table 1 of the manuscript. We have added the following to clarify:

"We assume no prior correlation between flux categories of CO₂ biosphere, CO₂ oceanic and CH₄ total. The spatiotemporal covariance components for each categories were included in B."

"The diagonal terms of R are the squared sum of measurement uncertainty and model representation error. We assume no correlation between the measurements. Therefore, all the non-diagonal terms of R are set to zero."

Section 2.1: Not reporting a posteriori uncertainties is a major weakness of the method. How do they know that a posteriori fluxes are indeed significantly better than the a priori fluxes? I appreciate that small uncertainties is not a perfect metric but it is useful.

AC: In figure 7, we see that the mean annual posterior fluxes of ratio are closer to the

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truth than the prior. We can assume the posterior uncertainties for RATIO and PROXY are of similar order given the facts that: (1) same amount of information is assimilated in both inversions; (2) we do not introduce any prior correlation between CO₂ and CH₄ fluxes; (3) PROXY has measurement information coming from SURFCO₂. Therefore, it is likely that the posterior fluxes from using the RATIO method have smaller uncertainties than the prior, and hence significantly closer to truth.

Section 2.2: Typo: assumed.

AC: The typo is corrected in the revised manuscript.

Section 2.2: The authors mention nothing about temporal and spatial correlations (see above comment about R and B).

AC: We have added the necessary information about temporal and spatial correlations (See our response to earlier comment about R and B).

Section 2.3: Did the authors sample the RemoTecv1.9 data for cloud-free scenes determined by small AODs and cloud fractions?

AC: We do not sample the RemoTecv1.9 data for cloud-free scenes. We have added the following to clarify:

"We do not sample GOSAT data for cloud free conditions, and therefore assimilate a rather optimistic number of GOSAT measurements. However, satellites such as Sentinel-5 will provide a comparable amount of data".

Section 2.3: Some brief details about the representation error would be useful to report in this paper rather than a simple reference to Basu et al, 2013.

AC: We have added a brief description of the model representation error calculation in the revised manuscript.

"The model representation error is the error made by our finite resolution model in simulating a sample at a specific location. Its size scales with the sub grid concentration

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variability, and is calculated using the local concentration gradients simulated by the model (Basu et al., 2013)”

Section 2.3: I'm not sure I completely follow the logic associated with the decision about not perturbing pseudo observations. It depends if they want to characterize their inversion system ability to infer fluxes.

AC: The aim of our study is: 1. To understand, in a Gaussian framework, the adverse effects of the biases introduced by a model-derived CO₂ field on the posterior CH₄ fluxes of a proxy inversion. 2. To understand whether the ratio inversion method can help us get better knowledge of the CH₄ fluxes in regions where the proxy method doesn't perform well.

As we explained already in the manuscript, our choice of not adding noise to the proxy and ratio measurements does not affect the comparison between the two methods. If we perturb the pseudo measurements with noise according to the data covariance matrix R, we will have to do several inversions with different noise realizations to catch the mean behavior. This multi-inversion mean would correspond to the results of a single inversion without noise. For this reason we do not perturb the data.

Section 2.4: The authors do not clearly explain in the abstract or elsewhere why their RATIO methodology uses the surface data. They do not explain why they are using these data.

AC: We have explained this in section 4 (paragraph 1):

“The method requires assimilation of surface measurements of CH₄ and CO₂ as an additional constraint, since a ratio alone is not sufficient to independently constrain the CH₄ and CO₂ fluxes.”

Section 2.4: There is no mention anywhere that the ratio data have a smaller systematic bias relative to the full-physics products.

AC: Full physics methane retrievals are outside the scope of our experiments. However,
C4663

it is true that Xratio has less bias than the full physics XCO₂ and XCH₄ retrievals, as the scattering-related biases tend to cancel out. We followed the suggestion by the reviewer and added the following line to the revised manuscript.

“Also, Xratio is less biased and has a larger number of measurements than XCH₄ and XCO₂ full-physics retrievals (Fraser et al., 2014)”

Based on the remainder of the paper it is not clear why the paper title, abstract etc is focused on inferring CH₄ fluxes even though the method clearly has a capability to infer CO₂ fluxes (see section 3.4).

AC: We agree with the referee and we changed the title of the paper to:

“On the use of satellite-derived CH₄:CO₂ columns in a joint inversion of CH₄ and CO₂ fluxes”

Discussion: There is a paragraph apologizing for not reporting uncertainties, which is clearly not good enough. Maybe they could compare/contrast the reporting of uncertainties from other methods.

AC: As outlined in the manuscript, this lack of posterior uncertainties in our variational approach is caused by the non-linearity introduced by the ratio method. However, the lack of posterior uncertainties in our synthetic experiment is partly compensated by the fact that we know the true fluxes. Furthermore we make the not unreasonable assumption that the posterior uncertainties of the RATIO and PROXY methods are of similar magnitude. The reason is that they make use of the same observational and a priori constraints. However, we agree with the referee that further discussion is needed. Therefore we have added a paragraph to the discussion section (please refer to our reply of the first specific comment).

Reference: Fraser, A., Palmer, P. I., Feng, L., Bösch, H., Parker, R., Dlugokencky, E. J., Krummel, P. B., and Langenfelds, R. L.: Estimating regional fluxes of CO₂ and CH₄ using space-borne observations of XCH₄: XCO₂, Atmos. Chem. Phys., 14, 12883-

12895, doi:10.5194/acp-14-12883-2014, 2014.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 8801, 2015.

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On the use of satellite-derived CH₄:CO₂ columns in a joint inversion of CH₄ and CO₂ fluxes

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Abstract. We present a ~~new~~ method for assimilating total column CH₄:CO₂ ratio measurements from satellites ~~, which have been retrieved using the proxy ratio approach,~~ for inverse modeling of CH₄ ~~fluxes and CO₂ fluxes using the variational approach.~~ Unlike conventional approaches, in which retrieved CH₄:CO₂ ~~ratios~~ are multiplied by model derived total column CO₂ and only the resulting CH₄ is assimilated, our method assimilates the ratio of CH₄ and CO₂ directly and is therefore called the ratio method. It is a dual tracer inversion, in which surface fluxes of CH₄ and CO₂ are optimized simultaneously. The optimization of CO₂ fluxes turns the hard constraint of prescribing model derived CO₂ fields into a weak constraint on CO₂, which allows us to account for uncertainties in CO₂. The method has been successfully tested in a synthetic inversion setup ~~using the TM5-4DVAR inverse modeling system.~~ We show that the ratio method is able to reproduce assumed true CH₄ and CO₂ fluxes starting from a prior, which is derived by perturbing the true fluxes randomly. We compare the performance of the ratio method with that of the traditional proxy approach and the use of only surface measurements for estimating CH₄ fluxes. Our results confirm that the optimized CH₄ fluxes are sensitive to the treatment of CO₂, and that hard constraints on CO₂ may significantly compromise results that are obtained for CH₄. We ~~see~~ find that the relative performance of ratio and proxy methods have a regional dependence. The ratio method performs better than the proxy method in regions where the CO₂ fluxes are most uncertain. However, both ratio and proxy methods perform better than the surface measurement-only inversion, confirming the potential of space borne measurements for accurately determining fluxes of CH₄ and other GHGs.

20 1 Introduction

In the past century, the concentrations of many potent greenhouse gases (GHGs) have increased in the atmosphere due to anthropogenic activities. The atmospheric dry air mole fraction of the greenhouse gas methane (CH_4), which has a global warming potential of 28–34 on a 100 year time horizon (Myhre et al., 2013), has increased from 700 ppb during the pre-industrial era to
25 ≈ 1800 ppb today (Ferretti et al., 2005). These atmospheric concentrations are unprecedented during at least the last 650 000 years (Spahni et al., 2005). The direct radiative forcing caused by the increase of methane since pre-industrial times is $+0.48 \pm 0.05 \text{ W m}^{-2}$ (Myhre et al., 2013), which amounts to 20 % of the present day cumulative radiative forcing due to all anthropogenic GHGs. Methane also influences atmospheric chemistry and it is an important control on the oxidising ca-
30 pacity of the atmosphere. Further details about [the methane budget](#) can be found in Kirschke et al. (2013). The atmospheric growth rate of methane has varied considerably in the last two decades ([Nisbet et al., 2014](#); [Bousquet et al., 2006](#)) ([Bousquet et al., 2006](#); [Nisbet et al., 2014](#)). Causes of these variations are still not fully understood, which calls for better monitoring of its sources and sinks using both top-down and bottom-up studies.

35 The top-down approach uses inverse modeling techniques to reduce the uncertainty in the bottom-up derived emission estimates on the basis of atmospheric measurements of CH_4 . In the past, several studies applied the top-down method [to assimilating](#) surface-based measurements from global monitoring networks [such as the operated by](#) National Oceanic and Atmospheric Administration–Earth System Research Laboratory (NOAA/ESRL), the Advanced Global Atmospheric Gases Experiment,
40 and Commonwealth Scientific and Industrial Research Organisation ([Houweling et al., 1999](#); [Bousquet et al., 2011, 2006](#); [Hein et al., 2006](#)) ([Bousquet et al., 2011, 2006](#); [Hein et al., 1997](#); [Houweling et al., 1999](#)). However, due to poor spatial coverage of the surface measurement sites, such inversions are effective in constraining the fluxes at sub-continental scales at best (Houweling et al., 1999).

Total column measurements of CO_2 and CH_4 (~~X_{CH_4} and X_{CO_2}~~ [X_{CH₄} and X_{CO₂}](#)) from satellites
45 have proven valuable for inversion studies of CH_4 and CO_2 , especially in regions where surface measurement sites are sparse ([Bergamaschi and Frankenberg, 2009](#); [Basu et al., 2014, 2013](#); [Houweling et al., 2014](#)) ([Basu et al., 2013](#)). For example, atmospheric retrievals from the Thermal and Near infrared Sensor for carbon Observations (TANSO) onboard [the GOSAT \(Greenhouse gases Observing SATellite \(GOSAT, Kuze et al., 2009\)\)](#),
[Kuze et al. \(2009\)](#) have provided valuable [constrains-constraints](#) on the fluxes of CH_4 and CO_2
50 (Basu et al., 2013; Fraser et al., 2013). The CH_4 absorption band at 1.6 micron allows retrieval of its atmospheric concentration with high sensitivity to the planetary boundary layer, where the signals of the sources are strongest. Besides a good sensitivity to the sources, the quality of the inversion-derived CH_4 budget depends strongly on the precision and accuracy of the measurements. It has been shown that systematic errors on regional or seasonal scales of less than 1 % can jeopardize
55 the usefulness of satellite measured CH_4 columns for estimating CH_4 budget (Bergamaschi et al., 2007).

High quality X_{CH_4} and X_{CO_2} retrievals require accurate knowledge of the light-path of the photons that are measured by the satellite. Scattering of light on atmospheric particles (aerosol particles and cloud droplets) may lead to significant light-path perturbations. The accuracy of X_{CH_4} retrievals depends to a large extent on how well the retrieval technique can account for such scattering induced perturbations. A commonly used technique is the so-called proxy method, which was originally developed for retrieving X_{CH_4} and X_{CO_2} using nearby spectral windows from SCIAMACHY (Frankenberg et al., 2005). Since atmospheric scattering affects both compounds in a similar way, light-path errors largely cancel out in the ratio. The retrieval-derived ratio ($\frac{X_{\text{CH}_4}^{\text{obs}} X_{\text{CO}_2}^{\text{obs}}}{X_{\text{CO}_2}^{\text{obs}} X_{\text{CH}_4}^{\text{obs}}}$) is multiplied with a priori knowledge of atmospheric CO₂ derived from a model ($X_{\text{CO}_2}^{\text{model}}$) to generate proxy column measurements of CH₄ ($X_{\text{CH}_4}^{\text{proxy}}$) (Eq. 1).

$$X_{\text{CH}_4}^{\text{proxy}} = \frac{X_{\text{CH}_4}^{\text{obs}} X_{\text{CO}_2}^{\text{obs}}}{X_{\text{CO}_2}^{\text{obs}} X_{\text{CH}_4}^{\text{obs}}} \times X_{\text{CO}_2}^{\text{model}}, \quad (1)$$

$X_{\text{CO}_2}^{\text{model}}$ is usually derived from the results of a CO₂ inversion using the surface measurements, such as CarbonTracker (Peters et al., 2007). It is assumed that: (1) CO₂ exhibits comparatively smaller unknown variations in the atmosphere than CH₄, and (2) residual differences in scattering between the spectral windows of CO₂ (1562 to 1585 nm) and CH₄ (1630 to 1670 nm) used in the retrieval are insignificant. Hence, CO₂ is used as proxy for changes in the light-path. Schepers et al. (2012) discuss the performance of the GOSAT RemoteC proxy retrieval in detail. This retrieval dataset has been used successfully in inversion studies for optimizing CH₄ fluxes (Monteil et al., 2013; Alexe et al., 2014) (Alexe et al., 2014; Monteil et al., 2013). In these studies the error in $\frac{X_{\text{CH}_4}^{\text{model}} X_{\text{CO}_2}^{\text{model}}}{X_{\text{CO}_2}^{\text{model}} X_{\text{CH}_4}^{\text{model}}}$ is assumed to be negligible compared to retrieval error in $\frac{X_{\text{CH}_4}^{\text{obs}} X_{\text{CO}_2}^{\text{obs}}}{X_{\text{CO}_2}^{\text{obs}} X_{\text{CH}_4}^{\text{obs}}}$. However, with the gradually improving quality of the GOSAT retrievals, errors in model-derived CO₂ may become a bottleneck for improving inversion-derived CH₄ fluxes (Schepers et al., 2012).

In some regions, the sparse network of surface measurement sites does not provide sufficient constraints on CO₂ fluxes, leading to possible biases in $\frac{X_{\text{CH}_4}^{\text{model}} X_{\text{CO}_2}^{\text{model}}}{X_{\text{CO}_2}^{\text{model}} X_{\text{CH}_4}^{\text{model}}}$ (Schepers et al., 2012). In this study we investigate a new method, called the ratio method, which circumvents the use of $\frac{X_{\text{CH}_4}^{\text{model}} X_{\text{CO}_2}^{\text{model}}}{X_{\text{CO}_2}^{\text{model}} X_{\text{CH}_4}^{\text{model}}}$ by directly assimilating the retrieved ratio of total column CH₄ and CO₂ into an inversion that optimizes CH₄ and CO₂ fluxes simultaneously. Thus, in the ratio method Eq. (1) is replaced by

$$X_{\text{ratio}} = \frac{X_{\text{CH}_4}^{\text{obs}} X_{\text{CO}_2}^{\text{obs}}}{X_{\text{CO}_2}^{\text{obs}} X_{\text{CH}_4}^{\text{obs}}} \quad (2)$$

Our motivation for implementing the ratio method is to find a representation of CO₂ in the inversion system, that is more consistent with both X_{ratio} and CO₂ surface measurements. It is noteworthy that Fraser et al. (2014) have also assimilated $\frac{X_{\text{CH}_4}^{\text{obs}} X_{\text{CO}_2}^{\text{obs}}}{X_{\text{CO}_2}^{\text{obs}} X_{\text{CH}_4}^{\text{obs}}}$. Also, X_{ratio} is less biased and has a larger number of measurements than X_{CH_4} and X_{CO_2} full-physics retrievals (Fraser et al., 2014).

Fraser et al. (2014) introduced the assimilation of satellite retrieved X_{ratio} in a maximum a posteriori (MAP) inversion system for constraining the surface fluxes of CH_4 and CO_2 . However, the transport model and inversion method used in their study are different from the ones used here.

We perform Observing System Simulation Experiments (OSSEs) to test the performance of the ratio method for reproducing the assumed true fluxes of CH_4 and CO_2 . The results are compared with inversions using proxy retrievals and only surface measurements. In the following Sect. 2, we elaborate on our inverse modeling setup, and describe our OSSE experiments. In Sect. 3, we analyze and compare the inversion-estimated posterior fluxes of CH_4 and CO_2 . In Sect. 4, we further discuss the significance and limitations of our findings and evaluate the future potential of the ratio method for application in inversion studies, leading to our final conclusions.

2 Method

2.1 Inverse modeling

We use the TM5-4DVAR inversion system in this study. It comprises of the Tracer Transport Model version 5 (TM5, Krol et al., 2005) coupled to a variational data assimilation system (4DVAR, Meirink et al., 2008). TM5 simulates the spatio-temporal distribution of a tracer in the atmosphere for a given set of fluxes and initial concentrations that are prescribed as boundary conditions to the model. We have ~~setup~~ set up a dual tracer version of TM5-4DVAR for simultaneous simulation of CH_4 and CO_2 . By combining the output of the two tracers, this model allows us to simulate X_{ratio} (see Eq. 2). The 4DVAR technique uses model calculated and observational dataset of X_{ratio} to optimize a state vector \mathbf{x} , consisting of surface fluxes of CH_4 and CO_2 . The optimum is found by minimizing a Bayesian cost function, defined as

$$J(\mathbf{x}) = \frac{1}{2}(\mathbf{x} - \mathbf{x}_b)^T \mathbf{B}^{-1}(\mathbf{x} - \mathbf{x}_b) + \frac{1}{2}(\mathbf{y} - \mathbf{H}\mathbf{x})^T \mathbf{R}^{-1}(\mathbf{y} - \mathbf{H}\mathbf{x}) \quad (3)$$

where \mathbf{x}_b is the a priori knowledge of \mathbf{x} , and \mathbf{H} is the observation operator, which converts the output of the model, forced by \mathbf{x} , to corresponding mixing ratios at the measurement sites of \mathbf{y} . Hence, $\mathbf{H}\mathbf{x}$ represents the model simulated counterpart of the observation vector \mathbf{y} . \mathbf{R} and \mathbf{B} are error covariance matrices for $\mathbf{y} - \mathbf{H}\mathbf{x}$ and \mathbf{x}_b , respectively. Each iteration of TM5-4DVAR is composed of a forward and an adjoint TM5 run (Errico, 1997). The forward run is used to calculate the value of the cost function for a trial state vector \mathbf{x}_j (using Eq. 3). The adjoint run provides the corresponding cost function gradient ($\nabla J(\mathbf{x})$). At the end of each inversion iteration j , $\nabla J(\mathbf{x}_j)$ and the state vector \mathbf{x}_j are fed into an optimizer module to calculate the state vector for the next iteration (\mathbf{x}_{j+1}). For linear inverse problems we use the conjugate gradient optimizer (CONGRAD, Lanczos, 1950), that has been used extensively for linear inversion problems (Monteil et al., 2011, 2013; Houweling et al., 2014; Basu et al., 2013) (Basu et al., 2013; Houweling et al., 2014; Monteil et al., 2014). Mathematically, it has the fastest convergence rate for linear inversion problems, but it performs

125 ~~may perform~~ poorly for non-linear ~~inversion problems, because it assumes that the shape of the cost~~
~~function is a multi-dimensional parabola.~~inversions

~~For non-linear problems we use M1QN3, a quasi-Newton algorithm based optimizer (Gilbert and Lemaréchal, 1989),~~
~~which is also commonly used in inverse modeling (Cressot et al., 2014; Krol et al., 2013; Muller and Stavrakou, 2005).~~

Our inversion setup for the proxy approach is linear. However, for the new ratio method the operator
130 H includes Eq. (2), and hence, the inversion becomes non-linear making M1QN3 a ~~more suitable~~
optimizer than CONGRAD. M1QN3 is a quasi-Newton algorithm based optimizer (Gilbert and Lemaréchal, 1989),
which is commonly used in non-linear inverse modeling (Cressot et al., 2014; Krol et al., 2013; Muller and Stavrakou, 2005).
It has the ability to rebuild the second derivative of the cost functions several times during its descent
to minimum, and therefore, performs better for non-linear inverse problems.

135 To compare the difference in convergence between M1QN3 and CONGRAD, we performed
additional proxy inversions using both optimization methods (see Appendix A). We find that M1QN3
has a slower convergence rate in comparison to CONGRAD, and therefore the number of iterations
needed to find the inversion solution is generally higher. Another drawback of the M1QN3 algorithm
that is available to us is that, unlike CONGRAD, it provides no information about the posterior flux
140 uncertainties in a ~~straight forward~~ straightforward way.

2.2 Truth and prior

The ~~assumed~~ assumed true CH_4 and CO_2 fluxes for our inversion setup are taken from Houwel-
ing et al. (2014) and Basu et al. (2013), respectively. The generation of pseudo observations \mathbf{y} is
explained in the next section. Concerning the state vector \mathbf{x} , CH_4 fluxes are optimized for a single
145 category representing the net flux from all the contributing processes at the surface, discretized per
model grid box and per month. For CO_2 , we optimize for fluxes from the biosphere and the ocean,
discretized in time and space like methane. We do not optimize emissions from other categories like
biomass burning and fossil fuel usage, as they are assumed to have relatively small uncertainties.
Table 1 shows the parameters used to calculate the error covariance matrix \mathbf{B} for the prior fluxes. We
150 assume no prior correlation between flux categories of CO_2 biosphere, CO_2 oceanic and CH_4 total.
The spatiotemporal covariance components for each category were included in \mathbf{B} . For details about
~~the~~ this implementation of \mathbf{B} in our inversion see Basu et al. (2013). We use one set of prior fluxes
 \mathbf{x}_b for all inversions, which was created by adding Gaussian noise to the true CH_4 and CO_2 fluxes.
The noise is generated using the a priori flux uncertainties accounting for spatial and temporal error
155 correlations, as described in Chevallier et al. (2007). Figure 2 shows the time series of the true and
prior fluxes for four Transcom regions (explained in Fig. 1). As can be seen, the assumptions regard-
ing the a priori flux uncertainties lead to realistic deviations from the truth in terms of seasonality
and net monthly flux.

2.3 Measurements

160 Pseudo surface observations are generated from a forward run of TM5 using the “true” fluxes as boundary conditions, and they are sampled at coordinates and times of samples collected by ~~the NOAA/ESRL~~ cooperative flask-sampling network run by NOAA/ESRL (Dlugokencky et al., 2009) in the period 1 June 2009 to 30 May 2010 at the sites shown in Fig. 1. In total, we use 3934 surface measurements of CH₄ (from 93 sites) and 1184 measurements of CO₂ (from 85 sites). Similarly, 165 synthetic total column measurements are generated at the times and locations of the GOSAT RemoTeC v1.9 proxy satellite retrievals for the same time period (Schepers et al., 2012). We do not sample GOSAT data for cloud free conditions, and therefore assimilate a rather optimistic number of GOSAT measurements. However, satellites such as Sentinel-5 will provide a comparable amount of data. The forward run of TM5 calculates 25 layer vertical model profiles at the retrieval coordinates. These profiles are converted into the corresponding total columns using the retrieval derived averaging kernels (see e.g. Monteil et al., 2013). In total, we use 443 523 GOSAT total column retrievals of both CH₄ and CO₂ (see Fig. 3).

The observational part of the cost function is calculated by weighing the mismatch between the model simulations and measurements ($\mathbf{y} - \mathbf{H}\mathbf{x}$) with the data error covariance matrix \mathbf{R} . The diagonal terms of \mathbf{R} are the squared sum of measurement uncertainty and model representation error. 175 We assume no correlation between the measurements. Therefore, all the non-diagonal terms of \mathbf{R} are set to zero. The model representation error is the error made by our finite resolution model in simulating a sample at a specific location. Its size scales with the ~~sub-grid~~ subgrid concentration variability, and is calculated using the local concentration gradient simulated by the model. Further details about the calculation of the model representation error in our setup can be found in Basu et al. (2013). For the measurement uncertainties we follow the recommendations of the data providers. For the GOSAT retrieved total column ratios, the uncertainty was calculated by error propagation of the instrument’s measurement noise of the CH₄ and CO₂ total columns given in retrieval data set. The uncertainties of proxy CH₄ total columns are also calculated in similar ways. In principle, they should be the ratio 185 uncertainties plus the ~~$\frac{\mathbf{x}_{\text{CO}_2}^{\text{model}}}{\mathbf{x}_{\text{CO}_2}^{\text{model}}}$~~ $\frac{\mathbf{x}_{\text{CO}_2}^{\text{model}}}{\mathbf{x}_{\text{CO}_2}^{\text{model}}}$ uncertainty (see Eqs. 1 and 2). However, the uncertainties from ~~$\frac{\mathbf{x}_{\text{CO}_2}^{\text{model}}}{\mathbf{x}_{\text{CO}_2}^{\text{model}}}$~~ $\frac{\mathbf{x}_{\text{CO}_2}^{\text{model}}}{\mathbf{x}_{\text{CO}_2}^{\text{model}}}$ are neglected in real world applications, and we follow the same procedure. Hence, in our experiment, the ratio and proxy columns have the same relative uncertainties. For computational efficiency, we assume no correlation between the measurements (i.e. all the non-diagonal term of \mathbf{R} are set to zero).

190 Formally, we should perturb the pseudo measurements with noise according to the data covariance matrix \mathbf{R} , following the same procedure as for the a priori fluxes. However, to catch the mean behavior one would have to do several inversions with different noise realizations. This multi-inversion mean would correspond to the results of a single inversion without noise. For this reason we do not perturb the data. It should also be noted that satellite measurements are simulated using the same 195 prior profiles as used for the real RemoTeC GOSAT retrievals. Since the same prior profile is used

in the inversion and in the generation of pseudo data, its contribution cancels out in the model data mismatch and therefore does not influence the results.

In the ratio inversion, the GOSAT measurements are in terms of X_{ratio} , whereas the output of the ~~Transport~~transport model is in terms of ~~$X_{\text{CH}_4}^{\text{obs}}$ and $X_{\text{CO}_2}^{\text{obs}}$~~ $X_{\text{CH}_4}^{\text{obs}}$ and $X_{\text{CO}_2}^{\text{obs}}$. The observation operator H transforms the absolute columns to column ratios using Eq. (2). For calculating the gradient of $J(x)$, the adjoint of H is needed for propagating the sensitivities of the cost function from X_{ratio} to the corresponding sensitivities of ~~$X_{\text{CH}_4}^{\text{obs}}$ and $X_{\text{CO}_2}^{\text{obs}}$~~ $X_{\text{CH}_4}^{\text{obs}}$ and $X_{\text{CO}_2}^{\text{obs}}$. This adjoint is derived by applying the adjoint coding rules described in Errico (1997). It should be noted that the problem is only weakly non-linear since the values of ~~$X_{\text{CO}_2}^{\text{obs}}$~~ $X_{\text{CO}_2}^{\text{obs}}$ vary in the narrow range of $\approx 350\text{--}400$ ppm in our calculations, and the inversion-derived adjustments to ~~$X_{\text{CO}_2}^{\text{obs}}$~~ $X_{\text{CO}_2}^{\text{obs}}$ are only a small fraction of that range.

2.4 Experiment

In this study, we perform OSSEs comparing different global inversion setups using the same truth and a priori fluxes. The inversions system is run at a $6^\circ \times 4^\circ$ horizontal resolution and 25 vertical hybrid sigma-pressure levels from the surface to the top of the atmosphere. Simulations are performed for the period 1 June 2009 to 30 May 2010. The transport in TM5 is driven by meteorological fields from the European Centre for Medium-Range Weather Forecasts (ECMWF) ERA-interim reanalysis project (Dee et al., 2011). Table 2 provides an overview of the inversions that have been performed, specifying the fluxes that were optimized, the optimizer that was used with number of iterations, and the type of measurements assimilated. The PROXY inversion requires ~~$X_{\text{CO}_2}^{\text{model}}$~~ $X_{\text{CO}_2}^{\text{model}}$ (see Eq. 1), which is calculated by sampling the output of a forward run of TM5 using posterior CO_2 fluxes from the SURFCO2 inversion and applying the GOSAT averaging kernel.

The TRU-DAT represents an inversion which assumes that we have perfect knowledge of ~~$X_{\text{CO}_2}^{\text{model}}$~~ $X_{\text{CO}_2}^{\text{model}}$. It is used as a best-case scenario for the proxy method. In contrast, ~~$X_{\text{CO}_2}^{\text{model}}$~~ $X_{\text{CO}_2}^{\text{model}}$ for PRICO2 was calculated using prior CO_2 fluxes transformed directly into observations using TM5 without optimization using CO_2 surface measurements. This inversion represents a worst case scenario for the proxy method. The RATIO inversion uses our new ratio method, assimilating surface CH_4 and CO_2 observations, and X_{ratio} for optimizing surface CH_4 and CO_2 fluxes. PROXY represents the common use of proxy retrievals in atmospheric inverse modelling. In PROXY, the same amount of measurements are assimilated in a series of two linear inversions: (1) optimization of CO_2 fluxes ~~w.r.t. the surface data with surface observations~~ (SURFCO2), (2) an inversion using surface CH_4 and ~~$X_{\text{CH}_4}^{\text{proxy}}$~~ $X_{\text{CH}_4}^{\text{proxy}}$ (PROXY). We use 50 ~~CONGRAD iterations~~ iterations of CONGRAD for both of these inversions. In RATIO, all the information is assimilated in a single inversion using 100 ~~iteration~~ of M1QN3 iterations.

230 2.5 Analysis

In Sect. 3, we analyze the monthly time series of posterior fluxes from different inversions using Taylor plots (Taylor, 2001) and mean annual departures ~~form from~~ the true fluxes aggregated over the ~~land-Transcom-Transcom land~~ regions. We only show the analysis of the fluxes over the land as the fluxes of CH₄ are negligible over the oceans. We define the following parameters to represent the
235 average deviation of the posterior fluxes from the truth over all the ~~land-Transcom-regions:-Transcom~~
~~land regions:~~

$$\begin{aligned}\kappa &= |\overline{cor} - 1|, \\ \gamma &= |\overline{\sigma/\sigma_{\text{truth}}} - 1|, \\ \beta &= |\overline{bias}|,\end{aligned}\tag{4}$$

240 where ~~eor-cor~~ is the cross-correlation between the posterior and true monthly flux ~~timeseries-time~~
~~series~~ for a Transcom ~~regionsregion~~, and $\sigma/\sigma_{\text{truth}}$ is the relative SD of the posterior and true monthly
flux ~~timeseries-time series~~ of a Transcom region. In the Taylor plots, $\sigma/\sigma_{\text{truth}} = 1$ and ~~eor-cor~~ = 1
represent the true fluxes and therefore, we subtract 1 from both the values in Eq. (4) to represent the
deviation of prior or ~~postrior-posterior~~ fluxes from true fluxes. Finally, ~~bias-bias~~ is the difference ~~of~~
245 ~~between~~ the posterior and true net annual flux of a Transcom region. It should be noted that κ and
 γ are dimensionless, and β has a unit of Tg yr⁻¹ for CH₄ and Pg C yr⁻¹ for CO₂. Table 3 lists the
values of these parameters for the inversions performed in this study. The closer these values are to
zero for an inversion, the better it is performing, With each parameter at zero the agreement between
the true and inversion-optimized fluxes is perfect.

250 3 Results

3.1 Ratio method implementation

Figure 3 summarizes the performance of RATIO (see also Table 2). The pseudo X_{ratio} measurements
have typical values in the range of 4.4 to 4.8 ppb ppm⁻¹. We observe that the latitudinal gradient of
CH₄ atmospheric concentration is a dominant mode of variation in X_{ratio} . The randomly generated
255 globally and annually integrated a priori CO₂ flux, combining land and ocean, is 2.01 Pg C yr⁻¹
larger than the true flux (truth = -4.65 Pg C yr⁻¹, prior = -2.640 Pg C yr⁻¹). As a result of this,
the a priori fluxes overestimate the global CO₂ increase. The global annual prior CH₄ flux is only
6.85 Tg yr⁻¹ lower than the truth (truth = 541.764 Tg yr⁻¹, prior = 534.905 Tg yr⁻¹), which is
a much smaller relative deviation from the true fluxes compared to CO₂. Hence, the percentage mis-
260 match between the modeled prior and measured X_{ratio} is mostly positive over the globe (Fig. 3c).
The figure also compares the prior and posterior misfits of RATIO to the “true” X_{ratio} . The measure-
ment uncertainty of X_{ratio} increases towards higher latitudes. We find a gradient norm reduction of

≈ 2000 for 100 iterations of M1QN3. As expected, the posterior mismatches are strongly reduced ~~in-comparioson~~ in comparioson to the prior, demonstrating that the ratio inversion system works
265 mathematically and that it is reasonably efficient in minimizing the cost function. The improved fit of measurements also leads to a convergence of the posterior fluxes towards the true fluxes, as will be discussed in detail in Sects. 3.3 and 3.4.

3.2 TRU-DAT and PRICO2

As explained in Sect. 2.4, TRU-DAT and PRICO2 represent best and ~~worse-worst~~ case scenarios
270 of the impact of errors in $X_{\text{CO}_2}^{\text{model}}$ $X_{\text{CO}_2}^{\text{model}}$ on the results of a proxy inversion. Here we analyze the differences between these inversions, which inform us about the sensitivity of the proxy method to errors in $X_{\text{CO}_2}^{\text{model}}$ $X_{\text{CO}_2}^{\text{model}}$. Figure 4 compares the performance of PRICO2 and TRU-DAT using Taylor plots. In these plots, each point represents a 12 month timeseries of CH_4 fluxes integrated over a ~~land-Transeom~~ Transcom land region. Compared to the prior ($\kappa = 0.286$ and $\gamma = 0.211$),
275 the posterior fluxes of TRU-DAT shows much better agreement with the true fluxes ($\kappa = 0.024$ and $\gamma = 0.042$). PRICO2 ($\kappa = 0.210$ and $\gamma = 0.258$), on the other hand, performs even worse than the prior in terms of γ . Figure 5 shows how well the TRU-DAT and PRICO2 inversions are capable of reproducing the true annual fluxes integrated over ~~land-Transeom~~ Transcom land regions. The β values are 2.370, 2.409 and 0.621 Tg yr^{-1} for PRIOR, PRICO2 and TRU-DAT, respectively. Again,
280 we observe that on average the results of TRU-DAT are closest to the truth, and that the results for PRICO2 are further away from truth than the a priori fluxes. This tells us that the performance of inversions assimilating proxy data is sensitive to our knowledge of the CO_2 fluxes. In practical applications, however, the CO_2 fluxes will first be optimized using surface measurements to obtain a better representation of atmospheric CO_2 concentrations. Inversions representing this approach
285 will be discussed in the next section.

3.3 PROXY, RATIO and SURFCH4

Next we analyze the difference between the proxy inversion (PROXY), using optimized CO_2 concentrations from SURFCO2, and our new ratio method (RATIO). For comparison, we also include results of SURFCH4 using only surface CH_4 measurements. The performance of these inversions
290 is analyzed as in Sect. 4.2, and the results are summarized ~~on-in~~ in Figs. 6 and 7. All three inversions improve the ~~eor-cor~~ of the posterior fluxes with the truth compared to the prior but have varied performance in improving $\sigma/\sigma_{\text{truth}}$. The prior fluxes of Boreal North America are closer to the truth than any of the posterior fluxes. However, it should be realized that the prior fluxes were created by adding random noise to the truth, which happens to be a small perturbation occasionally. This is why we
295 average results over all ~~land-Transeom~~ Transcom land regions to derive meaningful comparisons. The κ and γ values, representing the average ~~perfomence-over-land-Transeom~~ performance over

Transcom land regions, are shown in Table 3. We observe that RATIO and PROXY perform better than SURFCH4, confirming the importance of information provided by the satellite measurements.

Figure 7 shows the departures of the annual fluxes from the truth aggregated over ~~land-Transcom~~ region-Transcom land regions. The β values are 2.37, 1.40, 1.43, and 1.96 Tg yr^{-1} for PRIOR, RATIO, PROXY and SURFCH4, respectively (see Table 3). Overall, we find that the performance of RATIO and PROXY is similar. RATIO performs better than PROXY in 6 regions, and PROXY is better in the other 5 regions. The PROXY inversion shows the worst performance in Boreal North America, Temperate North America and Boreal Eurasia, and RATIO has the worst performance in Southern Africa.

~~Overall, we~~ We find that with the additional information provided by the satellite measurements RATIO and PROXY are able to reproduce the true fluxes better than SURFCH4. However, it is difficult to conclude if RATIO or PROXY performs better, as their ~~realtive~~ relative performances vary across the regions. As can be seen in Figs. 6 and 7, PROXY clearly has a poor performance over Temperate North America. Similarly, RATIO performs worse in Southern Africa than PROXY. These varying relative performances ~~is~~ are further investigated in the next subsection. Annual flux uncertainties of the fluxes are shown as error bars in ~~the~~ Fig. 7. It should be noted that unlike PROXY and SURFCH4, RATIO does not estimate posterior uncertainties. This drawback of RATIO will be further discussed in ~~the~~ Sect. 4. The reduction in uncertainty is larger for PROXY than SURFCH4 in the regions where we have less surface measurements (in Tropical South America, Temperate South America, Northern Africa). This can be attributed to the larger number of ~~satellites~~ satellite observations in comparison to surface measurements in these regions. In other regions, both inversions show similar uncertainty reductions due to a higher gradient norm reduction achieved by SURFCH4 (3.1×10^{10}) compared to PROXY (3.9×10^3). Both inversions are run for 50 iterations, but PROXY has a larger number of data to assimilate than SURFCH4, and therefore, it achieves a lower gradient norm reduction.

3.4 CO₂ fluxes

As explained in Sect. 1, the motivation for our ratio technique is to obtain a more consistent representation of the CO₂ concentration fields in the atmosphere. In this subsection, we address the question whether RATIO optimized CO₂ fluxes are indeed closer to the truth than those obtained using SURFCO2 (which are used for PROXY). Figure 8 shows the deviations of posterior CO₂ fluxes from the truth for RATIO and SURFCO2. In general, annual a priori CO₂ fluxes show large relative deviations from the truth compared to CH₄. This is a direct consequence of the assumed a priori flux uncertainties (see Table 1). The β values (Table 3) are 0.327, 0.185 and 0.134 Pg C yr^{-1} for PRIOR, SURFCO2 and RATIO, respectively. RATIO is able to constrain CO₂ fluxes better than SURFCO2. The difference between SURFCO2 and RATIO is explained by regions such as Temperate North America and Temperate South America, which are relatively poorly constrained by SUR-

FCO2. ~~RATIO is performing better in these regions with the help of satellite measurements. This difference in the performance can also be attributed to the high model representation error associated with the point measurements in Temperate North America and a~~ In Temperate North America, due to coarse resolution of the model in combination with large emission gradients, large representation errors are assigned to the simulated measurements. Also, we do not take the full advantage of surface measurement coverage of this region as we use only fully processed NOAA/ESRL flask measurements. A lack of surface ~~measurement stations~~ measurements can be the reason for poor performance of SURFCO2 in Temperate South America. We observe that RATIO is performing better in these regions with the help of satellite measurements.

Figure 9 shows how well the inversion-derived CO₂ fluxes reproduce the true seasonality. Compared with CH₄, the prior fluxes correlate well with the truth, despite their relatively large a priori uncertainties. This reflects the large seasonal variation in the biospheric CO₂ fluxes. For CO₂, the differences in the Taylor diagrams are dominated by variations in $\sigma/\sigma_{\text{truth}}$. Overall, RATIO ($\kappa = 0.125$, $\gamma = 0.225$) performs better than SURFCO2 ($\kappa = 0.180$, $\gamma = 0.241$). RATIO is able to reproduce the true seasonality for most regions except Northern Africa, Temperate Eurasia and Tropical Asia. In Temperate Eurasia, SURFCO2 performs very well. However, it performs worse than RATIO in Tropical Asia. In Tropical South America and Temperate South America, we find a similar performance of RATIO and SURFCO2. The prior for Europe does not deviate much from the truth, so the relative performance for the two methods cannot be judged adequately.

3.5 The link between CO₂ and CH₄

In principle, the performance of PROXY should improve with the performance of SURFCO2. If SURFCO2 reproduces the true CO₂ fluxes exactly, ~~then~~ the only source of error in $X_{\text{CH}_4}^{\text{proxy}}$ due to $X_{\text{CO}_2}^{\text{model}}$ will be the representation error of the finite resolution model used for generating spatio-temporal fields of CO₂. Also in the case of RATIO, the correctness of posterior CH₄ fluxes is dependent upon the correctness of CO₂ fluxes and vice-versa. For example, Figs. 8 and 9 show that Southern Africa has a poor performance of RATIO, and that SURFCO2 has a poor performance in Temperate North America for constraining CO₂ fluxes. This is also reflected in the poor performance of RATIO and PROXY in constraining CH₄ fluxes in these regions (Sect. 3.3). The performance of SURFCO2 varies regionally, which causes a corresponding pattern in the performance of PROXY. The same relation should hold for the posterior CO₂ and CH₄ fluxes calculated with RATIO. To quantify this relation, we define p_{CH_4} as a measure of the relative accuracy of RATIO and PROXY derived CH₄ fluxes, and p_{CO_2} as a measure of the relative accuracy of RATIO and SURFCO2 derived biosphere CO₂ fluxes for each Transcom region. They are defined as

$$\begin{aligned}
 p_{\text{CH}_4\text{CH}_4} &= \left| x^{\text{PROXY}}_{\text{CH}_4\text{CH}_4} - x^{\text{truth}}_{\text{CH}_4\text{CH}_4} \right| - \left| x^{\text{RATIO}}_{\text{CH}_4\text{CH}_4} - x^{\text{truth}}_{\text{CH}_4\text{CH}_4} \right|, \\
 p_{\text{CO}_2\text{CO}_2} &= \left| x^{\text{SURFCO}_2}_{\text{CO}_2\text{CO}_2} - x^{\text{truth}}_{\text{CO}_2\text{CO}_2} \right| - \left| x^{\text{RATIO}}_{\text{CO}_2\text{CO}_2} - x^{\text{truth}}_{\text{CO}_2\text{CO}_2} \right|, \quad (5)
 \end{aligned}$$

where the x 's denote timeseries of monthly fluxes integrated over land Transcom regions. The subscripts indicate the tracer, and the superscripts indicate whether the fluxes refer to the truth or inversion estimates. ~~p_{CH_4} and p_{CO_2}~~ p_{CH_4} and p_{CO_2} are arrays of 12 month timeseries for each ~~land Transcom regions~~ Transcom land region. They are defined such that: (1) ~~$p_{CH_4,i} > 0$~~ $p_{CH_4,i} > 0$ implies that RATIO is performing better than PROXY for CH₄ fluxes in the month i . (2) ~~$p_{CO_2,i} > 0$~~ $p_{CO_2,i} > 0$ implies that RATIO is performing better ~~then than~~ SURFCO2 for CO₂ fluxes in month i . (3) For values of ~~$p_{CH_4,i}$ and $p_{CO_2,i}$ less than~~ $p_{CH_4,i}$ and $p_{CO_2,i}$ less than 0 the reverse of (1) and (2) is true.

The upper panel of Fig. 10 shows ~~p_{CO_2} and p_{CH_4}~~ p_{CO_2} and p_{CH_4} series for Boreal North America. Lower ~~panels~~ panel of Fig. 10 shows the cross-correlations between ~~p_{CH_4} and p_{CO_2} for each land Transcom~~ p_{CH_4} and p_{CO_2} for each Transcom land region. As it can be seen, this value is above 0.7 (mean = 0.809) for all regions except for Australia (0.202) and ~~BEr~~ Boreal Eurasia (0.539). A lack of surface ~~measurement~~ measurements in these two regions can be the reason for the low correlation, as surface measurement stations are needed for good ~~performence~~ performance of both RATIO and PROXY (Sect. 4). Overall, we conclude that the relative performance of the proxy and ratio methods depends strongly on the relative performance of the surface-only and ratio CO₂ inversions.

385 4 Discussion

We have developed the “ratio” method for TM5-4DVAR inversions system. It is an inversion system for assimilating the ratio of satellite-retrieved total columns of CH₄ and CO₂ along with surface measurements for constraining their surface fluxes. The main advantage of ~~the~~ this method over the traditional proxy method is that it does not impose model-derived CO₂ concentrations as a hard constraint on the CH₄ flux optimization. Instead, our method allows optimization of CO₂ and CH₄ fluxes within a single consistent framework. This way we can benefit from the proxy retrieval, which has proven to be highly efficient in reducing the errors due to light-path modification by atmospheric scattering (Sect. 1), but at the same time, avoid projection of errors in ~~$X_{CO_2}^{model}$~~ $X_{CO_2}^{model}$ on the inverted CH₄ fluxes. The method requires assimilation of surface measurements of CH₄ and CO₂ as an additional ~~constrain, since,~~ constraint, since a ratio alone is not a sufficient ~~constrain~~ constraint for absolute values of CH₄ and CO₂ fluxes. For example, the inversions can reduce the absolute CH₄ and CO₂ modeled columns by the same factor and can still fit their ratio column to give a lower value of the cost function (Eq. 3).

The performance of the ratio method is tested in comparison with the traditional proxy method and surface-only inversions in an OSSE using the TM5-4DVAR atmospheric inversion system. Overall, we observe that the ratio method is capable of reproducing the true CH₄ and CO₂ fluxes better than the surface-only inversion. The performance of the ratio method in comparison to the proxy

method varies among ~~land-Transeom regions~~. Transcom land regions. The performance of inversions assimilating satellite data in this study is optimistic compared to inversions using real observations
405 as we have not introduced any systematic biases in our measurements. Also, as we do not filter out measurements taken in cloudy scenes and we use medium gain measurements in our inversion, we are optimistic about the satellite coverage in the tropics compared to real-life inversions. However, it is also true that satellite measurements are an important additional source of information about GHGs concentrations in these regions.

410 The ratio method is a more complicated inversion to solve than a proxy inversion as it is a non-linear inversion problem, and therefore the widely used CONGRAD optimizer cannot be used. In our setup, we use the M1QN3 optimizer, which is capable of handling the non-linearity. However, to inter-compare inversions using different optimizers requires attention as mathematically their mode of operation is different. For example, CONGRAD ~~constraints solves for~~ the largest spatial and
415 temporal scales in the first few iterations, gradually adjusting finer scales in subsequent iterations. M1QN3 works in similar manner, however, it has a much slower convergence rate for the finer scales than CONGRAD. Hence the overall convergence rate of M1QN3 is slower than CONGRAD, and to achieve the same gradient norm reduction it takes more iterations (Krol et al., 2013).

Another drawback of M1QN3 compared to CONGRAD is that no information is obtained about
420 posterior flux uncertainties, ~~since the method does not collect information about Hessian of the cost function like CONGRAD. Posterior uncertainties~~. They are essential for inverse modeling applications using real data to quantify the constraints on the fluxes imposed by measurements. This is true, despite the fact that several important sources of uncertainty, such as transport model uncertainties, are difficult to account for. Furthermore, the accuracy of CONGRAD's uncertainty approximation
425 may be rather poor for large optimization problems, limiting its use. An alternative method for calculating posterior uncertainties is to use a Monte Carlo approach (Chevallier et al., 2007). This method can be applied also to inversions using M1QN3, although the method is computationally expensive. So far we have not investigated possible alternatives for M1QN3. However, we would like to stress that there is a scope to find a more efficient optimizer for solving this non-linear optimization prob-
430 lem, and future studies into the application of the ratio method should put an effort in this direction.

Fraser et al. (2014) developed a method for assimilating X_{ratio} in the MAP inversion setup coupled to the GEOS-Chem global 3-D atmospheric chemistry transport model. Similar to our findings, their OSSEs show that the assimilation of X_{ratio} along with surface measurements of CH₄ and CO₂ can reproduce the true fluxes. However, there are some important differences with our study:

- 435 1. We focus on a comparison between the proxy and ratio approach and also perform a CO₂ inversion using surface measurements for calculating the model derived CO₂ fields used in the proxy approach. This way the propagation of errors from modeled CO₂ fields into proxy CH₄ measurements is also simulated. Instead, Fraser et al. (2014) add a constant or random bias to the X_{ratio} measurements.

- 440 2. Fraser et al. (2014) report posterior uncertainties of CH₄ and CO₂ fluxes derived from their X_{ratio} inversions. Although posterior flux uncertainties can in principle be derived from our method also, they are not reported here for computational reasons.
3. The ratio inversion system is weakly non-linear. The Fraser et al. (2014) ratio inversions assume linearity. We do a non-linear inversion using a suitable optimizer.

445 Now that we have demonstrated that the ratio method works in a synthetic environment, the next step is the application of the method to real satellite data. A first step in this direction is to validate GOSAT observed ~~X_{CH₄} over X_{CO₂} ratios~~ X_{CH₄}: X_{CO₂} with TCCON. After that we plan to apply the ratio method to real satellite data, and compare the outcome with inversions using the GOSAT proxy and full-physics retrieval products. With improved constraints on the CO₂ side of the problem, 450 as more space borne CO₂ measurements becoming available from GOSAT and OCO-2, the proxy method is expected to perform better for methane. In this case one would expect the results of the proxy and ratio methods to converge. Whether or not this will really happen depends on the mutual consistency of the various data streams. The ratio method provides an internally consistent setup (i.e. within a single inversion system) to test this and to identify remaining biases. It should be noted that 455 computationally, the ratio method has the advantage that it optimizes CH₄ and CO₂ fluxes together. This method can also be applied to other pairs of tracers, which are retrieved from close-by spectral ranges in the satellite measurement spectra. For example, CO total columns will be retrieved from TROPOMI (to be launched in 2016) using CH₄ as the proxy for atmospheric scattering, and there is a possibility ~~;~~ that our ratio method can be applied successfully to this pair of tracers.

460 5 Conclusions

We developed a new inverse modeling method within the TM5-4DVAR inverse modeling framework for direct assimilation of satellite observed ratios of total column CH₄ and CO₂. The dual tracer inversion solves for surface fluxes of CH₄ and CO₂. Our current implementation also assimilates surface measurements of CO₂ and CH₄ to further constrain the two tracer inverse problem. To 465 deal with the weak non-linearity introduced by the optimization of tracer ratios we make use of the M1QN3 optimizer, instead of the CONGRAD optimizer, which was used so far for inversions using proxy retrievals. Although the optimization of the ratio inversion using M1QN3 is about a factor of 2 less efficient than the corresponding proxy inversion using CONGRAD, we nevertheless find satisfactory gradient norm reductions (by a factor of ≈ 2000 in 100 iterations). We tested our method 470 in an OSSE setup. We observe good convergence of posterior model columns toward the true ratio columns, and the ratio method is able to reproduce the true CH₄ and CO₂ fluxes from randomly perturbed prior fluxes.

We performed additional inversions in our OSSE setup to compare the performance of inversions using proxy and ratio retrievals from GOSAT. In addition, we compare the performances of these

475 inversions, which also use surface measurements, with inversions that only use surface measurements. Additional inversions are performed to test the sensitivity of proxy inversions to the quality of the model derived CO₂ concentrations, which are used to translate the retrieved tracer ratios into total columns of CH₄. The performance of these inversions is evaluated by comparing the inversion-derived fluxes to a set of true fluxes from which the synthetic measurements were derived. The performance is assessed for monthly and annual fluxes integrated over the 11 ~~land-Transect~~ Transcom land regions. Our results demonstrate that the estimation of CH₄ fluxes using the proxy inversion is sensitive to errors in the modeled derived CO₂ concentrations.

We conclude that for most Transcom regions the ratio method is capable of reproducing the true seasonality and annually integrated CH₄ fluxes. However, it should be noted that availability of surface measurements is important for good performance of the ratio method. The relative performance of the proxy and ratio methods shows a relationship with errors in CO₂, with ratio method performing better in regions where the CO₂ fluxes are poorly constrained. In our synthetic simulations, the ratio inversion is capable of improving the CO₂ fluxes compared with the use of CO₂ surface-only measurements, which explains why it outperforms the proxy method in certain regions. This points to the applicability of the ratio method for improving CO₂ fluxes in these regions. Further research is needed to test the performance of the ratio method in ~~the~~ applications using real satellite data.

Appendix A: M1QN3 and CONGRAD

We tested the convergence rate of CONGRAD and M1QN3 using the setup of PROXY described in section 2.4. For this purpose, we carried out inversions with both optimizers for 30, 60 and 100 iterations and compared these to the standard inversion using 50 iterations. Figure 1.11 shows the corresponding posterior CH₄ flux departures from PROXY that are also shown in figure 7. We find that both the optimizers converge within 100 iterations. After 60 iterations, CONGRAD already reaches the solution, whereas M1QN3 shows slower convergence. Significant flux differences are found between the optimizers for inversions with 30 and 60 iterations. For CONGRAD, the difference between inversions with 50 and 60 iterations is negligible.

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Table 1. Covariance parameters of the a priori flux uncertainties per grid box per month used in the inversions. The uncertainty is expressed as a fraction of the a priori flux. Error correlations are defined by exponential (“e”) and Gaussian (“g”) correlation functions using the specified length scales (Basu et al., 2013).

Tracer category	Uncertainty (%)	Temporal (months)	Spatial (km)
CH ₄ Total	50	3.0-e	500.0-g
CO ₂ Biosphere	250	3.0-e	1000.0-g
CO ₂ Ocean	250	6.0-e	1000.0-g

Table 2. Summary of the inversions performed in this study.

Inversion	Measurements	Fluxes optimized	Optimizer (No of iterations)
RATIO	X_{ratio} , surface CH ₄ , CO ₂	CH ₄ , CO ₂	M1QN3 (100)
SURFCO2	surface CO ₂	CO ₂	CONGRAD (50)
PROXY	$X_{\text{CH}_4}^{\text{proxy}}$, $X_{\text{CH}_4}^{\text{proxy}}$, surface CH ₄	CH ₄	CONGRAD (50)
SURFCH4	surface CH ₄	CH ₄	CONGRAD (50)
TRU-DAT	$X_{\text{CH}_4}^{\text{proxy}}$, $X_{\text{CH}_4}^{\text{proxy}}$, surface CH ₄	CH ₄	CONGRAD (50)
PRICO2	$X_{\text{CH}_4}^{\text{proxy}}$, $X_{\text{CH}_4}^{\text{proxy}}$, surface CH ₄	CH ₄	CONGRAD (50)

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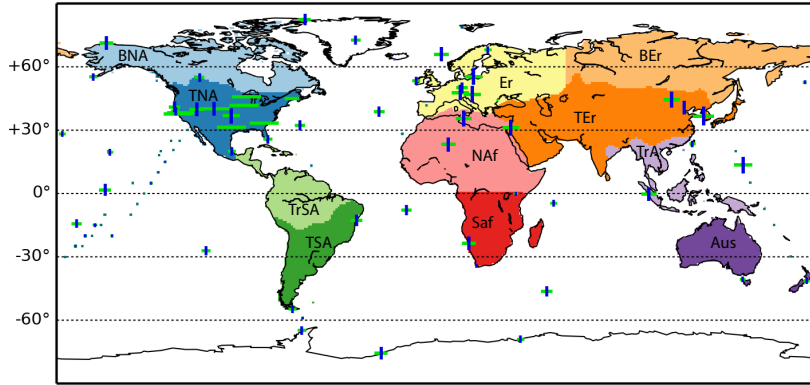


Figure 1. The dynamic symbols (blue-green crosses) show the location of the NOAA measurements sites included in inversions using surface measurements (see Table 2). The lengths of vertical blue and horizontal green bars are proportional to the number of CO₂ and CH₄ measurements, respectively. Continents are divided into 11 Transcom land regions (Gurney et al., 2002) which will be referred to in Sects. 4 and 3 as: Boreal North America (BNA), Temperate North America (TNA), Tropical South America (TrSA), Temperate South America (TSA), Northern Africa (NAf), Southern Africa (SAf), Boreal Eurasia (BEr), Temperate Eurasia (TEr), Tropical Asia (TrAs), Australia (Aus), and Europe (Eur).

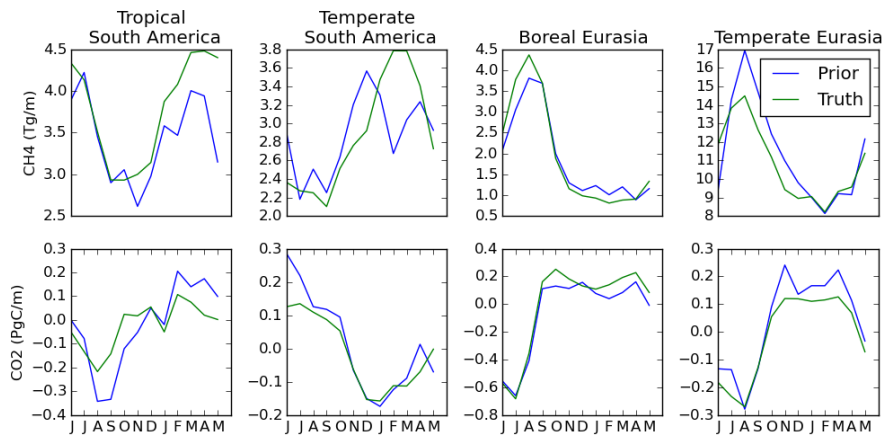


Figure 2. Timeseries of the true (green) and prior (blue) fluxes integrated over Tropical South America, Temperate South America, Boreal Eurasia and Temperate Eurasia. For CH₄, we show the total fluxes, and for CO₂, we show the biosphere fluxes. (see Table 1)

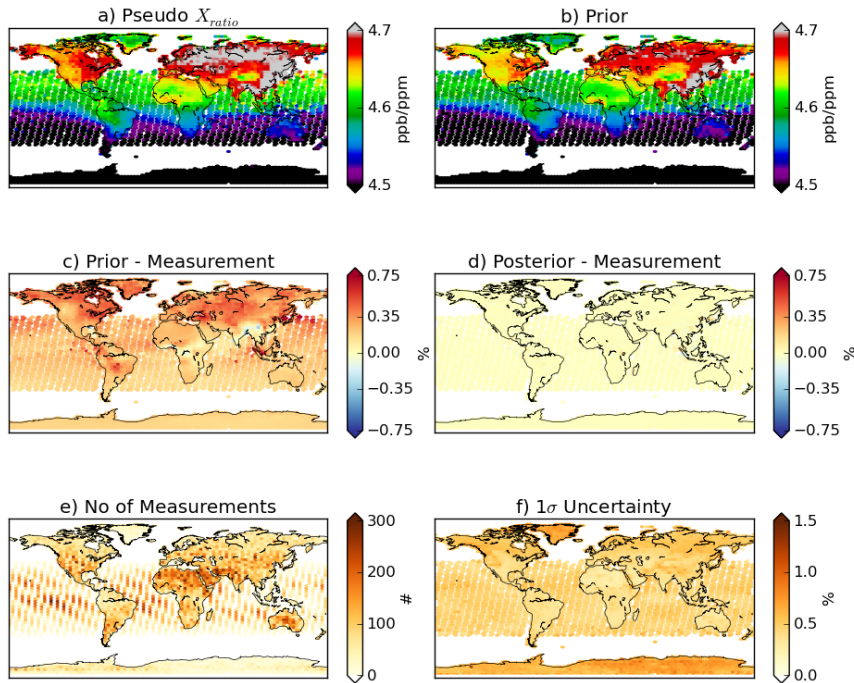


Figure 3. Fit of the RATIO inversion to the annually averaged “true” X_{ratio} pseudo measurements. (a) True pseudo X_{ratio} measurement, (b) a priori modeled X_{ratio} , (c) mismatch between the a priori model and the pseudo data, (d) the corresponding mismatch of the posterior model, (e) the number of GOSAT measurements, (f) the 1σ data uncertainty of X_{ratio} . The values represent yearly averages per $6^\circ \times 4^\circ$ (latitude \times longitude) grid box, except the bottom left panel which shows yearly integrals on $6^\circ \times 4^\circ$ (latitude \times longitude).

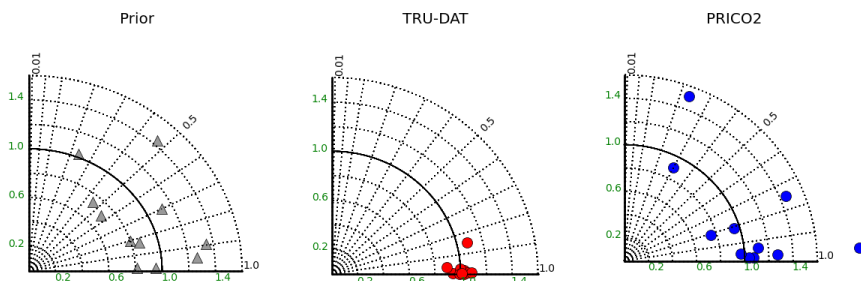


Figure 4. Taylor plots (Taylor, 2001) of monthly prior (grey triangles) and posterior CH_4 fluxes integrated over 11 ~~land~~-Transcom ~~land~~ regions for the inversions TRU-DAT (red circles) and PRICO2 (blue circles). In these plots, each dot represents a seasonal ~~eyele-variation~~ of a single Transcom region. The true fluxes are at the intersection point of the x axis and the bold arc (representing a $-cor = 1$ and $\sigma/\sigma_{\text{truth}} = 1$).

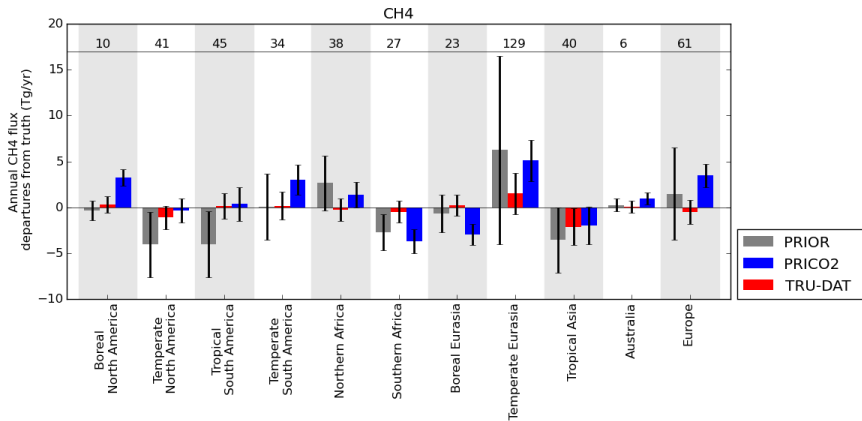


Figure 5. Annual prior and posterior CH₄ fluxes deviation-flux departures from the true fluxes at-land-for the Transcom land regions for the inversions TRU-DAT and PRICO2. The true fluxes are written at the top of the plot in Tg yr⁻¹. The vertical black lines on the bars show 1σ uncertainty of the corresponding values.

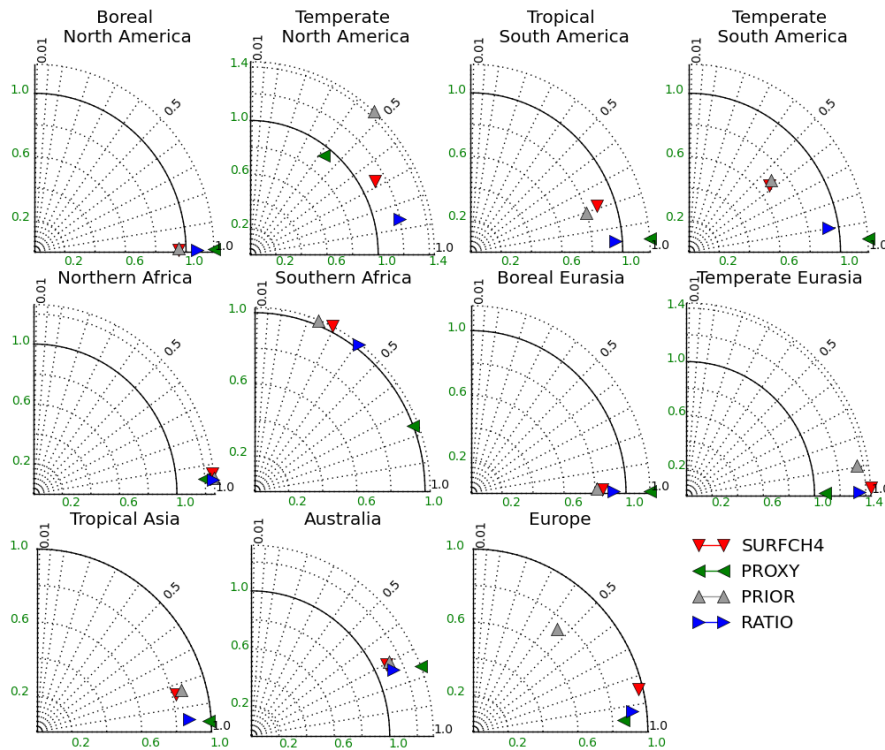


Figure 6. As Fig. 4 for the RATIO, PROXY and SURFCH4 inversions.

Table 3. κ , γ and β values of the inversions performed in this study (see Eq. 4 and Table 2). The β values have a unit of Tg yr^{-1} for CH_4 and Pg C yr^{-1} for CO_2 . κ and γ are unitless quantities.

Tracer	Inversion	κ	γ	β
CH₄				
	PRIOR	0.286	0.211	2.370
	RATIO	0.122	0.129	1.396
	PROXY	0.119	0.137	1.432
	SURFCH4	0.218	0.162	1.959
	TRU-DAT	0.024	0.042	0.621
	PRICO2	0.210	0.258	2.409
CO₂				
	PRIOR	0.232	0.392	0.327
	SURFCO2	0.180	0.241	0.185
	RATIO	0.125	0.225	0.134

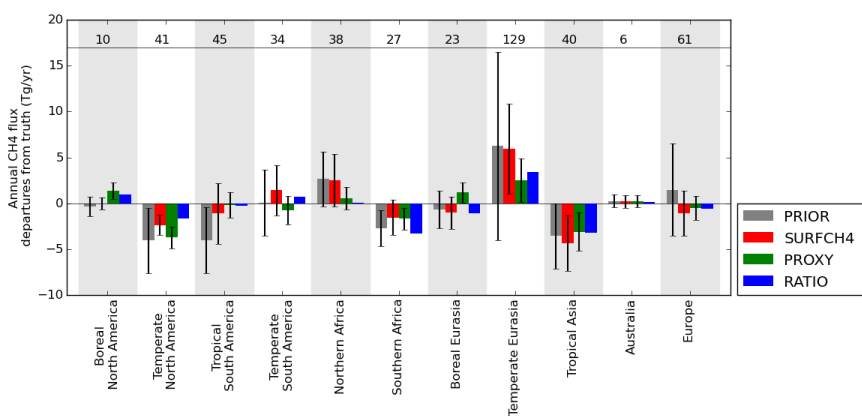


Figure 7. As Fig. 5 for RATIO, PROXY and SURFCH4.

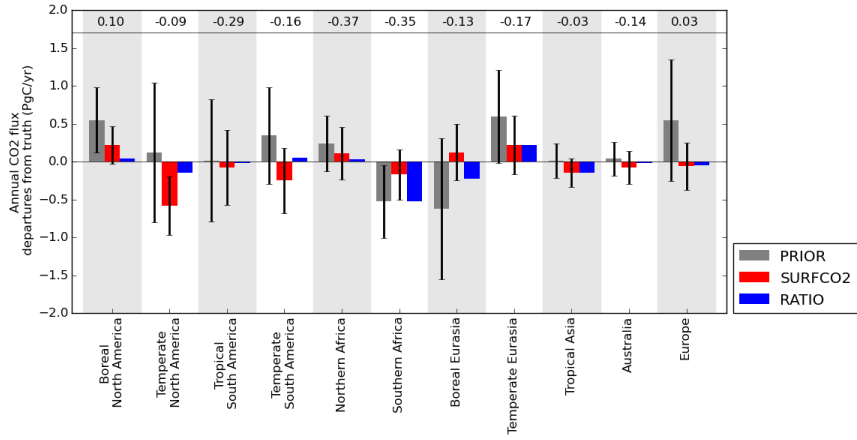


Figure 8. As Fig. 5 for the [biosphere](#) CO₂ fluxes in RATIO and SURFCO2 inversions.

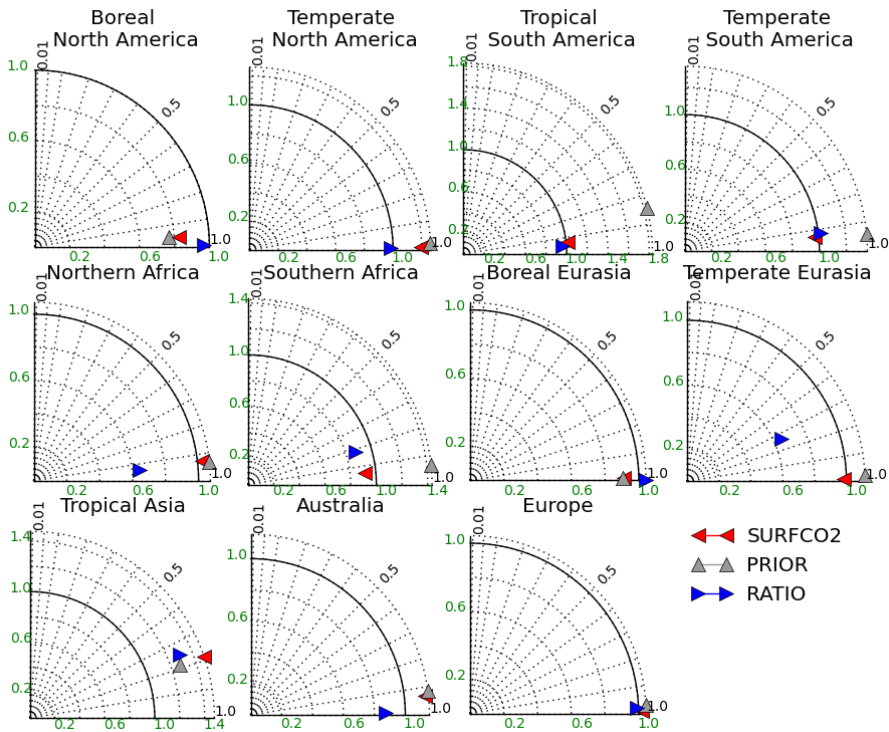


Figure 9. As Fig. 4 for the [biosphere](#) CO₂ fluxes in RATIO and SURFCO2 inversions.

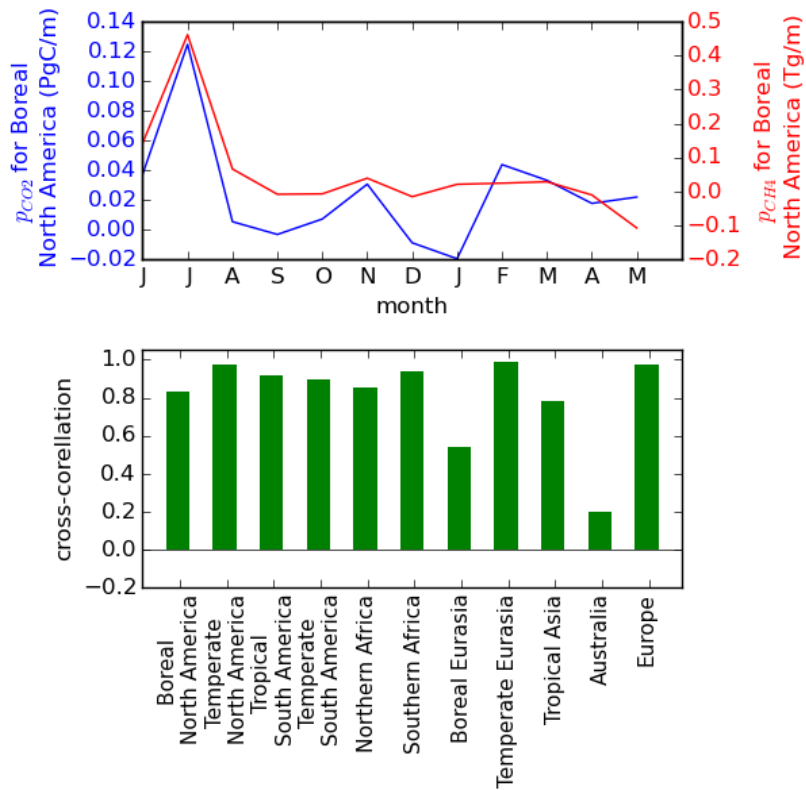


Figure 10. Top: p_{CH_4} - p_{CH_4} and p_{CO_2} - p_{CO_2} timeseries for Boreal North America. Bottom: cross-correlations between p_{CH_4} - p_{CH_4} and p_{CO_2} - p_{CO_2} for land-Transcom land regions (see Eq. 5).

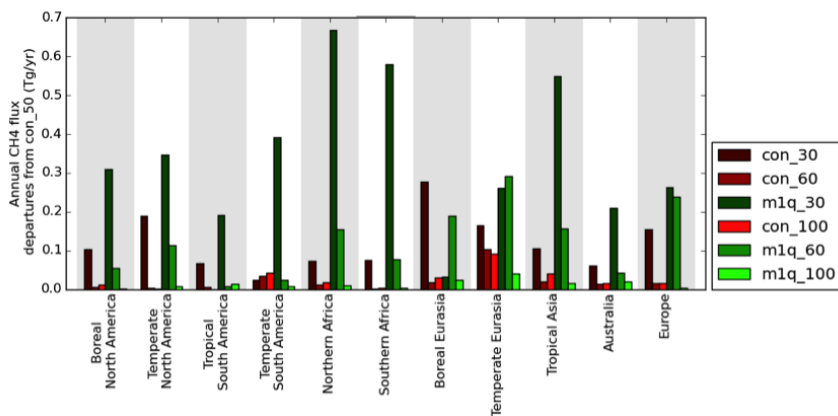


Figure 1.11. Absolute annual CH₄ flux departures of the inversion results from PROXY, which is run for 50 iterations using CONGRAD (see figure 7). The first part of label of each legend indicates the optimizer used for the inversion (m1q: M1QN3; con: CONGRAD), and the second part indicates number of iterations used.