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Interactive comment on "Natural and anthropogenic methane fluxes in Eurasia: a meso-scale quantification by generalized atmospheric inversion" *by* A. Berchet et al.

A. Berchet et al.

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1   Introductory replies

First we would like to sincerely thank the two reviewers for their efforts in reading and understanding our relatively long manuscript. We thank them for their fruitful comments and reviews.

Following their recommendations, in order to highlight the outcomes of our work and make our method more readable, we have modified and enhanced the structure of the manuscript. We rearranged the sub-part order in the method section, added mile-

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stones and refined the general structure. Now, the reader can follow our point from the subsection titles only.

The new manuscript structure is as follows:

- 1. Introduction

- 2. Marginalized inversion framework

  - 2.1. Motivations towards marginalizing
  - 2.2. Method outline
  - 2.3. Output analysis

    * 2.2.1. Network efficiency
    * 2.2.2. Solved spatial and temporal scales
    * 2.2.3. Posterior flux analysis
  - 2.4. Problem size reduction and filters

    * 2.4.1. Observation sampling
    * 2.4.2. Flux aggregation and constraints
    * 2.4.3. Plume filtering
- 3. Set up for an Eurasian domain

  - 3.1. The observation network: $\vec{y}^o$
  - 3.2. Estimates of the network footprints
  - 3.3. Prior fluxes and state vector: $\vec{x}^b$

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- 7. Conclusions

We have also included more physical discussion regarding the improved understanding of the emissions in the region of interest. The new version of the manuscript is attached to this document, with major modifications highlighted in red.

Every single comment from the reviewers is answered here. Comments on lexical or formulation issue are only succinctly answered.

Comments by referee $\#1$ (resp. referee $\#2$) are written in blue (resp. green).

2 General comments

1. I like the attempt at an objective quantification of uncertainty. Although I admit I struggled to understand the discussion of the marginalized Bayesian inversion. Reading the prior publications in this series (Berchet et al. ACP 2013 and Berchet et al. GMDD 2014) certainly helped although I think some effort could be paid to increasing the clarity of the presentation of the technique to those not previously familiar with the enhancements they have developed. Because so many steps are required in this technique, I would suggest incorporating a flow chart into the paper to give the reader something to hang onto and chart progress as they read their way through the convoluted steps required in pre-processing and actual inversion. As they are laid out now it is easy to get lost.

   The marginalized Bayesian inversion has been developed to dampen some possibly important flaws in regional inversion, recognized by the community. The idea behind it is rather simple: it is bound to incorporate the uncertainties in the uncertainties within the inversion computation. As it has been noted by both reviewers, the required steps to implement this basic idea are numerous. We

admit that the submitted manuscript lacked some clear insights into the method to be clearly understood. In the new version of the manuscript, we have included enough details, so the reader does not need to go into the prior literature.

2. One area I am a bit concerned about is the thresholds for 'hot spots' and ill-defined plumes (while understanding that their inclusion can be problematic). They are never defined beyond vague language and the exact thresholds could make their exclusion either relatively inconsequential or result in the loss of important information that could bias the estimates. I never got the sense that there was any attempt to understand how much impact the removal of hotspots and thin plumes has on the overall budget. Is it minor, major? This makes me wonder about how much improvement we gain from the inversion (with its approach that avoids poorly quantifying uncertainties) if we don't also attempt to quantify how important was the information excluded.

Hot spots and ill-defined plumes are critical in the inversion for two main reasons: 1) our representation of the atmospheric transport can generate temporal and spatial mismatches; 2) the time and location of hot spots generating plumes is not always exactly known (especially in Siberia where methane hot spots come from oil or gas leaks and from under-documented wetlands). These mismatches related to plumes causes very strong differences between simulated and observed methane concentrations. These high differences lead to unrealistic corrections in the flux after inversion.

Avoiding such unrealistic increments in the emissions motivates our filter of the plumes. The definition of these plumes can be complicated, as a strong hot-spot can be very well represented by the transport model if observed far enough away of the source. This is why the threshold we choose to define ill-defined plumes is representative of transport errors. All observations with a diagnosed transport

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error (after the maximum likelihood optimization) above a defined threshold (in our case 20 ppb) are excluded from the inversion.

We have clarified this point in the new version of the manuscript.

Besides, the impact of removing plume observations is only in terms of flux constraints. The more plume-like air masses and hot-spots are to be considered by our system, the less information we can deduce on the state of the emissions; and then the higher the posterior uncertainties.

3. Lastly, it appears that very few of the discrete samples are discarded (Figure 2 sites like SDZ, TAP, UUM) as compared to the continuous measurements. Does this demonstrate that the system is placing too much confidence in these samples? Considering how many samples are removed from the more continuous time series this seems strange. I wonder if this is a sampling bias and how will it influence the inversion.

Numerous continuous measurements are indeed excluded from our inversion. This is a critical point as a lot of resources is deployed to maintain the Siberian network. However, this feature can be explained and should not be considered as a sampling bias.

Flask sampling site locations were selected to monitor large-scale gradients and long-term trends. These locations are then mostly far away from the main sources, hence limiting the 'plume' effect. In contrast, the West-Siberian network was design to closely monitor wetland emissions and oil/gas ones.

As a consequence, Siberian sites are very often illuminated by close hot-spots

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and our regional transport model does not always reproduce the plume structure well. Studying the region with all the available in-situ observations would require a transport model with a higher spatial resolution, which we could not afford with our numerical resources.

Nevertheless, the system that we have developed deals with reduced number of observations as filtered out observations only cause the decrease of the network influence and the increase of the overall posterior uncertainties. This was tested in the prior literature with OSSEs.

A few comments have been added to clarify this point in the new version of the manuscript.

4. It is not clear why the minimum measured value per day is used as data filtering method. To exclude night time data, and take PBL height as selection criterion is fine. Something like an afternoon averaged value has been proposed in earlier studies. However, to select a minimum measured value will almost certainly bias the inversion towards too low emissions. A low-resolution model averages high frequency variability, not only the highs, but also the lows. In the model, the grid box where the measurement side is situated will almost certainly have emissions. Because of this, the simulation will not just represent to undisturbed background, but also local emissions.

Indeed, the main objective of our filtering method is to avoid biases toward too low emissions. However, as the mean operator is linear, averaging over the afternoon will conserve outliers representative of the local sources.

On the opposite, we notice that keeping the minimum observed concentration

of the afternoon is somehow equivalent to detecting the time when the PBL is at its maximum, hence when the atmospheric model is the less biased. This reasoning is only valid for methane and trace gases with almost only sources. For example, for CO2 in summer when photosynthesis consumes high amounts of CO2 in the afternoon, the averaging would probably be the best approach.

We slightly develop this point in the new version of the manuscript:
" Here we try to reduce the dimension of the observation space. At the regional scale, considering the spatial resolution of our transport model, only the synoptic variability of the observed signal is relevant. We then decide to keep only one piece of information per site and per day as it is commonly done at the global and continental scales. In addition, simulated vertical mixing close to the surface where observations are carried out is known to be flawed when the planetary boundary layer (PBL) is shallow (typically at night and in winter in Siberia). We then sample the observations during the afternoon when the PBL is higher than 500 m as suggested by prior studies (e.g., **?**) and we pick the observed and simulated mixing ratios at the time when the observations are minimum.
As surface emissions dominate on surface sinks for CH_4, keeping the minimum observed mixing ratio by afternoon is equivalent to detecting the time when the PBL is at its maximum, hence when the atmospheric model is the more accurate. This criterion filters out outliers generated by local influences which cannot be reproduced by an atmospheric transport model with a resolution larger than 25 km, and which only add noise to the system. "

5. Besides the comparison with independent data it would be useful to know how well the inversions reproduce the measurements that are used in the inversion. The risk of the rigorous sampling that is applied is that the measurement coverage becomes very irregular. How realistic are the optimized seasonal cycles at sites with only few data points? This will be hard to judge from the

limited data themselves, but nevertheless this cycle should look plausible, and not too much perturbed at times when data are available. Such comparisons could also serve as an evaluation of the data selection procedure. How poor or how well is the agreement between the model and data that were filtered out?

We agree with the reviewer for this point. Our filtering algorithm makes it difficult to compare prior and posterior concentrations. Nonetheless, digging into this direction for the validation of our model should have been done.

We have included a small discussion regarding this comment in the new manuscript:
" We can also use the data points which have been filtered out by our system in order to evaluate the inversion results. As the number of filtered out observations is high, sampling biases may be expected from the filtering procedures. In addition, as only a few data points are assimilated, unrealistic fluxes could have been inferred by the inversion to fit the assimilated data leading to a flawed reproduction of the remaining observations. As one can see in Tab. **??**, the marginalized inversion significantly improves the simulated mixing ratios at the sites where data is used as expected. As a proof on realistic flux prescription despite the filter on the observations, for all the remaining data, the model results are also well improved for unused data.
This confirms that our method does not create sampling biases despite the high number of filtered data points. It also confirms that the increments on the fluxes are realistic from the point of view of our network. "

6. Among the poorly quantified criteria that I mentioned earlier are the following: What are the criteria is used for,

- Aggregating fluxes.

- Aggregation of the boundary conditions.
- Plumes that can or cannot be resolved by the transport model.
- The rejection of unconstrained regions?
- The rejection of regions that are influence by the boundary condition.
- The statement that the observations constrain fluxes within a radius of 500 km.
- Separation between natural and anthropogenic fluxes.

All this pieces of information are essential as outcomes of our manuscript. They should be more clearly presented. Most of them are somehow included in the submitted manuscript, but mixed with other information.

We have modified the structure of the new manuscript, so the main outcome of our work are better highlighted.

- Aggregating fluxes $\rightarrow$ when $|r_{i,j}| = |\vec{P}^a_{i,j}| > 0.5$. Sect. 2.3.2
- Aggregation of the boundary conditions $\rightarrow$ Sect. 2.3.2
- Plumes that can or cannot be resolved by the transport model $\rightarrow$ $(\mathbf{R})^{i,i}_{max} > 20\,ppb$. Sect. 2.4.3.
- The rejection of unconstrained regions $\rightarrow$ $(\mathbf{K}_{max}\mathbf{H})^a_i < 0.5$. Sect. 2.4.2.
- The rejection of regions that are influence by the boundary condition $\rightarrow$ Sect. 2.3.2.
- The statement that the observations constrain fluxes within a radius of 500 km $\rightarrow$ rough estimate from Fig. 3.

- Separation between natural and anthropogenic fluxes → According to region grouping. Sect. 2.3.2. If only one type of emissions is included within a group, we consider that this type of emission is well separated for the related period and region.

3 Specific comments

1. p. 14591 line 26: total production of?

   The production of natural gas. Clarified in the new version of the manuscript.

2. p. 14591 line 29: absolute concentration? atmospheric mixing ratio?

   The atmospheric composition in terms of mixing ratios and isotopic composition.

3. p. 14592 l 11: Didn't Winderlich also use 3 other towers besides ZOTTO? (Demyanskoe, Igrim, & Karasevoe)

   Winderlich (2012) indeed uses 3 other tower sites. This statement has been corrected in the new manuscript. However, Winderlich mainly focused on the analysis of the ZOTTO signal and from the point of view of CO2.

4. p. 14592 l 29: Add in the acronym CTM here since it is referenced later but never defined (that I could see).

   Acronyms are now defined before being used.

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5. In the manuscript, 'tuple' is used. Is this naming convention coming from programming?

This term is indeed a bad habit from programming. We corrected all the occurrences of this term by 'couple of matrices'

6. p. 14595 l 29: How many Monte Carlo runs were performed?

60000 Monte-Carlo runs are performed to get a good approximation of the posterior uncertainties. This is what makes our work unique in a sense. Most other studies inquiring into the impact of ill-designed uncertainties only rely on a few Monte-Carlo draws.

7. It wasn't clear to me if the marginalized approach still has to assume a Gaussian distribution of uncertainties or not. If so, does the Gaussian assumption hold when you are forced to remove consideration of hotspots of methane?

The Gaussian assumption is indeed one of the flaws in most atmospheric inversions. We also rely on this assumption (this statement is clarified in the new manuscript). This assumption still needs to be generalized in our inversion system, as it has been done by some groups with non-marginalized inversion systems.

8. p. 14606 l. 7: Thin plumes in what respect?

The definition of what we call plume indeed needs to be clarified. This is done in the new version of the manuscript and in the reply to General comment nb. 2.

9. p. 14607 l. 21: Why is it possible to invert the accidental release plume at Elgin

(Berchet et al. 2013 ACP) but not here? The scale?

Actually, the accidental case of Elgin was not really inverted. As it was commented in Berchet et al. (2013), the system we used at thas time did not include the uncertainties on the uncertainty matrices.

The plume was not filtered out at that time because the transport in Europe was pretty well reproduced by our model and because the point source was sufficiently distant from most observation sites (but still influenced observations).

10. p. 14611 l. 26: Indeed, this is where a study like this has great utility – Pointing out where observation sites would be ideally located.

Our method main outcome is unfortunately not significant new insights about methane regional emissions, but about how to better design a monitoring network. This is more emphasized in the new manuscript.

11. p. 14613 l.15: But the hotspots are removed so how is it possible that the inversion is seeing 'punctual leaks and purging releases of gas'?

Hot spots are not always filtered out, has we developed in the reply to General comment nb. 2. It happens that plumes can be assimilated in the inversion systems. In particular, in summer when the PBL is well mixed, CTMs do better in simulating trnasport. That is why some conditions can favour the detection of plumes.

12. p. 14614 l2: 'a not negligible' - confusing. Try 'some' or 'a small' or 'a significant' whichever best fits.

We corrected this in the new manuscript.

13. Fig 3: To make it easier to read, make the outlines of the stars white. Also a scale is needed for the size of the markers. As it is now they vary in size but the reader has no information about what a big one means vs. a small. Also add labels to the colour bars so we know which is which.

This will indeed greatly increase the readibility of the figure. We recomposed a new figure.

14. Fig 7: Please add in bigger numbers and some sort of colourbar label.

Added in the new manuscript.

15. P14590, line 3: Methane influences OH also directly (thus not only via O3)

We modified our statement to avoid such ambiguity.

16. P14591, line 26: What is meant by the 'absolute composition in CH4'?

This statement was misleading. We meant the atmospheric composition of methane (including mixing ratios and isotopic composition)

17. P14592, line 7: What is meant by 'relative contributions to the fluxes'?

We mean the relative contribution of a specific type of fluxes (e.g., wetland) to the total regional fluxes. This is now clarified.

18. P14592, line 14: What is meant by the 'data assimilation Bayesian theory'?

There are different ways of assimilating data. We use the Bayesian inference to do so.

19. P14592, line 17: 'the likely under-estimation... some inventories": a reference is needed here

Winderlich (2012) used NitroEurope emission data, which included EDGAR v4.1 anthropogenic fluxes.

20. P14592, line 18: 'However, atmospheric... local constraints surface fluxes': This depends on the kind of site and is not true for example for a station like South Pole.

Indeed, at the very beginning of the use of data assimilation to infer surface fluxes, most atmospheric inversions were computed at the global scale to deduce year-to-year global budgets. We have to precise that we are implicitly referring to regional inversions.

21. P14593 line 1: It is kind of obvious that uncertainties in reproducing transport come from transport errors.

This sentence indeed sounds weird. We slightly modified the introduction to make our point more accurate.

22. P14594 line 8: 'To do so, we look for the pdf of the state of the system with some knowledge about the atmospheric composition and on the state distribution.': What are we supposed to learn from this sentence?

Same as above, we have reformulated the introduction to the method and now mostly explain what is new in our method.

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23. P14599, line 27: How can the method account for sampling bias. I didn't find that information back in section 2.1

Sampling biases can never be totally accounted for. However, the method as we define it reflects missing values directly into increased uncertainties and decreased influence. We have clarified this point.

24. P14601, line 13: A version number and reference is required for GFED.

OK.

25. P14603, line 15: 'state vector' i.o. 'observation operator'.

OK.

26. P14603, line 19: What matters is not the mean residence time of the air, but the amount of methane that is oxidized within the domain. This could easily be a few Tg/yr. If it cannot be assigned to an atmospheric sink, then it will be accounted for as a reduced source. I understand that you don't want to optimize the sink, but I don't think that the sink can be completely ignored.

The OH sink is indeed a critical point in methane atmospheric inversions. However, regional inversions mostly analyze the synoptic signal, which is caused by regional sources. The atmospheric sink is responsible for very large scale gradients. In our configuration, large gradients are mostly attributed to the prescribed boundary conditions. The regional inversion is then more likely to modify boundary conditions in relation to the atmospheric sink, rather than the regional inversions.

This was another reason for filtering out the regions that cannot be separated from the boundary conditions.

27. P14604, line 14: The statement that GOSAT is the only remaining source of data seems incorrect. For example, NIES has also an aircraft program in Siberia.

This is correct. NIES, which is the main provider of observations for our work, also carries out aircraft profiles above the city of Surgut. LSCE, with the help of the Institute of Atmospheric Optics (Tomsk), also has an aircraft long-term monitoring program over Siberia.

However, in the context of regional inversions, using aircraft flask samples would have been a difficult exercise. As the posterior flux uncertainties are high, as much as the uncertainties in CTM vertical transport, comparing observed and simulated concentration vertical distribution would not lead to clear insights. This is why we decided not to use these data sets.

GOSAT was then the only remaining data set covering high latitude with a satisfactory accuracy.

28. P14604, line 27: 'and associated to... priori profiles' What is meant here?

The full data set is described in Parker et al. (2011) and in Cressot et al. (2014). The retrieval algorithm needs CO_2 vertical profiles and prescribed averaging kernels to compute CH_4 total columns. We have reformulated the paragraph to make it clearer.

29. P14609, line 15: 'Then, amongst... from each other". What criterion is used to make this distinction?

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The description of the method is to be clarified, so that all the criteria are properly defined. This is done in the new version of the manuscript.

30. P14610, line 10: 'indicating that... anthropogenic from wetland emissions". What is the criterion for deciding that fluxes can or cannot be separated?

Same as above. All the criteria are clarified now.

31. P14610, line 22: 'to the real fluxes'. I guess you mean to the inversion-estimated fluxes?

We mean the actual flux we are trying to find out. Actually, we first carried out our inversions with out-of-date EDGAR emissions which were significantly underestimating oil and gas industry emissions. In this configuration, the inversion was finding posterior fluxes in the range of those we find now.

We do not show this kind of tests as we extensively tested our method on OSSEs in a previous method paper.

32. P14611, line 10: which co-located emissions? And how do you know that the regional flux estimates are accurate?

Accurate was ill-chosen. Actually, as we tested our method on various OSSEs, we are confident about the posterior uncertainties of our inversion. The main point we clarify in the new version of the manuscript is that our system provides consistent posterior uncertainties, while classical inversions tend to dramatically under-estimate their posterior uncertainties.

33. P14612, line 8: How are 'significantly constrained' fluxes extrapolated.

We should have added a simple formula to clarify this. We consider that, whenever the portion of constrained fluxes is larger than 20% of the total fluxes, the regional budget is constrained overall. When this proportion is below 20% we consider the inversion does not provide valuable information on the regional budget. The 20% can be discussed but taking higher values would prevent doing much extrapolations.

For constrained period, we simply apply the average correction factor for constrained fluxes to unconstrained fluxes.

34. P14613, line 17: 'This could explain... August' I think it is worth checking how well the inversion resolves anthropogenic and natural sources for this month using posterior covariances.

The anthropogenic leaks are one hypothesis. Wildfires are another one. Isotopic measurements could really help for separating emission sources. However, using covariances, we notice that Lowland emissions are correlated with West Russian emissions (where numerous wildfires occur in August 2010), with a noticeable increase in the emissions.

35. P14614, line 3: How is the 27TgCH4 derived?

This number is derived from EDGAR database v4.2 (year 2010) for OECD European countries, and Central European ones. This is now specified.

36. P14615, line 10: 'We observe a mean shift...' But the GOSAT data were corrected for a mean offset with the model. Therefore it depends on which model estimate you take for this correction whether the shift that is discussed here

leads to an improved agreement with GOSAT or not.

This point was not clear in the previous version of the manuscript. GOSAT retrievals are indeed corrected with the model used to compute the total columns. However, this bias is corrected at the global scale. It is possible that GOSAT measurements suffer biases depending on the latitude (Cressot et al. 2014). This is why we do not discuss GOSAT in terms of absolute value, but only compare differences in total columns.

In the end, the average increment produced by the inversion is very low and cannot be interpreted.

37. P14615, line 11: A mean shift cannot be compared with a single column retrieval precision.

This is true. This statement is incorrect. We should take the number of observations into account when computing observational uncertainties on the shift. Actually, the standard deviation on the posterior minus prior shift itself proves that GOSAT is not sufficient for a regional use.

We clarified this statement in the new version of the manuscript.

38. P14617, line 1: But the MERLIN mission doesn't yield measurements at high spatial resolutions (only a narrow line ∼50 km length per aggregated sample).

MERLIN resolution can indeed be seen as pretty low. However, from our point of view, for Siberia, such a resolution, with a good coverage and precision will drastically improve the number of usable observations for the region.

We have slightly modified our statement to make this idea clearer.

39. P14590, line 2: 'climate forcing' (without 's')

OK.

40. P14591, line 26: 'literature' (remove a 't')

OK.

41. P14591, line 26: 'composition <of> CH4'

OK.

42. P14592, line 1: 'variation' (without 's')

OK.

43. P14593, line 13: 'in-/out-coming to/from': What?

The side observation sites are used to constrained the global air masses which enter the domain or get out of it. Doing so, the total mass balance for the whole domain is improved.

We have reformulated the sentence.

44. P14593, line 19: 'The maximum likelihood criterion'

We mis-used some phrases in the manuscript. The maximum likelihood criterion is the basics of our estimation. The maximum likelihood is found with a quasi-Newtonian descent method. This has been corrected throughout the new version of the manuscript.

45. P14600, line 2: 'AT night or WHEN the PBL is thiNner than'

    OK.

46. P14603, line 11, 'hot spotS'

    OK.

47. Figure 3, caption: 'On the left column, ... boundary conditions" This sentence is broken, and needs revision.

    We have reformulated this sentence.

48. Figure 4: This figure needs resizing. Right now it is difficult to see what they are supposed to represent.

    The resizing will be done during the proof-reading process before publication in the two-column journal format. We think this figure should be displayed on a plain page.

Please also note the supplement to this comment:
http://www.biogeosciences-discuss.net/11/C9347/2015/bgd-11-C9347-2015-supplement.pdf