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Interactive Comment

Interactive comment on "Mapping pan-Arctic methane emissions at high spatial resolution using an adjoint atmospheric transport and inversion method and process-based wetland and lake biogeochemical models" *by* Z. Tan et al.

Z. Tan et al.

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General Comments Though the authors already carried out an extensive work and analysis, the following points need clarification and revision publication in ACP.

Response: We appreciate the valuable comments from the reviewer. These comments help us improve the manuscript in both readability and scientific values.

1.1 Satellite observations and bias correction Using satellite observations in an inversion system is a difficult task. Using SCHIAMACHY at high latitudes in support to sur-

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face in situ observations is even more difficult. The authors acknowledge this difficulty and apply filters on satellite data. They also worked on bias correction to minimize any misuse of satellite data in the inversion. However, in its current form, some questions remain unanswered and should be discussed. 1. Satellite bias is corrected along natural parameters (latitude, air mass factor, etc.) before inversion. Using the same data for debiasing and then for the inversion can be very hazardous. One should make sure that the bias patterns are totally decorrelated from the patterns used in the inversion (concentration gradients in this case). As methane emissions are dominant in tropical regions, concentration patterns could be somehow correlated with satellite bias. In this case, you risk misleading the inversion or at best reduce the number of usable information in the satellite observations. Has it be tried to include the bias correction in the inversion procedure?

Response: We have not tried to include the bias correction in the inversion procedure. In previous studies, some included and some did not. There is no claim that including the bias correction in the inversion procedure is better than the ones not including or vice versa. Given the risk that the further optimization of bias correction functions in the inversion cycle could cause bias correction to incorrectly account for the uncertainties brought by unaccounted model errors or even the uncertain sources and sinks (Houweling et al., 2014) and the inclusion also makes the inverse modeling system more complex, thus the inclusion was not chosen in this study. But as the pan-Arctic inversions are our focus, we did make an effort to detect the bias using independent observations. Specifically, we used the observed CH4 vertical profiles from the NOAA/ESRL aircraft mission over Alaska, the NIES aircraft mission over Siberia and the NASA/ARCTAS aircraft mission over northern Canada to build a relationship between the satellite bias and specific humidity averaged over the lower 3 km. It should make the debiasing process more reliable. See Fig. 3 for details.

2. Though efforts are done to deploy new observation sites around the Arctic ocean, satellite datasets could fill some gap in the observations. In my opinion, this paper

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has all the elements to partly address this question and should address it. What is the impact of using satellite data on the inversions? This could be estimated by computing the sensitivity matrix (Cardinali et al., 2004). It could also be inquired into by comparing inversions with and without assimilating satellite observations.

Response: Thanks very much for this suggestion! Accordingly, we have used a Monte Carlo stochastic approximation method to calculate the inversion uncertainty with and without assimilating satellite retrievals. It shows that assimilating satellite retrievals does reduce the inversion uncertainty.

1.2 Inversion system and uncertainties 1. The description of the system is somehow hard to follow. Section 3.3 should be clarified, in particular, concerning the nesting procedure and the spin-up periods. It looks like observations are used several times in the different inversions, spin up and nesting procedure. This could artificially increase the weight of the observations multiply used, compared with those used only once. Please discuss this point. It may be necessary to stop the spin-up period when the inversion period starts to avoid multiple use of information, biasing the inversion.

Response: The surface sites in the pan-Arctic were used in both global and nested-grid inversions. It could increase the weight of the NOAA/ESRL observations. But if it was not used in global inversions, we believe the boundary conditions of the nested-grid inversions would have much more errors. Since the NOAA/ESRL sites in the pan-Arctic provide much less observations (sometimes less than 1/50), this double counting should introduce much less errors than the method the reviewer suggested. Also, using surface measurements in both global and nested-grid inversions can be found in other previous studies such as Wecht et al. (2014). In addition, we have rewritten the description of the optimization and spin-up processes.

Wecht, K. J., Jacob, D. J., Frankenberg, C., Jiang, Z. and Blake, D. R.: Mapping of North American methane emissions with high spatial resolution by inversion of SCIAMACHY satellite data, J. Geophys. Res. Atmos., 119, 7741–7756,

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doi:10.1002/2014JD021551, 2014.

2. The global inversions are used as boundary conditions for the regional inversions. It would be interesting to see the impact of the higher resolution on the inversion results. Could the posterior fluxes from the global and the regional inversions be compared for equivalent regions? Anyway, I have some concerns about the way the nesting is carried out. If I understand well, the nested regional model is run on a grid, which does not extend north of 80°. This means that the transport across the Arctic ocean is totally excluded from the regional inversion. Thus, for instance, ZEP only sees the influence of the global boundary conditions as it is really close from the side of your regional domain. ALT is excluded from the regional domain while it is expected to provide some regional information, etc. In the best case, this is a pity of missing some potential information with air masses crossing the Arctic ocean and reaching remote sites. In the worst case, it totally biases the regional inversion and, at the end, the regional is not better (or maybe worse) than the global inversion. This problem must be addressed, especially as you use a relatively scarce network with Arctic sites relatively close to the border of the regional domain. That being said, I finally do not see what exactly brings the regional inversion to this study.

Response: We acknowledge that the exclusion of the North Pole in the nested grid could introduce some uncertainty to our estimates but do not agree with the reviewer's claim that this exclusion can totally bias the regional inversion and make the regional not better than the global inversion. We argue that, due to the following reasons, our regional inversion can do a much better job in helping understand CH4 emissions from the pan-Arctic. First, as we replied to one specific comment below, studies showed that in the summer time which we are interested in, vertical and zonal transport are much stronger than meridional transport. It is true that ALT is excluded from the regional domain. But we do not think that the exclusion of this site would make important regional information missed. The ALT site is located in a region far from possible CH4 emission hotspots. And because satellite retrievals in northern Canada are much more

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abundant than the ALT measurements, even if they are of less quality, the regional information they can provide is much better. Thus the scenario to damage our inversions as pointed out would hardly occur. Compared to coarse grid inversions, high-resolution inversions have many advantages: 1) because the footprint of satellite retrievals becomes more consistent with the finer grid cells, the chance they can be represented well in the GEOS-Chem model is much larger; 2) the impact of earth topography on the usability of satellite retrievals (tessellation error) is largely reduced. In summary, it is very unlikely that there is a large bias in the regional inversion due to the exclusion of the North Pole.

3. Concerning the prior uncertainties in the inversion, the current system uses a regularization term γ to control the weight of prior information compared with observations. How this term is computed? Is it based on a $\chi 2$ criterion? Couldn't the same procedure be used to also adjust the in situ vs satellite observations? It has been proven that prior uncertainties play a key role in inversion, and wrong uncertainty matrices can lead to totally biased or inconsistent results. Furthermore, a critical point in inversions is a correct specification of posterior uncertainties. Posterior fluxes without posterior uncertainties are mostly worthless numbers produced by very elaborated black boxes (to caricature...). The authors acknowledge this issue and try to address it by comparing inversion results for 6 different wetland prior fluxes. I am confident that these different scenarios can be sufficient to qualitatively discuss the performance of the inversion. In addition, it seems that the 6 scenarios are sufficient (by chance?) to reproduce a realistic range of uncertainty when comparing to Berchet et al. (2014) numbers for Siberian Lowlands. However, as the author try to draw some conclusions about the emissions from lakes, dominated by other sources, uncertainties might be too high. This is especially critical as the regional inversions seem kind of unsound. Additional inversions with different observation and prior uncertainty matrices would be necessary to really address this issue.

Response: The term γ is determined by analyzing its influence on the minimum of the

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cost function. It is a usual way to balance the prediction error and assimilation error in adjoint methods. More details can be found in Hakami et al. (2005), Yumimoto and Uno (2006) and Kopacz et al. (2009). For the emissions from lakes, we showed in Fig. 6 of the revision that the agreement between the GEOS-Chem model and SCIMACHY over a yedoma permafrost region (circled by a black polygon in Fig. 1) gets much better when the emissions from lakes were considered. There is a non-negligible possibility that the missed emissions by the DLEM scenario are from lakes because as illustrated, 56% of the water-inundated landscapes in this region are lakes. And it is possible that emissions counted for wetlands in other wetland models actually are from lakes. But we are cautious to draw a conclusion that CH4 emissions from lakes must be included in inversions or are significant across the pan-Arctic because there is still very large uncertainty. But the point is that the inversions in this study can shed light on this source at large spatial scales that are unachievable from field observations and the inversions are more reliable than biogeochemical models.

Hakami, A., D. K. Henze, J. H. Seinfeld, T. Chai, Y. Tang, G. R. Carmichael, and A. Sandu (2005), Adjoint inverse modeling of black carbon during the Asian Pacific Regional Aerosol Characterization Experiment, J. Geophys. Res., 110(D14), D14301, doi:10.1029/2004JD005671.

Kopacz, M., D. J. Jacob, D. K. Henze, C. L. Heald, D. G. Streets, and Q. Zhang (2009), Comparison of adjoint and analytical Bayesian inversion methods for constraining Asian sources of carbon monoxide using satellite (MOPITT) measurements of CO columns, J. Geophys. Res., 114, D04305, doi:10.1029/2007JD009264.

Yumimoto, K., and I. Uno (2006), Adjoint inverse modeling of CO emissions over eastern Asia using four-dimensional variational data assimilation, Atmos. Environ., 40(35), 6836–6845, doi:10.1016/j.atmosenv.2006.05.042.

1.3 Structure, content and title of the manuscript The manuscript in its current form lacks some consistency between the title, structure and content. The title makes the

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reader expects an atmospheric inversion accounting for lake and wetland emissions. Section 4.1 deviates in my opinion from the main topic of the paper. What is the objective of this section? In the current state, it looks like an enumeration of aggregated emissions on global regions and compared with previous work. Though by itself not uninteresting, I don't think it is relevant for Arctic inversions. Maybe the entire section could be moved to supplementary materials (or to a different paper dedicated to global inversions). On the other hand, Section 3.4 seems to me a key part of the manuscript. But the authors chose to put it only at the end of the method section with only limited details. I consider the satellite measurements play a key role in this work, especially as the Arctic in situ sites are very scarce during the inversion window. As noted by the authors, bias correction is essential for using both satellite and surface measurements. An amended version of the manuscript should include an extended discussion on the bias correction, on the performance of the different models, on the relative weight of satellite data in the inversion compared to surface measurements. This discussion is already partly done in Section 3.4 but should be extended and moved to Section 4. Some elements of Section 4.1 may also be used for this discussion. The title should render the use of satellite observations as it is not common in Northern latitude.

Response: We have changed the title to "Inverse modeling of pan-Arctic methane emissions at high spatial resolution: What can we learn from assimilating satellite retrievals and using different process-based wetland and lake biogeochemical models?" In the revision, we mainly focused on the following questions: 1) how large the impacts do the wetland biogeochemical models have on pan-Arctic CH4 inversions and in which direction can the wetland biogeochemical model can be improved for the use of inverse modeling? 2) can the inclusion of CH4 emissions from lakes improve the results of inverse modeling? 3) can the assimilation of satellite retrievals reduce the uncertainty of the posterior estimates? and 4) to compare the possible debiasing method for global or pan-Arctic scale inversions? And we have moved the most part of description about the optimization steps and results of global inversions to supplementary materials.

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Technical Comments The following points are mostly technical points that need reformulation or some clarification. p. 32471 l. 20: the last sentence might over-sell the paper or is too vague

Response: This sentence has been deleted.

p. 32472 l. 24: I think putting together ")(" should be avoided as much as possible. There are other occurrences of this typo point in the manuscript

Response: We have revised all these occurrences of ")(" in the manuscript.

p. 32474 l. 9: inversions are even more sensitive to uncertainty matrices; that should be at least partly addressed

Response: Thanks very much! In this revision, we have calculated the uncertainty of posterior estimates of methane emissions from the pan-Arctic. It shows that by using satellite retrievals the uncertainty is reduced.

p. 32476 l. 10: are the outliers numerous? What is the impact of this filtering on the inversion?

Response: We only find one outlier that can pass other quality tests in our study period. Thus we expect this filtering only has a trivial impact on the inversion.

p. 32476 l. 19: the selection is relevant, but some details on how it is done are needed for the reader. Couldn't the excluded sites be used for evaluation? A map of all the sites excluded from the inversion, assimilated in the inversion and used for validation should be provided (at least in the supplementary material), with the borders of the nested model.

Response: For the global scale, we excluded the same sites as in Alexe et al. (2015). We have added this citation for reference. For the nested model, we now added a new figure (Fig. 1) to show the sites assimilated in the inversion and used for validation. There are no surface sites excluded from both assimilation and validation in the nested

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inversions.

Alexe, M., Bergamaschi, P., Segers, A., Detmers, R., Butz, A., Hasekamp, O., Guerlet, S., Parker, R., Boesch, H., Frankenberg, C., Scheepmaker, R. A., Dlugokencky, E., Sweeney, C., Wofsy, S. C. and Kort, E. A.: Inverse modeling of CH4 emissions for 2010–2011 using different satellite retrieval products from GOSAT and SCIAMACHY, Atmos. Chem. Phys., 15, 113–133, doi:10.5194/acp-15-113-2015, 2015.

p. 32476: Maybe I missed it but I couldn't find anywhere whether surface observations are continuous or flask measurements.

Response: The surface observations are weekly flask measurements. We have added this information in this section.

p. 32478 l. 17: Can you give an exact definition of "lake"? This seems obvious, but the difference between wetlands and lake could be very tiny in some conditions? Does the map of lakes evolve with time?

Response: The lakes north of 60° N were retrieved from Global Lakes and Wetlands Database (GLWD). This map does not evolve with time. Tan and Zhuang (2015) have detailed description of the lake map processing. According to GLWD, lakes are defined as permanent still-water bodies (lentic water bodies) without direct connection to the sea. And wetlands are by nature transitional between terrestrial and aquatic ecosystems and have the presence of standing water for some period during the growing season, either at the surface or within the root zone. At least in GLWD, there is no double counting of lakes or wetlands. And we have acknowledged the possible uncertainty introduced by the double counting in the revision. We have added the definition of "lake" into this section.

Tan, Z. and Zhuang, Q.: Arctic lakes are continuous methane sources to the atmosphere under warming conditions, Environ. Res. Lett., 10, 054016, doi:10.1088/1748-9326/10/5/054016, 2015.

p. 32479 l. 10: Is there any citation comparing GEOS-4 and GEOS-5? As you use different meteorological forcings for the different inversion windows, it could have an impact on the results. The two datasets are probably very consistent and the impact is probably very limited, but this should at least be mentioned.

Response: In our revision, the GEOS-4 meteorological forcing was only used for constructing initial conditions on January 1, 2004. Thereafter, all inversions used the GEOS-5 meteorological forcing, including global scale and nested grid inversions. Additionally, in the revision, we moved the start time of nested grid inversions from July 1, 2004 to July 1, 2005. With such a change, we expect that any signals that could be caused by the inconsistency between GEOS-4 and GEOS-5, if any, should have disappeared after the transport and assimilation processes of one and a half years.

p. 32479 l. 14: if I understand well, for instance, if an air mass from Canada crosses the pole and reaches a site in Siberia, you wouldn't be able to recover any information on the emission with your way of dealing with the pole? It would be then mixed with "boundary" polar conditions? You might lose a lot of information on Arctic emissions considering the fast transport of air masses over the Arctic Ocean. Wasn't it possible to implement the procedure of the global system in the nested system?

Response: We did not include the polar area for the following reasons. First, in GEOS-Chem, with the concern of numerical stability, there is a special treatment of advection in the polar region (Lin and Rood, 1996), but this treatment has not been applied and tested for the nested grid. Second, according to Miyazaki et al. (2008), the Northern Hemisphere (NH) extratropics during summer has slow mean-meridional circulation and inactive wave activity but strong vertical transport. Thus there should be very few air masses from Canada crossing the pole and reaching a site in Siberia or vice versa. Third, it is true that the boundary conditions of the nested model could miss the signals out of boundaries. But this is the drawback of all the similar applications, regardless whether it is in North America or in the pan-Arctic. The possible solution is to construct the boundaries by real data but it is out of focus of this paper. Instead, we

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have acknowledged this problem in our discussion and called for the improvement of the GEOS-Chem model.

Lin, S.-J. and Rood, R. B.: Multidimensional Flux-Form Semi-Lagrangian Transport Schemes, Mon. Weather Rev., 124, 2046–2070, 1996.

Miyazaki, K., Patra, P. K., Takigawa, M., Iwasaki, T. and Nakazawa, T.: Global-scale transport of carbon dioxide in the troposphere, J. Geophys. Res., 113, D15301, doi:10.1029/2007JD009557, 2008.

p. 32480 l. 10: people unfortunately do not always define Arctic the same way... Please give your definition, so that the reader knows on which region your emissions are defined.

Response: We have removed the word “Arctic” here and the nested domain has defined in the previous paragraph (180°W–180°E and 80°N–56°N).

p. 32482 l. 22: does the system guarantee that it is not stuck in a local minimum? I guess it does, but mentioning only the 0.5% criterion might be insufficient

Response: Yes, the system guarantees that the iteration is not stuck in a local minimum. We have mentioned in the sentence “optimization changes its course automatically if local minimum reaches”.

p. 32483 l. 14: BIC seems a reasonable score but it is not commonly used, so please give a little bit of details on it.

Response: We added some descriptions of the method: “The BIC criterion is widely used for regression model selection and awards a model that fit measurements with the least model parameters.”

p. 32483 l. 25: Does filtering outliers influence the bias correction? What is the portion of data filtered out along this criterion?

Response: The grid squares with RSD in excess of 20 ppb are not outliers but just

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as indicated by Turner et al. (2015) they are more likely dominated by bias in prior emissions or strong local emissions. If these values are included, the bias correction will either remove local emission signals or account for biases not belonging to SCIAMACHY retrievals.

p. 32484 l. 15: is there a known reason for the opposite dependence of model-data differences in East Asia? This only comes from wrong emission inventories or is there a relation with regional meteorology or other?

Response: According to Peng et al. (2016), the EDGAR dataset could overestimate anthropogenic CH4 emissions from China.

Peng, S. S., Piao, S. L., Bousquet, P., Ciais, P., Li, B. G., Lin, X., Tao, S., Wang, Z. P., Zhang, Y., and Zhou, F.: Inventory of anthropogenic methane emissions in Mainland China from 1980 to 2010, Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-139, in review, 2016.

p. 32484 l. 22: I do not understand why you need these polynomial trends? Is it that you use monthly or 2-weekly flask measurements and extrapolate them to hourly residuals? If so, I think this might be a problem for the inversion. Extrapolating data before inversion can only bring additional uncertainties.

Response: In the revision, we directly compared the weekly flask measurements (the data records include the measurement date and UTC information) to the model.

p. 32486 l. 3: Please remind the inversion windows here. It is not always clear when the satellite data are used.

Response: The global scale inversion window is from January 2004 to December 2004 and January 2005 to December 2005. The inversions of the second time window are for analysis.

p. 32488 l. 20: it would be easier for the reader to draw a picture if the same area were compared.

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Response: Our results cannot directly compare with Monteil et al. (2013) because they only reported the CH_4 emissions from the areas north of 50°N.

p. 32489 l. 13: without uncertainties on the posterior, it is hard to see the impact and the confidence of the inversion. The subsequent discussion is thus very speculative in my opinion. The DLEM scenario with no lakes only shows the limitation of inversion methods, I think... I do not really get the choice of DLEM. The way you put it, it only confirms that the inversion has not enough information to redistribute fluxes. But the missing fluxes could also be wetland fluxes.

Response: We have calculated posterior uncertainty in the revision.

p. 32490 l. 18: both numbers looks pretty high, especially for the total column. What the difference between observed and prior total columns? Is the improvement significant? I think this is the most important here. If with the inversion, you only shift the total columns of 1 ppb without the lakes and of 2 ppb with the lakes, you got a signal; but conversely, if the inversion shifts the total columns by e.g., 30 ppb without the lakes and 31 ppb with the lake, you got nothing...

Response: We have drawn another figure to show the difference. As shown in Fig. 6, there are visible differences.

p. 32490 l. 22: I think this citation is not relevant. They could have achieved 15 ppb of improvement if taking wrong prior fluxes...

Response: We have removed this citation.

p. 32491 l. 26: Berchet et al. (2014) did find methane emissions of 1–13 $TgCH_4/y$ from Siberian wetlands, which is amazingly consistent with your figure.

Response: Our newly estimated methane emissions from Siberian wetlands are 1.6–7.6 Tg yr-1.

Tab. 1: Maybe you could add correlation coefficients as you show one R in Figure 1.

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Response: We have added it.

Figure 1c: it would be interesting to compare on the same figure before and after optimization and to have the same figure for all debiasing method (probably in supplementary material to avoid having dozens of figures...)

Response: As shown in Table 1, the fitting between model and SCIAMACHY does not differ too much among several methods, e.g. between "Latitude only" and "Latitude + Humidity". Thus such plots probably will not bring much information.

Figure 4: Could you please add the prior and posterior uncertainties? Why does the seasonal cycle vanishes after 1998 in the Tropics? As for Section 4.1, I am not sure this figure is really relevant regarding the topic of the paper

Response: We think you are right – this figure seems irrelevant to our topic. It only shows the process of initial condition construction. We have removed it in the revision. For the vanishing of the seasonal cycle after 1998 in the tropics, it is related to the discontinuation of the biomass burning emission dataset. In GEOS-Chem, the GFED3 dataset covers only from 1997 to 2010 and all simulations before 1997 have to use the data of year 1997. Compared to the other years, biomass burning emissions have more apparent seasonal cycle in 1997.

Figure 8-9: Please add the prior RMS for each different scenario, so that one can see the improvement after inversion.

Response: For both figures, we have added the prior RMS for each different scenario.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 32469, 2015.

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