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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Retrieval of methane source strengths in Europe using a simple modeling approach to assess the potential of space-borne lidar observations" *by* C. Weaver et al.

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The authors thank both reviewers for their helpful comments that should clarify our message.

Response to Reviewer 1's comments

Rev1: P.19563, Line# 10ff: A bit concerned about this fundamental assumption. The daily variations are likely to be caused by local emissions or synoptic transport as

revealed by the Transcom continuous experiment for CO2 as well as CH4 simulations later using Eulerian chemistry-transport model (Patra et al., JMSJ, 2009

That means, to be able to derived emission information one need to know the transport perfectly at hourly timescales and at very fine resolution. I have my own reservation whether or not 0.5x0.5 longitude-latitude boxes are good enough for that.

Additionally, flexpart may not be a good model for the species that have long lifetime and are globally well mixed.

AC: Yes, we were initially concerned that surface methane time series at the ground-based observing stations would be dominated by local sources and that FLEXPART (along with the 0.5 degree GFS met fields) would be unable to capture the fine spatial scale transport. But there are stations where our model is able to simulate the ground based methane observations - here the long range synoptic transport dominates and must have been correctly simulated.

P.19563, Line#26: How good is this 150m of emission height approximation, when you have sites respresenting wide variety of sites. For example the air column above Jungfraujoch site, at 3580m, can be very thin compared to the column above Mace Head.

AC: Yes, the 150m emission height is typically used by experienced FLEXPART users emitting at the earth surface. I suppose next time we could base the emission height on the boundary layer height.

P.19563, Line#27: If you have assumed 20 days lifetime for the CH4 particles, why again this OH chemistry? The OH chemistry with instantaneous lifetime of longer than 3-4 years in the mid-high northern latitudes should have non-measurable impact on the forward simulations.

AC: Our results should not be dependant on the inclusion of the OH chemistry package in FLEXPART. We simply turned it on for completeness.

P.19564, Line#2: What is the rationale for having a model top up to 400 hPa - is it the typical height of the tropopause over your model domain or the experimental design for measuring CH4 column below 400 hPa? Please clarify.

AC: Thanks, we added a sentence: Our configuration of the FLEXPART model does not loft many particles (molecules of methane) above the 400 hPa level so its contribution to the column perturbation is insignificant. The partial column methane above 400 hPa is spatially homogeneous and is included in the Background term.

P.19564, Line#6: may be 'i-th' is better. AC: It should have been superscripted – I will check for that when its proofed.

P.19565, Line#2: I think it is not the effect of horizontal resolution, more important probably is how the meteorology is paremeterised in Flexpart. Also, I am not convinced by the methodolgy applied for correlation analysis. Looks like you correlating concentration at hourly time intervals, please correct me if i am wrong. I would like you to separate diurnal and daily variability and then make the correlation analysis, as in Patra. et al. (2009). That will enable you to understand the Flexpart transport quantity better, in my opinion.

AC: Thanks, we added a sentence: 'Differences between FLEXPART simulated concentrations and the observations can be attributed to: parameterization schemes employed by FLEXPART, errors in the GFS winds and errors in the observations themselves.'

We did investigate the impact of diurnal effects on our analysis by doing correlations separately for both daytime and nighttime conditions – we found little change in the correlation coefficients – so we did not pursue this.

P.19566, Line#22: Ideally yes, you want trajectory to travel over all boxes, but what about employing covariance between boxes with spatial correlation?

AC: Since, the transport patterns are in the form of filaments rather than a diffusive, we

did not want covariance between different boxes.

P.19567, Line#3-4: I am surprised that a good agreement is claimed here with MACC! Can you be quantitative, say for spatial patterrn or strength?

AC: Not sure how to improve the existing wording.

P.19567, Line#27: What is the connection here with tulip industry? Was there a link suggested by someone? Give reference or else delete

AC: No comment

P.19568, Line#5ff: Lacks discussion, please discuss the role of the baseline determination on your results

P.19568, Line#2: How? Column upto 400mb is only about 60% of the airmass, and recent analysis show that CH4 column above the tropopause is also important for variability in total column CH4.

See sentences added on P.19564

P.19568, Line#2: What about systematic bias, which is what the most dangerous for source/sink inversion (Patra et al., JGR, 2003), and GOSAT inversiions struggling hard to deal with the retrieval bias (e.g., Basu et al., ACP, 2013; Maksyutov et al., ACP, 2013).

AC: Until the MERLIN/GSFC instruments are built we don't have any information on how to characterize the instrument systematic bias so we really can't address this. There is a potential systematic bias from data temporal/spatial sampling and rejection of data due to cloudy conditions. These are accounted for in the new section: 'Application to Remote Sensing'

Reviewer #2

Unfortunately, this sensitivity analysis is based on forward simulations of XCH4 for only

2 days, and is missing important elements such as the expected spatial and temporal coverage of the satellite data.

AC: Our sensitivity experiments now consider 20 days in summer and 20 in winter. See new section: 'Application to Remote Sensing'

Furthermore, a severe limitation of the analysis is that it takes into account only the vertical column between the surface and 400hPa, i.e. ignoring 40% of the total column. Although most of the CH4 variability is indeed expected in the boundary layer, the upper troposphere and stratosphere may also contribute to variations of the CH4 mixing ratios averaged over the total column, which needs to be further analyzed. I assume that the column averaged mixing ratios shown in Fig. 7 represent just the average between the surface and 400hPa.

AC: Thanks, we added a sentence: Our configuration of the FLEXPART model does not loft many particles (molecules of methane) above the 400 hPa level so its contribution to the column perturbation is insignificant. The partial column methane above 400 hPa is spatially homogeneous and is included in the Background term.

The paper completely lacks any discussion of vertical sensitivity (averaging kernels), expected spatial and temporal data coverage and potential systematic errors of the LIDAR instruments. The authors only compare their calculated relatively small signal in daily XCH4 of the assumed 50% reduction of CH4 emissions over Germany and the Netherlands (âĹij3 ppb) with the expected instrument precision (âĹij14 ppb), and estimate that at least monthly averaged measurements would be required for detecting the emission reduction, assuming a reduction of the random error by 1/ôŔřŰ(N), while not discussing any systematic errors which might become limiting.

AC: See last comment of Review 1

Furthermore the signal in the monthly average XCH4 might be smaller than in the shown daily maps. Overall, the presented very short analysis is not sufficient to quan-

titatively assess the sensitivity of the space-borne LIDAR measurements to changes in surface emissions. A much more detailed analysis would be required which should also include Observing System Simulation Experiments (OSSEs) taking into account the major aspects of the instrument (and the inverse modeling system), especially vertical sensitivity, expected spatial/temporal data coverage and systematic errors of the LIDAR instruments (as well as systematic errors of the modeling system), as e.g. performed for SCIAMACHY

AC: The new section: 'Application to Remote Sensing' describes an OSSE-like sensitivity study. It accounts for any potential systematic bias from data temporal/spatial sampling and rejection of data due to cloudy conditions. Until the MERLIN/GSFC instruments are built we don't have any information on how to characterize the instrument systematic bias so we really can't address this. Our current OSSE-like study looks at 20 days in summer and 20 in winter. We will extend this time duration when more funds become available.

Although the presented analysis of European CH4 emissions based on the inversion of measurements from 9 surface stations is capable to identify some major CH4 emission regions (Netherlands, and coal mining areas in Poland), the derived emissions per 'tile' and 45 day time step seem to have considerable noise (i.e. very large variations between the time steps, which are probably not very realistic), which is a common problem of many inverse modeling systems. It would be useful to better analyze these fluctuations and try to better separate the signal from the noise (e.g. by analyzing averages over larger areas and longer time periods).

AC: I suspect that if we averaged our results over larger grid boxes and longer periods we would get the same results if we analyze over larger grid boxes and longer periods. To reduce the noise we would need more monitoring stations or better treatment of the methane transport from regions outside our 262 grid domain.

The analysis of only 7.5 months is very short. At least one full year should be sim-

ulated to allow clearer conclusions about the contribution of seasonally varying emissions from wetlands. A significant limitation of the presented inverse modeling system seems to be the treatment of the background, which is assumed to vary linearly over 45 days, and which therefore is not capable to account for variations in the background concentrations on shorter timescales. It seems likely that the general rather poor model performance for the mountain stations (e.g. observations at Jungfraujoch and Plateau Rosa are often several 10 ppb below model simulations / retrieved baseline; Fig.4) is largely due to variations of the background concentrations, which are not properly simulated. Furthermore, it is noted that the retrieved baselines show considerable discontinuities between the 45 day-periods (Fig. 4). The simulations could be significantly improved by using background fields from global CH4 inversions (see e.g. [Rödenbeck et al., 2009]).

AC: We tried to address the problem of outside transport by using CH4 fields from a 3D Eulerian transport model. But inclusion of these fields significantly degraded our results. Our analyzed surface CH4 concentrations were very different than the observations – so Figure 4 looked terrible. This is why we resorted to a simple linearly varying background. If we continue this study we will look at CH4 from global inversions [Rödenbeck et al., 2009]. Thanks

The CH4 inversions from the MACC project are not a very good reference for the specific purpose of this paper (since the MACC inversions, aiming mainly on the global scale, have been performed on coarse model resolution and do not assimilate European surface observations). It would be more appropriate to compare with CH4 inversions on higher spatial resolution based on European surface measurements (e.g. [Bergamaschi et al., 2010; Manning et al., 2011]). None of the existing regional inversion studies is cited or discussed in the paper (beside the mentioned European inversions, there are also many inverse modeling studies for the US (e.g. [Kort et al., 2008]).

AC: We chose the MACC for comparison because it did not assimilate surface obser-

vations. We have included and discussed Bergamaschi et al., 2010; Manning et al., 2011, Thanks.

In many parts to the paper the discussion seems not very well elaborated. E.g. on page 19569 the authors state "This will definitely be an improvement over the current passive satellite instruments.", without giving any reference.

AC. Removed

According to Table 1 the measurements from Kollumerwaard are reported on the NIST scale, while all other measurements are reported on the NOAA04 scale. It is not discussed in the paper, if and how the data have been converted to a common CH4 calibration scale.

AC. The difference between the NIST and NOAA04 scale is between 0.1% - 0.3% - so several ppb. Since this is an offset, we did not convert the data to a common scale since this bias would be included in the retrieved background values.

Citation: Dlugokencky, E. J., R. C. Myers, P. M. Lang, K. A. Masarie, A. M. Crotwell, K. W. Thoning, B. D. Hall, J. W. Elkins, and L. P. Steele (2005), Conversion of NOAA atmospheric dry air CH4 mole fractions to a gravimetrically prepared standard scale, J. Geophys. Res., 110, D18306, doi:10.1029/2005JD006035.

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