Reply to review 2

This work documents an interesting case study in which a Lagrangian Particle Dispersion Model (LPDM) is used together with high-resolution satellite data in an inversion. The paper describes an efficient method for producing SRR 'footprints' using an LPDM from a large number of satellite retrievals, which would ordinarily be resource-intensive and time-consuming. The new method is applied to a northern high-latitude region and results are compared to an inversion using ground-based observations within the same region. The authors found that the satellite-based inversion produced posterior fluxes which were very close to the prior, and put forward suggestions for why this was the case.

Overall this is an interesting study and a nice documentation of a method that will likely become more and more important as satellite resolution continues to increase. The method is well-described on the whole and the figures are OK. I have some comments and suggestions regarding some aspects of the method and the presentation, but I am happy to recommend publication of the manuscript after these are addressed.

We thank the reviewer for this comprehensive review. We insert our replies (indicated in blue) to each comment below.

Major comment

Throughout the document, the authors correctly refer to the fact that LPDMs are not usually used for this type of satellite-based inversion – e.g. 'LPDMs have not been used to any significant extent with satellite observations' (line 48).

However, I do think some discussion of any previous satellite-based LDPM inversions is merited. It should be made clear exactly what exactly the novel parts of this new methodology are. For example, Ganesan et al. (2017) performed a GOSAT-based inversion over India using a different LDPM. There should be some discussion of how your method differs to and updates theirs, as is necessary due to the increased volume of data available from TROPOMI compared to GOSAT. The novel aspects of your method should be highlighted more explicitly (e.g. the use of $P_n = Pa_n w_n$, in Eqn. 7, which is nice).

We agree that we should specifically mention the previous studies using Lagrangian models to model satellite observations and discuss how our study differs from these. We now refer to these studies in the introduction L51 and include a line after Eq. 5 discussing how our method compares to what has previously been done.

Specific comments

Line 85: 'SRRs have only been calculated for point observations ...'. For FLEXPART, although this is not true of other Lagrangian models.

We were referring to FLEXPART, but we agree that this statement is misleading and that there are examples of studies using LPDMs to model satellite observations (e.g. studies by Wu et al. 2018 and Ganesan et al. 2017). We have now corrected this statement to state emphasising instead the use of an LPDM with large numbers of satellite observations.

Line 101: How exactly might chemical loss be represented during the backward simulation? Via an assumed fixed lifetime, or via actual representation of OH concentration and chemical loss rates? Is there likely any effect from not including this here?

Linear chemistry can be accounted for in the backwards simulations. In this study, we have accounted for the loss of CH_4 due to reaction with OH by using pre-calculated 3D and monthly fields of OH concentrations, which were computed by the GEOSChem model. The loss of CH_4 due to OH in this study, however, is very minimal since the particle trajectories were only calculated for 20 days backwards in time. The effect of accounting versus not accounting for OH in this case is likely negligible owing to the high latitude of the domain and the length of the trajectory calculations.

Line 195: Is it possible to include a map of the variable resolution of the optimised flux grid?

We now include maps of the variable grid used for the TROPOMI and the ground-based observation inversions in the Supplement (Fig. S1).

Line 255: Can you provide a measure of the chi-square value for the various retrievals to test the convergence?

For the retrieval product that we use, namely WFMD v1.8, there is no chi-square value provided. Instead the data provider gives a binary quality flag (0 = good, 1 = bad) based on a number of criteria (Schneising et al. Atmos. Meas. Tech., 2019) and we have selected only the “good” data.

Line 295: I am a little surprised at the limited posterior uncertainty reduction. You say that this is to be expected due to limited observational coverage due to cloud cover and other issues, but you say earlier that there are over 3500 super observations per day, with TROPOMI having good sensitivity down toward the surface. Is the satellite coverage quite heterogeneous? Perhaps a figure showing the super observation coverage density by the satellite over the region for the spring and summer periods would be useful (perhaps alongside the SRR map in Figure S6).

We have included figures showing the number of observations per 14-day time interval for the TROPOMI super-observations and for the ground-based observations (see Supplementary Fig. S2). For the TROPOMI observations, we show the number per time interval as well as latitudinal intervals of 5° to indicate how the number of observations also varies by latitude. The number of observations strongly decreases with latitude, especially in March and October, as expected. There are also fewer observations from the mid May to mid July in the latitudes north of 50°N owing to cloud cover.

Concerning the low uncertainty reduction in the inversion using TROPOMI, this has a number of reasons other than the number of available observations. First, the TROPOMI XCH4 observations are more uncertain compared to the ground-based observations. The median uncertainty for XCH4 was 16 ppb compared to 8 ppb for the ground-based observations, and the model-observation errors are weighted by the inverse square of the uncertainty. Second, the satellite SRRs are smeared out over larger regions, compared to the SRRs for ground-based observations, which are focussed over smaller regions. This leads to stronger deviations in the modelled mixing ratios relative to the background (i.e., if there are sources in the SRR region) for the ground-based observations compared to the satellite observations. Since the cost function (in the inversion) includes the quadratic difference between observation and modelled mixing ratio, a few large differences have more impact than a large number of small differences. This means that the ground-based observations (with more focused SRRs) have more impact in the inversion.

Line 295: If you were expecting such poor satellite coverage over a difficult-to-observe region, why did you choose this region for your case study?

We were actually surprised that there is almost no uncertainty reduction over the domain in the inversion using TROPOMI. We had chosen this study domain as it extends as far south as 40°N meaning that at least in the southern part of the domain there should be sufficient number of observations, and because this domain contains quite important CH4 sources from coal, oil and gas, as well as important wetland fluxes (namely, in the Western Siberian Lowlands). A further, non-scientifc reason for choosing this domain was because the project funding this work was focused on high-latitude CH4 sources, so we were obliged to look for a domain including high latitudes.

Figure 10: I think there are some data points falling outside of the boundary of the Taylor diagram (around NSD = 2.5; R = 0.5). These are easily missed by the reader and the Taylor diagram should be expanded to include them if possible. Also please include a label on the angular axis.

We have expanded the radius of the Taylor diagram to include the points that were "off-scale" in terms of NSD. We also include the label on the angle, i.e., "Correlation".

Supplementary Figure: I'd be interested to see a plot showing 'posterior – prior' XCH4 for the results shown in Figure 2. Is there any spatial variability in the change to XCH4 produced by the inversion in this case or is it simply a homogeneous addition/subtraction of background CH_4 within that latitude band?

We have now included a figure in the Supplement (Fig. S4) showing the differences between the prior and posterior XCH4 for March and for July. There is some spatial variability in the change a posteriori relative to a priori: for March the increment is more negative in the north versus the south, for July the increment is small negative in the north and east and slightly positive in the west and south.