Reply to Review 3

We thank the reviewer for the comprehensive review and reply to each of the comments below.

Comment 1:

The paper by Bakels et al. (2024) describes the new FLEXPART v11. We presume the reviewer is referring to section 9.2 of Bakels et al. (2024), which describes how in FLEXPART v11 a simulation can be started using a custom particle initialization, which in principle could be used to represent satellite retrievals. However, there is no code provided in FLEXPART v11 to generate such a custom particle initialization, and modelling satellite observations is just mentioned as a possible application. In fact, this possible application is mentioned as it is exactly the work described in this manuscript, that is, how to make particle releases for a satellite retrieval in order to calculate total column SRRs. The reason why this manuscript refers to FLEXPART v10.4, and not to v11, is because the developments started before FLEXPART v11 was ready, and because only the FLEXPART v10.4 code is able to be used directly to calculate total column SRRs. It is planned, however, to release code in the future to initialize particle releases for satellite retrievals for use in FLEXPART v11.

Bakels et al. (2024) also describes the Linear Chemistry Module (LCM), which was implemented by me (Rona Thompson). The LCM is only a forward in time simulation mode and does not provide source receptor relationships (SRRs), which are needed for the inversion and are described in this manuscript. In the LCM mode, the whole domain is filled with virtual particles, which are then sampled to represent observations, either in-situ observations or satellite retrievals.

Comment 2:

The main purpose of the manuscript is to describe the methodology for calculating total column SRRs that could be implemented in any Lagrangian particle dispersion model, and not specific to FLEXPART. We prefer not to include details of the new routines in FLEXPART v10.4 in the manuscript, however, we now include these in the supplement.

Notice specifically that, in FLEXPART v11, there were no code changes made that are specifically related to modelling satellite column SRRs. However, the new v11 feature allowing to define particle releases with complete flexibility (section 9.2 of Bakels et al., 2024) makes it possible to define these releases, exactly as described in this study, such that FLEXPART v11 will determine satellite column SRRs. One may therefore consider this study as describing the pre-processing step that is necessary to produce the FLEXPART v11 input files for satellite column SRRs. In FLEXPART v11 itself, no changes are needed, other than for the output of the SRR files for satellites.

Notice also that this could be used, for instance, to provide corresponding input data files to produce SRRs for slant column measurements. Again, no changes would be needed in FLEXPART v11 (other than for the output of the SRR files for satellites), and the particle release files could be produced in a very similar way as described in this paper – just accounting for the additional complexity of the slant column geometry.

Comment 3:

In Eq. 6, *t*, is the residence time of the particles in the grid cell as also described in Seibert and Frank (2004). We have modified Eq. 6 and instead of mass we now use the transmission function, which represents the fraction of mass remining in a particle which may change if there is atmospheric chemistry (as described in Seibert and Frank (2004)). We have also changed the following equations for the calculation of $\mathbf{H}^{col}$ accordingly.

Comment 4:

We have updated the equations (as mentioned in response to the previous comment). The main point is that the equation for the total column SRR can be expressed as a sum over all particles in all layers when the information on how each layer's SRR ($\mathbf{H}_n$) contributes to the total column, i.e. the weighting by $a_n w_n$, is incorporated into the initialization of the particles. Here we incorporate that information by varying the particle density in each layer where the number of particles in layer n is $P_n = P a_n w_n$ where P is the total of number of particles released per retrieval.

In Eq. 11 (formerly Eq. 10) there is no division by density, since what is wanted is the number of particles terminating in a given grid cell relative to the total number of particles released. This gives a unitless fraction. This fraction is calculated for all grid cells in 3D space and together defines a weighting matrix for the influence of the initial mixing ratios on the column observation.

Comment 5:

In the original FLEXPART code, releases could only be made for rectangular volumes which were aligned with the meridians and parallels (the particles are randomly distributed in these volumes). However, the satellite pixel geometry can be rotated with respect to the meridians and parallels and may not be rectangular. In the case of TROPOMI, the pixels are trapezoids and are rotated. Therefore, to make a release that represents the exact geometry of the satellite pixel, we perform an affine transformation. This involves the following steps:

1. Calculate the angle of rotation of the satellite pixel relative to the meridians
2. Calculate the equivalent lat and lon bounds of the unrotated pixel
3. Calculate the angle of distortion from satellite pixel
4. Calculate the equivalent lat and lon bounds of the undistorted and unrotated pixel
5. Distribute the particles randomly within this rectangular volume
6. For each particle, reapply the rotation and distortion so that the particles all fall within the original satellite pixel.

This is performed in the subroutine releaseparticles_satellite.f90 which is available from the open gitlab repository: https://git.nilu.no/flexpart/flexpart.git

We now include the above information in the supplement. Since the affine transformation is a whole algorithm we do not include all the equations in the supplement but refer the reader to the code in the aforementioned repository.
Also notice again that all of this can be considered as a pre-processing step for defining the particle release file in FLEXPART v11, since in this version the individual particle release positions can be specified in a file part_ic.nc. These positions are completely independent of a particular grid geometry. This was not possible in the older FLEXPART v10.4 version still used in this paper.

Comment 6:

The super observations are created for rectangular grid cells, which are aligned with the meridians and parallels. We now specify this in Section 2.2.

Indeed, the denominator of the third term on the RHS of Equation 12 should be S and not M. This is a typo in the writing of the equation only, the expression in the code is the correct one.

Equation 14 (formerly Eq. 13) cannot be simplified further, as the reviewer suggests, because:

$$\frac{1}{S}\sum_{m=1}^{M}\sum_{n=1}^{N} s_m a_{mn} w_{mn} x_{mn}^{pri} \neq \sum_{n=1}^{N} \overline{a_n w_n} \overline{x_n^{pri}}$$

since $xpri_n$ and a_n are unique for each retrieval. (Note division by S on the RHS is not needed since the mean of a_n, w_n and $xpri_n$ are used).

Comment 7:

The prior error covariance matrix also includes temporal correlation of prior flux errors. The temporal correlation is calculated using exponential decay with time with a correlation length of 28 days. We have added this information to Section 3.1.3.

Comment 8:

We include a short new section, 3.2.1, on the inversion diagnostics and put figures of the cost per iteration in the Supplement.

We think the reviewer is referring to the reduced chi-squared value, which is twice the final cost divided by the number of degrees of freedom, which has an expected value of one if the uncertainties are appropriately chosen (Rodgers, 2000; Tarantola, 2005). In practice, however, it is not always possible to achieve a value of one, and as pointed-out by Chevallier et al. (2007), the reduced-chi-square criterion can be ambiguous and alone is not a sufficient criterion for assessing the appropriateness of the uncertainties. In any case, the reduced chi-square values were as follows:

1) using TROPOMI: 1.08

2) using ground-based observations: 4.86

Comment 9:

Indeed, the TROPOMI observations provide little constraint on the fluxes at this high latitude. The reason for this is that the TROPOMI XCH4 observations at this latitude are more representative of background air and not so sensitive to fluxes at the surface. For spring and autumn, depending on atmospheric temperature structure, this can be because the boundary layer is shallow, and below the level of peak sensitivity for the TROPOMI instrument (as can be seen from the averaging kernels).

The observation uncertainties (i.e., for the super-observations) are determined from the retrieval information, namely, these are calculated as the area-weighted quadratic sum of the individual retrieval uncertainties (as determined from the retrieval algorithm). The median observation uncertainty was around 12 ppb and did not vary much between months. In addition, we consider a background uncertainty estimate. In total, the observation space uncertainties had a median value of 16 ppb and inter-quartile range of 14 – 18 ppb. We consider this to be a very reasonable estimate of the uncertainty of the column average mixing ratios at this latitude and not an overestimate. Of course, increasing the observation space uncertainty would further reduce the constraint on the fluxes.

We did perform a number of sensitivity tests but did not include these in the paper, since the focus is rather on the methodology of using satellite observations in an inversion based on a Lagrangian transport model. We tested using an uncertainty of 50% of the prior flux value (as was also used in the inversion using ground-based observations) and then because the posterior fluxes did not differ from the prior fluxes, we increased the uncertainty to 100%. The result remained the same, i.e., the posterior fluxes remained very close to the prior fluxes. We now include this information in Section 3.1.3.

Comment 10:

As we state in our reply to comment 9, we consider that the prior and observation uncertainties are appropriately chosen. Furthermore, for the inversion using TROPOMI we used twice as large prior uncertainties compared to the inversion using ground-based observations.

Comment 11:

At this northern latitude, the answer is that TROPOMI provides little constraint on the surface fluxes. This result has also been found by an independent study using Eulerian atmospheric transport model and an ensemble data assimilation algorithm (Tsuruta et al., Remote Sens., 15, 1620, https://doi.org/10.3390/rs15061620, 2023).

We have used our FLEXPART code and the inversion framework, FLEXINVERT, also in a study using TROPOMI over Europe and find there that TROPOMI provides a more

significant constraint on fluxes in southern, western and central Europe, but not a very large constraint in northern Europe.
However, in general it is to be expected that satellite observations have a weaker constraint on the fluxes than ground-based observations. First, the satellite observations are more uncertain compared to the ground-based observations (the median uncertainty for XCH_4 in our study 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.

Minor comments:

We have added titles to Fig. 2 and the units are given in the figure caption.

We have added country labels to Fig. 1