Response to Anonymous Referee #2

We thank Referee #2 for the constructive assessment of our paper. In the following, we respond to the individual points raised in italic letters (in blue), the reviewer comments are left in normal font (in black).

The manuscript describes a regional inverse modeling system that uses atmospheric observations to estimate GHG fluxes. Synthetic experiments are performed to evaluate the system. A number of specific aspects need to be addressed before I can recommend accepting the manuscript for publication.

Main comments:

Regarding the number of regions: I fully agree with referee #1 in that a prior error of 100% for different regions with a changing number of regions will result in a decrease of the uncertainty of the spatially integrated fluxes, as the errors are assumed uncorrelated.

One possible method would be to inflate the variance accordingly such that the prior error of the spatially aggregated flux does not change with the number of regions. However this has an impact on the comparisons between flux estimates using a different number of regions to be solved for.

Authors' response: In our results, increasing the number of sub-regions can increase or decrease the total (sum of the sub-regions) posterior error, and similarly increase or decrease the posterior uncertainties. The optimization procedures can behave in a complex way with the number of sub-regions.

For example, it is possible for the atmospheric transport to correlate the flux signals from different sub-regions. If a parcel trajectory passed through different sub-regions with non-zero fluxes over the 5-day period, then the flux signals from the different sub-regions could be mixed (correlated) within the parcel. Integrating the contributions from all the parcels, the flux errors for the different sub-regions in combination with transport errors could be correlated in complex ways (particularly when the errors are biased).

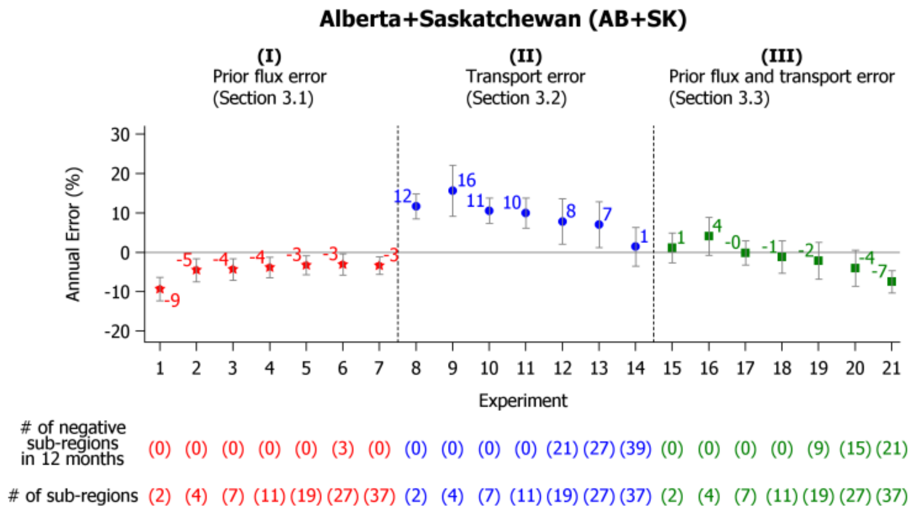
As it is likely not the case in our model that the 'errors are uncorrelated', the posterior uncertainty of the whole region in our results could go up or down as it is divided up into a larger number of sub-regions (and not always 'go down').

These characteristics in the posterior uncertainties can be seen more clearly in the revised version (see below) of Figure 5 in which the scales of the y-axes have been changed to improve readability. The number of negative sub-regions for the corresponding number of sub-regions is shown on the 2nd row from the bottom in the figure. These explanations will be added to the manuscript revision.

This points to the value of evaluating the inversion model for many different setups to assess the stability and robustness of the inversion model.

Figure 5 (re-plotted to improve legibility)

a **MCMC**



CFM

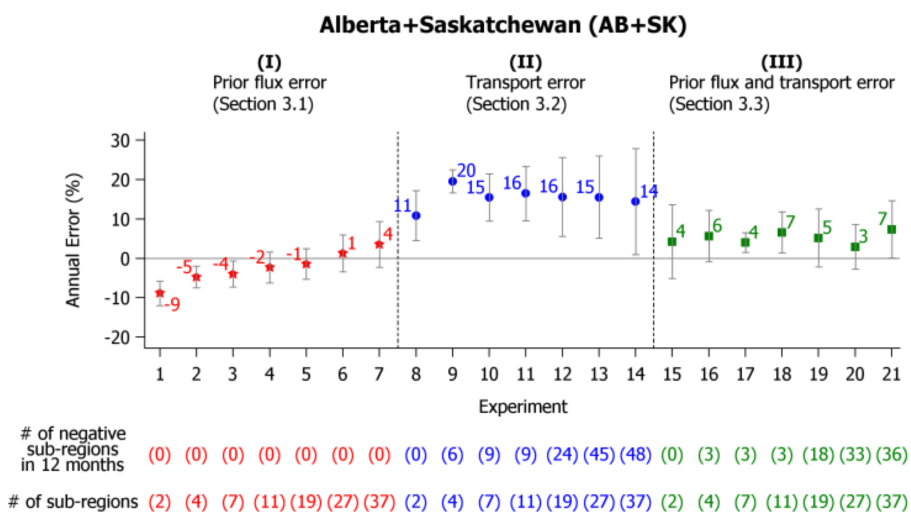
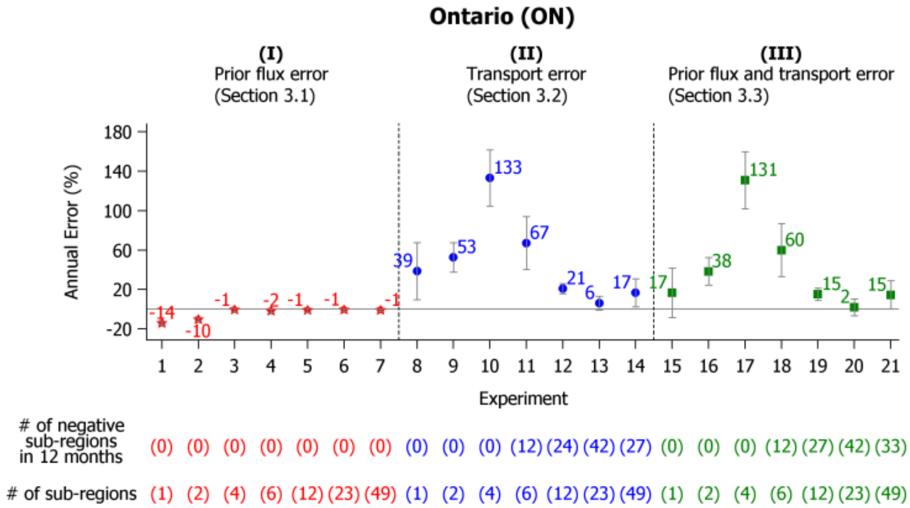
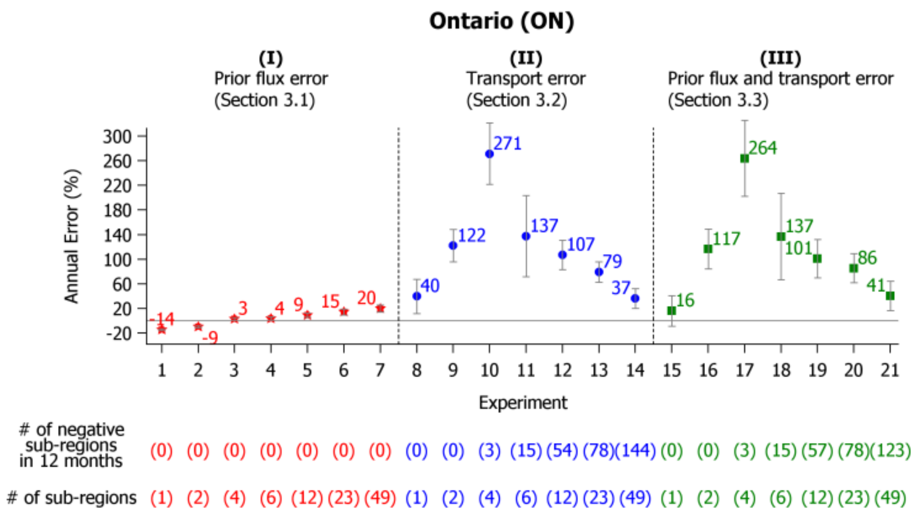


Figure 5 (re-plotted to improve legibility)

b ***MCMC***



CFM



This might also be part of the reason that posterior estimates shown in Figure 7 are closer to the prior when the number of regions is larger.

Authors' response: Actually the posterior estimates are closer to the target (appear to be close the prior) shown in Figure 7 when the number of regions is larger as there are more degrees of freedom to fit the target observations.

The method to calculate the baseline is a bit problematic: Using CT2011 predicted fossil fuel CO2 extracted at locations 5 days before arrival at the observation site does not separate the outside-domain influence from inside-domain influence, where domain means the regional domain of interest.

A better method would be to sample the 3d CO_2 field at the locations when trajectories first leave that regional domain. To assess the impact, at least a map showing a distribution of the locations of the trajectories at the time step 5 days prior to the measurement should be provided (may be included in an appendix).

Authors' response: We have tested with different numbers of days backward and found most of the particles are located quite far away from the inversion domains 5 days ago.

A figure is attached (Figure C1) and will be included in the supplementary section. It shows that the total number of particles in each 0.5ºx0.5º grid 5 days before arriving at each station summing up from January to December 2009. The figure includes the percentages of the particles remaining inside the inversion domain. There are about less than 2 percent of particles inside the AB+SK and ON domains. Most of these particles that remain inside the domain are actually above the mixing height. Therefore, the impact of the surface sources inside of the inversion domain (from more than 5 days ago) should be negligible. In fact, most of the synoptic variability in the concentration time series at the measurement sites has been captured over a 5-day integration period.

Sampling the 3d CO_2 field at the locations when the trajectories first leave the regional domain could miss the footprint contribution from the particles re-entering the regional domain within 5 days. Therefore, we opted for capturing the full 5-day footprint and sampled the 3d CO_2 field at the end of the 5-day simulation period.

Flux error is referred to in the manuscript as the difference between the posterior flux and the target flux (true flux). However, in inverse modeling, usually the statistical uncertainty in the posterior flux estimate is used as an estimate of the expected error in the retrieved flux. In a synthetic experiment, the actual difference between retrieved and true fluxes can be regarded as a realization of this posterior uncertainty. Note that the flux error as referred to in this manuscript is thus expected to be within the 1-sigma uncertainty range for 68% of the cases, i.e. 32% on average are expected outside the range. I suggest also assessing the statistical posterior uncertainty, and including these as error bars in the respective figures.

Authors' response: The statistical uncertainty has been shown as the error bars in Figure 5 and as the statistical distribution of the posterior estimates in Figure 7. The error bars can be seen more clearly in the revised version (see below) of Figure 5 in which the scales of the y-axes have been changed to improve readability. The number of negative sub-regions for the corresponding number of sub-regions is shown on the 2nd row from the bottom in the figure. These explanations will be added to the manuscript revision.

The appendix appears as another version of the explanation of the simulations, rather than only providing information that is additional to the main manuscript content. For example, eq. A1 and A2 of the appendix are identical to eq. 1 and eq. A2 of the main text.

Authors' response: eq. 1 and eq. 2 will be moved to the Appendix.

Contradicting description of the MCMC method: In line 209 it is mentioned that the MCMC method is applied without prior constraint (no regularization), then in line 225 it is mentioned that assumed distributions of lambda_prior are used, which indicates that a prior constraint is used.

Authors' response: In CFM, the posterior fluxes will be forced (regularized) back to the prior if the model-data mismatch uncertainty is much larger than the uncertainty of the prior flux. To avoid ambiguity, the word "regularization" will be removed. It will be clarified that the MCMC method assumes 'λ_p follows normal distribution with a mean of 1 and a variance of 1 for $(\sigma_{prior})^2$, which corresponds to a 100% allowable error'. This serves as an implicit prior constraint on λ_p, but there is no explicit prior constraint in the MCMC optimization (Equation 1) corresponding to the regularization term Equation 4 of CFM.

Detailed comments:

L45: "different atmospheric transports" -> "different atmospheric transport models"

Authors' response: Corrected.

L75: "CarbonTracker fossil fuel CO2" here a reference should be given

Authors' response: A reference has been included.

L83: add "Also" at the beginning of the sentence starting with "Other . . ."

Authors' response: Added.

L94-95: This is not a typical use of estimation error and uncertainty. If these terms are to be used to refer to synthetic and real data inversion, this should at least be made very clear. However I would not recommend using the terms that way.

Authors' response: We will replace the confusing sentence 'The sensitivity of the estimation error (when the truth is known in synthetic experiments) and uncertainty (when the truth is not known in reality) needs *to be closely examined in any inversion setup' with: 'Using synthetic data experiments to examine the sensitivity of inversion results to the different model components (prior fluxes, model transport, observations, etc.) could lead to better understanding of the inversion estimated uncertainties.'*

L181: Note that CFM approaches can also involve simulations, at least when the number of unknowns is large (e.g. pixel-based inversions).

Authors' response: The sentence 'In addition to the more common analytical-based CFM approach, we include a simulation-based method for flux estimations, MCMC' will be replaced with 'Two methods are used in this study, one based on the MCMC method and one based on minimizing the Cost Function as in Gerbig et al., 2003; and Lin et al., 2004.'

L189: add "The" before "Inversion"; also check throughout the manuscript for missing articles.

Authors' response: Corrected. We will check throughout the manuscript.

L233: the variance should have units corresponding to the square of the synthetic observations, i.e. if the observations are in ppm (for dry air mole fractions), the variance should have units ppmˆ2.

Authors' response: Added the unit in ppm^2.

L289: Note that Gerbig et al. (2003) used temporal and spatial correlation in the measurement uncertainty related to transport error, thus their "D_epsilon" was not diagonal.

Authors' response: We will change the sentence to 'The error covariance matrices are not known, consequently $\boldsymbol{D}_{\epsilon}$ *and* $\boldsymbol{D}_{prior}$ *are assumed to be diagonal matrices following e.g. Zhao et al., 2009; Stohl et al., 2009.'*

Fig. 5 caption: The numbers next to the symbols and the two rows of numbers in brackets below the x-axis should be mentioned/explained in the figure caption. Also the error bars shown in Fig. 5 (b) should be explained.

Authors' response: Explanations will be included in the figure caption as "The bottom two rows of numbers show the numbers of negative sub-regions in all 12 months for year 2009 and the corresponding numbers of sub-regions used in the inversions."

Why are there no error bars in Fig. 5 (a)?

Authors' response: The error bars were included in Fig. 5(a) but they were relatively narrow compared to Fig. 5(b) due to the same scale of the y-axis. The Fig. 5(a) will be improved by using a different scale.

What exactly is shown as the y-axis, is it the difference between posterior and target (truth) after spatial aggregation to the respective region (AB+SK and ON) and after temporally aggregated from monthly to annual?

Authors' response: Yes. We will add this to the figure caption.

Why then are the error bars based on the standard deviation of the monthly errors, and not on the annual errors?

Authors' response: We have calculated that the annual posterior fluxes with statistical uncertainties do not cover the targets as shown on the right column of Fig. 7. The error bars calculated based on the annual posterior fluxes are even smaller than the standard deviation of the monthly errors.

L384: "large degrees of freedoms" -> "large number of degrees of freedoms"

Authors' response: Corrected.

L396: "Bielgers" -> "Bieglers"

Authors' response: Corrected.

L444: "transport errors are in our experiments are" remove the first "are"

Authors' response: Corrected.

Figure 6: it should be mentioned which transport model is used in the pseudo observations. Either in the caption or in the text near line 460. I assume that CT2011 transport was used, corresponding to the case with prior flux and transport error.

Authors' response: Yes. The synthetic observations were from CT2011 (based on the transport model TM5) and model results in this study were simulated by FLEXPART. We will mention this in the caption also.

L501, also L610-614: 32% of the estimates are expected to not include the truth within the 1-sigma uncertainty range, thus it is not required that all estimates include the truth within their uncertainty range.

Authors' response: Although it is the case that '32% of the estimates are expected to not include the truth within the 1-sigma uncertainty range', Figure 7c shows the differences between the target fluxes and posterior fluxes are all outside the uncertainty range. This suggests the posterior uncertainties are underestimated. We will clarify the statement (L501) from:

'An important feature in Fig. 7 is that the monthly posterior uncertainties (colored bands) could be underestimated as the uncertainties do not always cover the target fluxes, particularly for ON region.'

To:

'An important feature in Fig. 7c is that the posterior errors are systematically larger than the inversion model's monthly posterior uncertainties (colored bands). Thus the posterior uncertainties appear to be underestimated.'

Similarly change: 'Fig. 7' to 'Fig. 7c' in line 610 to be clearer.

L568: The sensitivity experiments should be added to table 3 so that it is clear which station was omitted in which experiment.

Authors' response: We have added Table 3d.

L669: The fact that aggregation error does not play an important role is due to the fact that target fluxes and prior fluxes are very close to each other in terms of spatial pattern. It should be clearly discussed as to how far this difference is expected to really represent differences between prior flux and true flux.

Authors' response: We calculated the source distribution (SD) errors for the two inversion regions using the following equations. The first equation calculates the source distribution (SD) error resulting from the difference of the fossil fuel CO_2 spatial distributions between CT2010 and CT2011. The second equation calculates the error resulting from the difference of the spatial distributions between CT-CH_4 (scaled to represent fossil fuel CO_2 as described in Section 4) and fossil fuel CO_2 CT2011. CT-CH_4 represents the anthropogenic CH_4 fluxes provided from CarbonTracker Methane.

$$\frac{\sum_{grid} |CT2010\text{-}CT2011|}{\sum_{grid} CT2011} \times 100\%$$

$$\frac{\sum_{grid} |CT\text{-}CH_4 - CT2011|}{\sum_{grid} CT2011} \times 100\%$$

Table below shows the source distribution (SD) errors for the two inversion regions investigated in this study. This result shows that even if the source distribution (SD) error (the cause of aggregation error in the inversion) is large (>100% by this measure), it does not play an important role in the inversions as the results shown in Sections 3.1 and 4. Recall that Sections 3.1 and 4 show the results of flux error only cases. The results indicate that it is possible for the inversion to estimate many sub-regions if there is no transport error even with large source distribution error as calculated below.

Inversion region	*Prior flux*	*Target flux*	*SD Error*	*Section*
AB+SK	*fossil fuel CO_2 CT2010*	*fossil fuel CO_2 CT2011*	*28%*	*3.1*
ON	*fossil fuel CO_2 CT2010*	*fossil fuel CO_2 CT2011*	*29%*	*3.1*
AB+SK	*Anthropogenic CH_4 CT-CH_4*	*fossil fuel CO_2 CT2011*	*182%*	*4*

We will include this explanation in the revision.

L693-696: this general statement on the nature of regional flux inversion should be backed up by references. Note that this statement is quite in contradiction to typical regional inversion results (see e.g. Lauvaux et al.2016).

Authors' response: The discussion will be clarified to indicate that the spatial resolution in the inversion model depends on many factors including spatial and temporal resolution of the observations and model transport errors (we can resolve large number of sub-regions in the absence of model transport error). We will also include a discussion of the state of the art inversion model. We will change the paragraph to:

*In ON, number of sub-regions resolvable is ~4. Since the sub-regions are crowded around southern Ontario, they appear to be not fully resolved by the dispersion model. This is another indication that regional inversion cannot go below some spatial limit (maybe ~3°x3° or 5°x5° or larger) in our setup. The important factors appear to be transport error and the 'observational constraint' is from 3 sites with daily (afternoon well mixed condition) observations. For example, a 20% error in the wind (v=10m/s) over 1 day period would result in an error in the footprint location of ~2m/s*1day=173km. For the multi-day model dispersion period, it could potentially propagate transport and dispersion errors over regions of ~3°x3° or 5°x5° (depending on emission distribution and transport). Note that in the absence of transport error (Prior flux error only case), the inversion model can resolve a much larger number of sub-regions.*

In contrast, (state of the art) inversion models operating on different spatiotemporal scales could potentially resolve finer scales. For example, Lauvaux et al.2016 using a denser network of hourly observations combined with additional constraints of correlation in the prior uncertainty seems to be able to estimate pixel level fluxes over smaller urban scale domains.

L698-707: This discussion should include a discussion of pixel-based inversions (solving for spatially resolved fluxes at high resolution but using spatial (or temporal) correlation in the prior uncertainty) as it is state of the art nowadays.

Authors' response: As what we have learned in this study, without the transport error, it appears to be possible to resolve a large number of sub-regions with simple diagonal prior uncertainty specifications. This does not hold true when the transport error is large. The covariance matrices of the model-data mismatch and prior fluxes are not perfectly known in practice. Flux errors and transport errors are folded together to calculate the prior concentrations. Therefore, uncertainties in the posterior will likely arise from the assumptions made about those covariance matrices.

Discussion of pixel-based inversion has been added as detailed above. We believe current pixel-based inversions are similarly sensitive to model transport errors and availability of sufficient observations as discussed here.

Technical note: it would be much easier for reviewers if the captions for figures and tables were not separated from the figures and tables.

Authors' response: Captions would be included immediately below the corresponding figures and tables in the revision.

References:

Lauvaux, T., Miles, N. L. and Deng, A.: High resolution atmospheric inversion of urban CO2 emissions during the dormant season of the Indianapolis Flux Experiment (INFLUX), J. Geophys. Res., 121, doi:10.1002/2015JD024473, 2016.

Tarantola, A.: Inverse problem theory and methods for model parameter estimation, SIAM, Philadelphia, PA, USA, 2005.

Thompson, R. L., Gerbig, C., and Rödenbeck, C.: A Bayesian inversion estimate of N_2O emissions for western and central Europe and the assessment of aggregation errors, Atmos. Chem. Phys., 11, 3443–3458, doi:10.5194/acp-11-3443-2011, 2011.

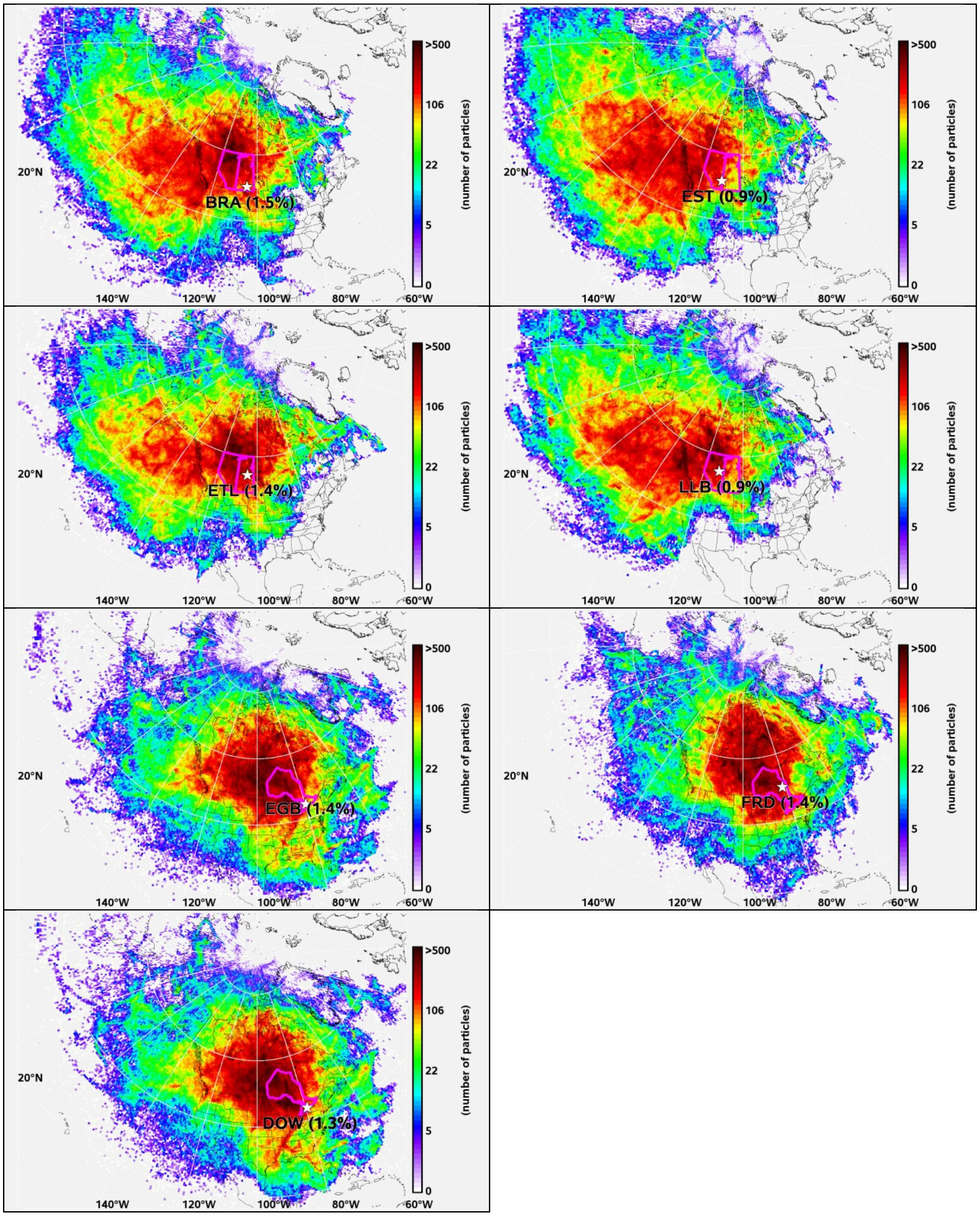


Figure C1. The spatial distributions of the total number of particles in each 0.5°x0.5° grid on the 5th day before arriving at the seven stations summing up from January to December 2009. The number in bracket shows the percentage of particles that is below the mixing height and inside the inversion domain (magenta).