

Response to Reviewer 2, Grant Allen.

Reviewer comments are in italics text; responses in plain text.

Summary:

The paper is a very thorough examination of various model-and-measurement-based approaches to establishing upwind urban-background mole fractions of methane and carbon dioxide for use in Lagrangian (anthropogenic) flux calculations using urban measurement network measurements. It rightly highlights the significant challenges that such methods are typically (if not always) subject to, which include PBL mixing/dilution, biospheric attenuation, significant problems with inventories/priors, global model backgrounds, and model transport error. Some key conclusions of the paper are that an upwind column method appears to provide optimal backgrounds, but with caveats that summertime presents (expected) challenges concerning temporal PBL development and potential negative biases due to biospheric influences. These conclusions are not at all surprising, but they are very useful to others following this work and attempting to conduct urban GHG flux closure. The results are highly specific to the Washington/Baltimore area but the authors are very upfront about that and rightly suggest that conditions need to be assessed on a case-by-case basis. I would also assume that the uncertainties presented here are highly specific to the design of the measurement network/sampling (and this environment) and cannot be taken to be more broadly representative for other areas – and the paper does point this out.

The paper is generally very well written and well-presented. It represents incremental scientific advancement that is very important to others attempting similar important work. I recommend publication after only small potential modifications and perhaps some thought to the specific comments below which may help increase the impact of the paper for others.

We thank the reviewer for the thorough reading, analysis, and suggestions for improving the manuscript.

Specific comments:

I/I think the conclusions section could be clearer than it is. There is no central guidance in the conclusions section on what method(s) – I.e I think the upwind column method? – is/are optimal, and which are highly sub-optimal. Summarising some of what was concluded in the discussions section would be very useful, and arguably more important than discussing the RMSE and bias errors, which are highly specific to this singular environment and case study. Moreover, I wasn't entirely clear after reading the paper whether the UCF might be optimal in all conditions/seasons, or whether other methods may be better under specific conditions?

We agree and we have now added some statements to clarify our recommendations and findings in the conclusion section, first paragraph. We specifically note that the upwind-afternoon background performs well for CO₂ in winter and all year for CH₄. The upwind column background performs better for CO₂ in summer and equally well for CH₄ and winter CO₂. We also point out in the 2nd conclusion paragraph that model-based backgrounds seemed to be

unbiased and perform well (although not as well as the upwind-observation-based) for CO₂, but not at all well for CH₄.

2/ The paper could offer more guidance on what the authors consider might be an optimal network design in future, especially concerning how to place upwind measurement stations. Given that the central conclusion here is that a measurement-based background is optimal (I hope I've read that correctly?), can you go any further here to talk about whether model-based backgrounds should ever be trusted/useful, and/or whether towers with measurements at more/various heights might aid background, especially considering the biospheric problem where sinks are obviously land-based – for example, could a mix of surface sites and towers go some way to addressing the biospheric problem? It seems to me that upwind surface measurements may be more important than anything else here. Residual layers at higher altitudes are of course also important, but I would imagine that after ventilation from the day before, upwind surface measurements and free tropospheric knowledge may be more important than vertically-resolved measurements all through the background PBL?

We thank the reviewer for this suggestion and agree that our study should provide some guidance on network design and additional useful measurements. We have now added an additional paragraph to the Conclusion:

“Our study allows us to give some guidance with regard to background for researchers establishing urban GHG tower networks. First, establishing stations upwind of the area of interest in a configuration that has been shown to capture incoming air from the predominant wind directions is crucial. For our network, a synthetic design study by Mueller et al. (2018) identified locations whose observations best correlated with “true” background. Second, the best-performing background for summertime CO₂ required integrating the upwind tower observations with knowledge of boundary layer height and observations in the free troposphere. We used existing free tropospheric observations from the NOAA/GML aircraft network, which provided measurements every two weeks at best. More frequent observations would have better captured synoptic-scale variability above the PBL and likely improved the Upwind Column background. Some capacity to conduct such airborne measurements should be considered in urban studies. Third, model-based backgrounds should still be considered, especially in cases where they can either be optimized in the urban inversion directly or informed by a nested inversion framework that allows upwind fluxes to be estimated rather than assumed. We did not extend our study to optimizing the modeled backgrounds using the tower observations, but it would be one way to adjust the modeled background and improve performance.”

On the second point, we agree with the reviewer’s thinking that additional observations aloft to help inform the column-based background would be very useful, even for our study where we relied on relatively sparse existing NOAA flask measurements. This information helps construct a better vertical column. We agree with the latter point the reviewer makes, in that we do not believe that additional vertically resolved measurements within the PBL will change the background much, because many air parcels enter the domain above the PBL and currently the FT is probably the least accurate part of the column.

3/ Many of the biospheric problems discussed here exist simply because of the way the goal is defined, which is to deconvolve anthropogenic flux components. However... this problem could be turned around if the goal is about understanding "net" urban GHG emissions. Given the growing agenda on Net Zero carbon, urban biospheres are an integral part of the equation/solution. You could argue that understanding net emissions are an important result in and of itself. It might be interesting to discuss the pros and cons of this. The surrounding biosphere creates a problem for establishing representative upwind backgrounds (fully recognised and discussed), but the urban biosphere also creates a problem for deconvolving anthropogenic urban emissions – I don't think the latter problem is recognised or mentioned in the paper, but there are also good reasons why we might want to know what it is in a "net" sense.

We do not agree with the specific premise of the first statement here, i.e., that the biospheric problems in our study exist simply because of the way the goal is defined. Rather, we agree with the statement later in this paragraph: the biospheric problems discussed here are those that confound the use of an upwind tower observation as a background in summer for CO₂. This issue is caused by biospheric fluxes near the upwind towers, and is not related to urban biospheric fluxes inside the domain. Whether we are solving for a "net" enhancement in the domain or just the anthropogenic enhancements, this is still a problem, as it affects the calculation of the total enhancement, whether that is positive or negative, and independent of the goal of the study in terms of partitioning any net CO₂ enhancement between anthropogenic or biospheric components.

We fully agree that the urban biosphere does indeed create a problem for deconvolving anthropogenic emissions, and the reviewer is correct that we do not discuss this at all in this study. We do discuss the impact it has on the SNR (and perhaps this is the problem the first statement is referring to), because it causes small enhancements in summer, and we have added a sentence in the legend of Fig. 9 to make clear that the enhancements shown are "net", and not fossil-only. We also added a sentence in the discussion of SNR below figure 10 to point out that the SNR would be larger in summer if only FF enhancements were considered, for example if a biosphere model was used to determine fossil enhancements alone (of course, this would introduce error on that correction due to improperly modeled biological fluxes).

As the reviewer points out, we do not really focus on whether the goal is to diagnose emissions from a particular sector or process. However, the reviewer makes a good point that this could be mentioned in the text so we have now added a sentence to point this fact out (last sentence of the discussion now, clarifying that while the background determination is challenging in summer, it is additional to the challenge of separating biospheric from anthropogenic fluxes within the domain).

4/ In the conclusions section, a suggestion is posed on using "ensembles" of the different methods/models as a proxy for error/uncertainty. I would disagree with this. This would not really be an ensemble, as each method/model is systematically entirely different. Ensembles usually represent variations (e.g. monte carlo simulations or parameter space) of a systematically-consistent approach (e.g. perturbing winds, prior uncertainty space etc in one model/method). You wouldn't really get a statistically-relevant ensemble by comparing apples

with pears this way, and it's a very different approach to e.g. comparing outputs of different climate models (which the IPCC would call an ensemble). I'm not even convinced it would give a max/min range of uncertainty that could be useful. I can see why it's attractive to comment on how uncertainty may be better defined but I'm far from convinced that the above would be fit-for-task. I would recommend removing this, or if not, then to discuss the above caveats or suggest alternative guidance on how to establish uncertainty. But this is just a suggestion.

We pondered over how to define an uncertainty on the background for this study – after investigating the various backgrounds and evaluating their performance, we believe that a reader who might be interested in this topic would really just want to know the uncertainty on the background value. This uncertainty could then be incorporated into the model-data mismatch or observational error in an inversion for example.

We agree that the spread of several very different background representations does not represent the true uncertainty; this is unknown. But we do believe that the five realistic representations we have chosen can indeed be used as an ensemble whose spread may be used as a proxy for uncertainty, because they are all different realizations of the same physical system (the background value). We have already evaluated these five as best we can and believe they are unbiased and well-performing according to various metrics, and specifically we do not carry out this exercise for CH₄, where we know that four of the six backgrounds are not realistic. We also have shown that in winter, the magnitude of the model spread (mean over a month) in winter is similar to the error we found by comparing with observations (Fig. 8), giving us confidence that the spread of the five backgrounds is not unrealistically small or large. Thus, we have chosen to retain the comparison of model spread to the enhancement (the SNR in the Discussion), as we still believe this is a useful comparison.

However, in order to address the reviewer's concern and clarify to the reader the above issues and caveats, we have now added text at the beginning of the Discussion to add the caveat about this not being a true probabilistic ensemble, as follows:

“Unfortunately, the true uncertainty of the background is unknown. However, we can observe the differences between the various realistic and plausible representations of the true background that we have constructed for CO₂. We limit this set of plausible backgrounds to the first five backgrounds listed in Table 1 (i.e., omitting the Upwind Aft background, which we found to be biased in summer). Although this set of five background time series does not represent a formal probabilistic ensemble, the spread of these members can still inform us as to the confidence we have in any one of them or their mean.”

Also, as the reviewer suggested, we have removed from the Conclusion the recommendation of using different backgrounds to define an “ensemble” as a proxy for uncertainty.

5/ The uncertainties are presented in concentration space (for background). But nothing is given in terms of how this may manifest as flux error. I guess SNR is a proxy for this to some extent and I guess this paper's scope is on background evaluation, so I don't strongly suggest that flux error should be included, but if there is anything you can say about that, it may be helpful.

We believe that translating background error into flux error is indeed beyond the scope of the work here. We did include the SNR analysis for the purpose of trying to understand the impact of background errors on the enhancement uncertainty which in turn impacts flux errors. We also focus much of the evaluation of the various backgrounds on bias because bias will have the largest impact on flux errors from atmospheric inversions. We have now added the following sentence at the end of the Conclusions:

“We specifically focus our evaluation metrics on bias, as biases will have the largest impact on posteriors from atmospheric flux inversions (as compared with random errors).”

Technical corrections: None – thank you.