Anonymous Referee #3

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The authors thank the reviewer for the careful reading of the manuscript and comments, which we believe have strengthened the paper substantially.

#### Overview:

The paper of Karion et al. reports measurements of atmospheric mole fractions of CO2, CH4 and CO from the CARVE tall tower in central Alaska. The measurements, mainly from the highest of the three avaiable heights, are used to infer fluxes of CO2 and CH4 and a regional budget of CH4. The work forms part of the US Carbon in Arctic Reservoirs Vulnerability Experiment (CARVE) and draws on modelling methods and results from other papers published on the project. The simultaneous measurements of CO, a tracer for combustion emission sources, allow the authors to identify and eliminate events, which are associated with anthropogenic emissions or biomass burning and thus determine the natural fluxes of CO2 and CH4. The WRF-STILT modellling framework, with a priori maps of CO2 and CH4 fluxes, is used to calculate the modelled mole fraction enhancements. Very good agreement is obtained between the modelled and measured CO2 enhancements using fluxes taken from the POLAR-VPRM regression model. The performance for CH4 is poorer, which the authors attribute to the simpler CH4 flux models used with their much coarser spatial resolution and lack of temporal variability. A key finding is the significant biospheric fluxes of both CO2 and CH4 during the autumn and winter, which is supported by the cited paper of Zona et al. Overall, the paper reads well and should be published, after addressing the specific and technical comments below.

# Specific Comments:

## Abstract

I found a slight disconnect in the sentence (page 34873, line 21) "CO2 signals at the tower are larger than predicted, with significant respiration occurring in the fall that is not captured by PolarVPRM" with the "remarkably good agreement with tower observations" in the previous sentence. It seems to undermine the "remarkably good agreement". Perhaps, "However" or similar qualifier is needed at the start of the sentence beginning "CO2 signals".

The wording in the abstract has now been changed to clarify this.

## Modelling

The WRF-STILT modelling uses 500 particles per timestep (p 34880, line 13). There is no information provided in this or the cited paper by Henderson et al. as to why this number of particles is used. I am familiar with the UK NAME Lagrangian particle dispersion model, which appears to use far more particles: 20,000 (Ganesan et al., Atmos. Chem. Phys, 2015) to 33,000 particles (Manning et al., J. Geophys. Res., 2011). What is the reason for the difference? Is it to do with spatial resolution or a

computational issue? What effect does using more or less particles have, e.g., on the uncertainty?

STILT is computationally more intensive than Flexpart or Hysplit, which usually are run with a larger number of particles. I am not personally familiar with UK NAME but I imagine that it is also less computationally intensive than STILT. STILT has been run with 500 particles in this and other studies, and for the model runs presented here, there was no sensitivity analysis on the particle number performed. There have been two previous studies that have shown that 500 is generally a sufficient number of particles for this kind of application. Hegarty et al. (2013) tested STILT with 500 and with 5000 particles, and found little difference between the two cases in replicating a dispersion experiment: Hegarty, J. D., R. R. Draxler, A. F. Stein, J. Brioude, M. Mountain, J. Eluszkiewicz, T. Nehrkorn, F. Ngan, and A. E. Andrews (2013), Evaluation of Lagrangian Particle Dispersion Models with measurements from controlled tracer releases, *J. Appl. Meteor. Clim., 52*, 2623-2637.

In addition, Gerbig et al. (2003) (see section 3.1) also performed a sensitivity test using STILT with different numbers of particles between 50 and 1000 to estimate the error due to the stochastic nature of the model; at 100 particles it was estimated to be 13% and random (i.e. with no correlation between different receptors).

Gerbig, C., J. C. Lin, S. C. Wofsy, B. C. Daube, A. E. Andrews, B. B. Stephens, P. S. Bakwin, and C. A. Grainger, Toward constraining regional-scale fluxes of CO2 with atmospheric observations over a continent: 2. Analysis of COBRA data using a receptor-oriented framework, J. Geophys. Res., 108(D24), 4757, doi:10.1029/2003JD003770, 2003.

## Local source mixing

I did not altogether find the discussion on the influence of (very) local sources convincing (p.34886, lines 22-25). For CH4, the authors argue that the measurements from the highest level of the CARVE tower are decoupled from the ground "despite its low height a.g.l., the tower (my insertion) is elevated above the surrounding area and likely is not affected by very local CH4 sources, such as wetlands". In the very next sentence on CO2, the tower "is surrounded by trees and other vegetation" and these cause a larger CO2 cycle. These statements seem to contradict. If I have misunderstood this, the text needs to be rewritten.

We have now added text in the site description referring to the land cover immediately surrounding the tower, which is mostly deciduous and evergreen forest, with wetland regions  $\sim\!20\text{--}30$  km away. The nearby forest vegetation is likely to be a source/sink for CO2 but not a substantial source of large methane emissions. We have also clarified the text in those lines to explain that the night-time CH4 signal would be trapped in a shallow layer near the wetlands which are at lower elevations and farther away.

# CH4 flux models

The CH4 models are denoted as "uniform land-based flux" and "elevation-based". The cited paper of Chang et al. (2014) provides information on the "elevation model" and its use of four ecosystem/ land cover categories: Highlands (plateaus and uplands); Lowlands (plains, lowlands, and flats); the North Slope (Arctic coastal plain and Arctic foothills); and Mountains (ranges and mountains). It was not obvious from Chang et al what the uniform land-based model was (constant in time and spatially across these categories?). The elevation model gives a marginally better performance, which is to be expected as it does represent, to a certain extent, where wetlands and associated methane emissions are located. More information is needed in the present paper on these models.

We have tried to clarify now that we are using an elevation map from NGDC as a very simple spatial representation of fluxes, and not the ecosystem categories from Chang et al. We do use the uniform flux model, as do Chang et al., which assumes a constant methane flux over all land regions, varying only monthly (with our monthly scaling to our data). We also scaled the elevation model similarly, only on a monthly basis.

## Background concentrations

The determination of the background is a key factor in the analysis. With the focus of the CARVE project on the carbon-cycle and natural fluxes of CO2 and CH4 in Alaska, I can see why a background based on clean air masses from the Pacific was chosen. However, as also noted by Referee 1, this impacts on the data capture, especially in the winter when the winds are predominantly from the East (Fig 4). Given that the uncertainty in the background concentration is the major term, I also endorse referee 1's comment about the robustness of the flux estimates for methane, especially in the autumn and winter and the conclusion about the significance of the autumn/winter fluxes for the annual CH4 budget.

In response to this and the other reviewers' comments, we have conducted a sensitivity analysis to some of the choices made in selecting the background conditions for each observation. We refer to our response to Reviewer 2 for some figures with the different flux estimates and more discussion. We agree with the reviewer on the need to emphasize the large uncertainty of any conclusions related to the wintertime analysis. We have now changed the language in section 4.6 and the Conclusion to emphasize the high uncertainty associated with the wintertime emissions and point to the need for more observations on late fall and wintertime methane fluxes.

#### Context

This paper forms part of a series of papers on the CARVE project and the reader is referred to these. I note that the main project description paper (Miller et al., 2015) is in preparation. The introduction clearly refers to the carbon-cycle but this context

becomes lost later in the paper. As also noted by referee 1, it would be useful to be more explicit about the CO2 and CH4 sources earlier (and which are relevant to this study). This would then explain why anthropogenic sources and biomass burning were not of interest (but could be) and hence excluded. The leakage of CH4 from oil and gas facilities is currently very topical.

We now give some more background on the known sources of CH4 and CO2 in the arctic in the introduction ( $2^{nd}$  paragraph) with some associated references.

### *Technical comments:*

Throughout the paper, R2 is said to be the correlation coefficient. Formally, it is the coefficient of determination and not the (Pearson product-moment) correlation coefficient, which is R (see, for example,

http://stattrek.com/statistics/dictionary.aspx? definition=coefficient\_of\_determination. This occurs on p. 34892, line 12 and also in Table 1 and Figures 7 and 8.

This error has been fixed.

There are a number of typographical other comments:

- There is no reference to Figure 4, which presumably should be in Section 4.2.
- p. 34874, lines 2, 11, 14: Check the date order of citations (e.g., should be Schuur et al., 2008, 2009, 2015)
- p. 34874, line 3: Remove "in" from "focused on in its ...."
- p. 34876, line 9: Insert comma after "2011" in "October 2011 17 km north of Fairbanks, AK,"
- p. 34876, line 16: remove "out" from "to change out flask packages". This also occurs in the Acknowledgements (page 34894, line 21)
- p. 34882, line 11: Longitude in "tagged with the mole fraction from the Pacific boundary curtain at their exit latitude, longitude, and time" should be "altitude"
- p. 34890, line 26: Insert "in the" in "any improvement (in the) correlations"
- p. 34892, line 1: Suggest rephrasing to "not only low-lying wetlands and forests, but also extensive upland and mountain regions"
- p. 34892, line 8: Insert "in origin" after "biogenic"
- p. 34892, line 22: Insert "results" after "The model"
- p. 34892, line 23: Replace "that repeated all three years" with "that was repeated in all three years"
- p. 34892, line 27: Insert as indicated "to have fluxes from interior Alaska in its observation footprint" or similar
- p. 34893, line 20: Insert as indicated "fluxes in this region are likely to be highly heterogeneous"

We thank the reviewer for the close reading and finding these errors - they have been corrected in the text.