

Anonymous Referee #1

Received and published: 13 January 2016

We thank the reviewer for the thoughtful comments and careful reading of our manuscript. We address the points below. The reviewer's comments are italicized and the responses are in plain text.

This is a very well written and relevant study that should be published in ACP. Both the measurement and modelling approaches are appropriate and well described, and the results are presented in a (mostly) convincing way.

I have only a few points which I would like the authors to consider, separated into main and minor points.

Main points:

1. The local surroundings of the site should be described in more detail, e.g. by complementing Figure 1 with another zoom onto the site showing both topography and land use in an area of say 50-100 km around the tower. Figure 4, for example, resents wind roses for two seasons but it is unclear how strongly these winds might be influenced by the local topography. Some short description of the site is given later in the results section (p 16, lines 21-24), but this kind of information should be presented earlier in Section 2.

We have now replaced the right panel of the original figure with a more zoomed-in version, which is 44.6 km high (0.4 degrees latitude) and 47.5 km wide (1 degree longitude), and has a different elevation scale than the left panel. This shows the immediate vicinity of the tower more clearly than the previous version. The land use for the area in the second figure is mostly deciduous and evergreen forest, with some woody wetlands and urban (Fairbanks), which is now mentioned in the text.

2. A "Pacific boundary" is used as background for the measurements which only works for air masses advected from the west. Data with more than 25% of particles originating from a position east of 160° have been discarded. Since winds are primarily from the east between October and April (wind rose in Figure 4), much of the data in this period has to be excluded from the analysis when applying this filter. This is clearly not ideal, especially given the fact that (according to Fig 1a) the site has significant sensitivity to the eastern parts of Alaska. How does the average footprint change as compared to Fig. 1a when this filter is applied? Is the eastern part of Alaska still covered? Is this part not relevant for methane fluxes?

The reviewer makes a good point here, in that the average influence shown in Fig. 1a is for all the observations at the tower. We now show the new influence contours for both the full data set (as before) and the filtered data set on the figure. As the reviewer predicted, it shows that there is less influence over eastern Alaska and Canada once the filters are applied.

Is there not better alternative than the “Pacific boundary” that would allow preserving more data during wintertime? Section 3.3. should mention what fraction of data has to be discarded due to this procedure. The consequences of this choice of background are not very clear to me. As stated on P18 line 12, “the choice of background is crucial to any analysis of the measurements (of CH₄)”. This background appears to be only rarely/poorly defined in winter but at the same time the conclusion is drawn that significant fluxes of methane persist through fall and winter. How robust is this conclusion given the uncertainties in the wintertime background?

We have now conducted a sensitivity analysis of the background calculation on the CH₄ flux estimates in response to the comments from this and other reviewers. Please see the response to Reviewer 2 for figures showing the effect of the background filter. We discuss the results of the analysis in the text in Section 3.3, and we also note that 50% of the observations are eliminated because of this background filter. Although this is a large percentage of the observations, we believe it is a necessary choice given that this method of calculating the background at least for this subset of observations is the best, based on its agreement with the observations; it captures synoptic scale variability in the observations which would otherwise be interpreted as a regional signal. We also agree that given the few observations retained in winter, we have added language to Section 4.6 to emphasize the large uncertainty in the winter fluxes and emphasize the need for further study in winter.

Furthermore, it is not always entirely clear which analysis is based on the filtered or the full data set. Section 4.1 mentions explicitly that no filtering is applied, but what about Section 4.2? Figure 5 presents measured time series for the whole observation period together with background values derived from the “Pacific background”. Obviously, background values are only shown where possible (i.e. filtered), whereas observations represent the complete time series. This should be stated explicitly in the caption. From this figure it actually appears that even in winter there are only few gaps in the background, which seems to be incompatible with Figure 4 showing that winds between October and April were mainly from the east and with the statement on P13 - L23 that “in many winter months, fewer than 6 days of observations remained after the data filtering.

We have clarified this in section 4.2, that our analysis uses all the data, as we are characterizing the site itself. The filters were only applied when enhancements above background were needed. We also have added clarification in the Figure 5 caption. We changed the background to dots instead of a line, which shows the gaps more clearly.

3. The manuscript never spells out clearly what sources of methane are expected in Alaska. The first time the reader learns about the potential importance of wetlands as a CH₄ source is Section 4.1. Up to that point it remains pretty much unclear what sources of methane are expected, and therefore the motivation for producing an elevation-based flux map as described in Section 3.5 remains unclear.

There would certainly be better options than surface elevation to describe watershed hydrology (if that was the purpose), but there are also data available for wetland extent in Alaska (see Whitcomb et al., C. J. Remote Sensing, doi:10.5589/m08-080, 2009) that could serve as a proxy for CH₄ source areas. What about wild animals? What about oil, gas and coal mining? Alaska's economy seems to be dominated by the oil and natural gas industry (<http://alaska.gov/kids/learn/economy.htm>)? Fairbanks is mentioned as a potential source of anthropogenic emissions, but what about other CH₄ emissions from fossil fuel extraction in Alaska? A recent study has indicated that high Arctic soils may represent a net sink of CH₄ (Jørgensen et al., Nature Geoscience, 2015, doi:10.1038/ngeo2305). How does that relate to the results presented here? As opposed to CO₂, the modelling of CH₄ was much less successful, suggesting that the sources are not well represented by the two flux maps and/or that temporal variability of CH₄ emissions is high. This poor understanding of the CH₄ fluxes is briefly mentioned at the end of Section 4.5 but should also be emphasized in the conclusions.

We have now expanded a paragraph describing the site in the introduction. We have added background information that we were missing before in this section, regarding the expected contribution to the methane fluxes from different sources, including wetlands (and now referring to previous work illustrating late fall emissions), and wild animals and oil and gas exploration. We don't have a way to partition total methane observed at the tower between these different sources. Animals and fossil fuel exploration are not thought to be a large factor in total Alaskan methane emissions, based on inventories and previous studies (Bruhwiler et al., Kirschke et al.). Geologic seepage is a source as well (e.g. Walter et al.) (all the above now cited in the text). The soil sinks estimated in Jørgensen et al are small in magnitude relative to the fluxes shown here, but are now mentioned as a possible additional factor in the text. We also now mention in the conclusion that the spatial and temporal representation of CH₄ fluxes is likely incorrect based on poor agreement with the hourly observations.

4. Autumn bursts of CH₄ and CO₂ fluxes coinciding with soil freezing in the high-Arctic Tundra have been described by Mastepanov, M. et al. Large tundra methane burst during onset of freezing. Nature 456, 628–630 (2008) and Mastepanov, et al. Revisiting factors controlling methane emissions from high-Arctic tundra, Biogeosciences, 10, 5139-5158, doi:10.5194/bg-10-5139-2013 (2013). It would be good to place the results in context with these studies.

We have added references to this literature now in 4.6, as our study supports these observations.

Minor points:

- P4, L4: "focused on in its" -> "focused on its"
- P8, L20: What does "drift-corrected" mean? Is this a drift of the signal offset or of the span?

These two changes have been made (the drift is the offset).

- - P9, L27: Are the STILT sensitivities in terms of dry air mole fractions?
Yes.

- - Section 3.2: The three nested domains of WRF are described, but the simulation domain of STILT remains unclear. Is STILT only simulated in the inner domain, or is it run in a nested configuration as well (as is possible e.g. with FLEXPART)?

STILT is run after WRF fields are generated, and does not require a grid on its own. It can run over the entire WRF domain, using the WRF fields available at the grid point of the particle location. After particle trajectories are calculated, the footprints are gridded in post-processing, which can occur at whatever resolution the user requires. For this work, we used a relatively coarse half-degree grid over the entire WRF domain. We have added a sentence in section 3.2 to clarify.

- - P12, line19-20: I didn't understand what "the RMS residuals of the boundary curtain" are.

This is now explained further in the text. They are the residuals that are computed when the boundary curtain is calculated from measurements. A smoothing function is applied and the residuals are the residuals between the data and the fit.

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- - P13, L11: What is the source of the NOAA NGDC elevation data?

The caption of Figure 1 now includes more detail and a reference.

- - P15, L19: "diurnal cycles of the CO₂" -> "diurnal cycles of CO₂"
- - P19, L23: Probably it would be clearer to say "more negative" instead of "lower".
- - P21, L6: The lack of correlation may not only be due to a poor spatial representation but could also be due to temporal variation of the fluxes.
- - P22, L14: "also have" -> "also has"
- - P23, L23: As mentioned above, it is quite likely that the soils in some regions act as sinks rather than as sources.

These changes have been made.

- - Figure 1: Since Figure 3 shows the influence of different regions, it would be very useful to include in Figure 1 the borders of these regions (at least between Lower Alaska and the North Slope and Canada).

We believe the description in the text is sufficient and the figure would become too cluttered, so we have chosen not to include this border in the map.

Anonymous Referee #2

Received and published: 18 January 2016

C11683 ACPD 15, C11683–C11687, 2016

The authors present an analysis of CO₂ and CH₄ fluxes using three years of data collected from the CARVE tower in interior Alaska. The manuscript is well-written and figures are easy to read. The measurements are of high-quality and made using established methods. There are some methodological issues with the analysis that need to be addressed but if these comments can be resolved, the paper should be published in ACP.

Major comments:

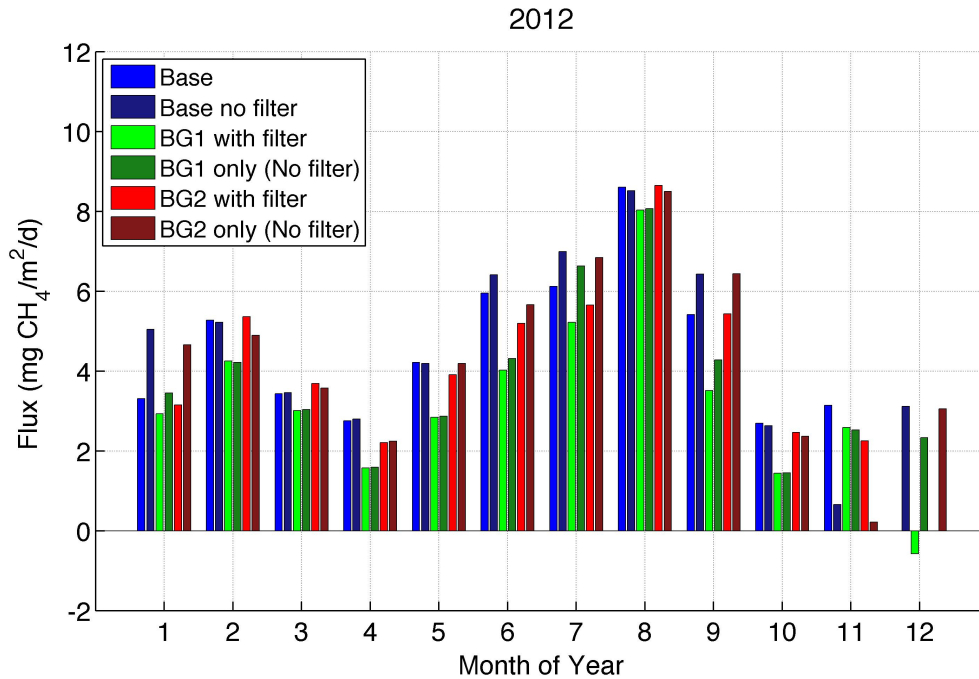
- 1. The method used for the background identification has some issues. Firstly, the authors use a Pacific curtain as the background for calculating enhancements in the measurements. This only applies to air masses that are transported from the West. The authors first discard the subset of particles that did not exit from the West or exited from too far North-West and allow up to 25% of particles to be discarded in this way. Can it be clarified that both the effect on the surface influence as well as on the background are removed in this way? This would lead to up to a 25% error in the analysis, as of course the measurements would still be sensitive to these directions. Secondly, as the authors correctly discuss that the choice of background is critical (especially for CH₄), what is the potential impact of these choices on the results? Can a sensitivity analysis be done? Can the authors use a better method for identifying the background, in the absence of having an additional tower that samples the background (i.e. using model simulations for other directions)?*

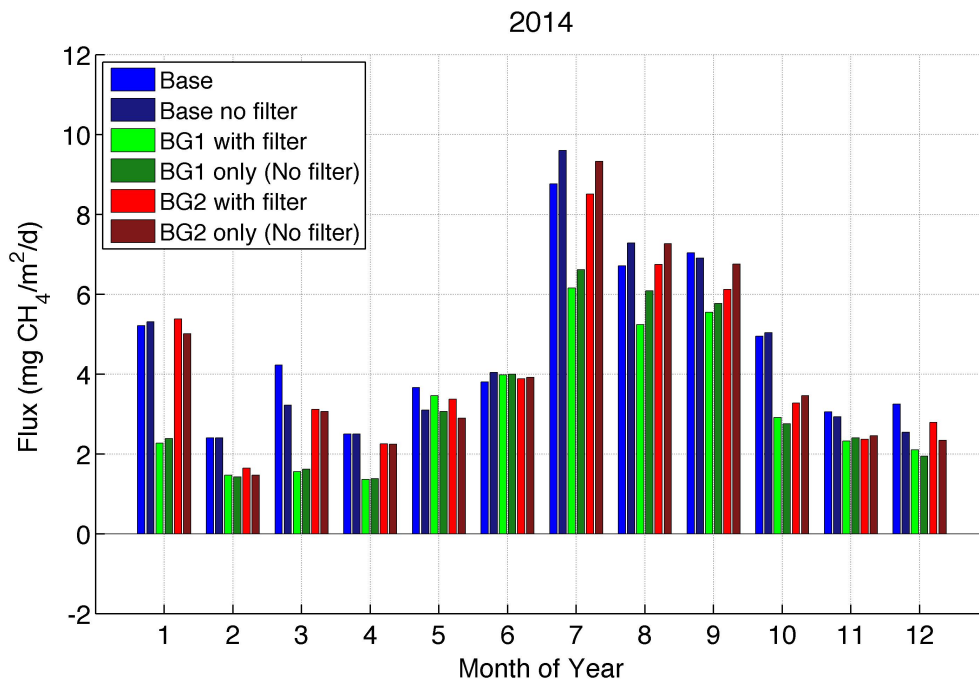
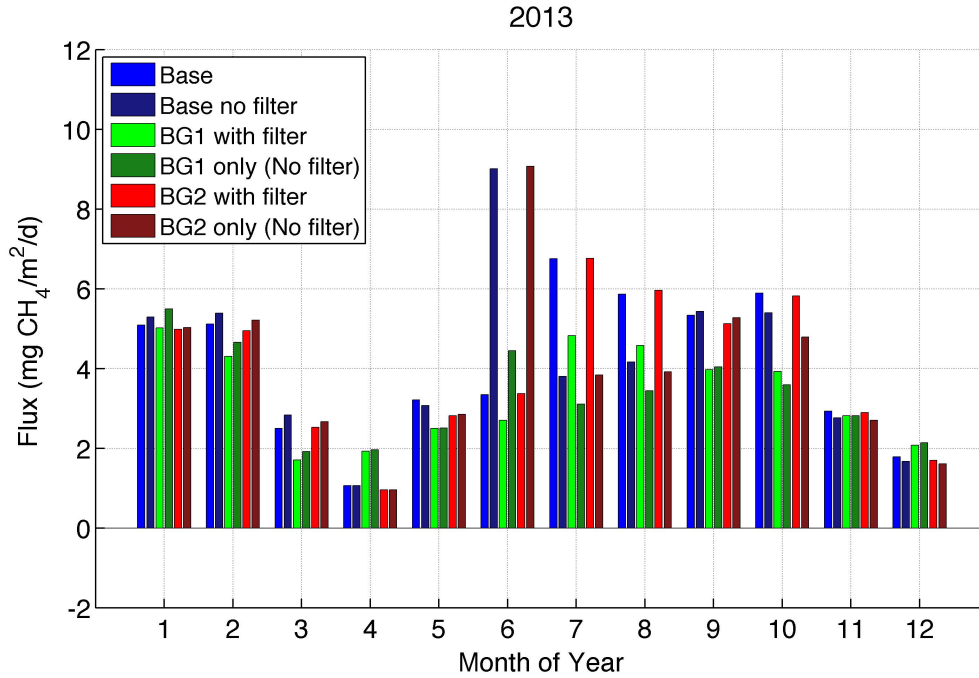
We now clarify in the text that only the background calculation is affected by discarding 25% of the particles. This has no effect on the footprint itself in our analysis, only in the background mole fraction assigned to that footprint. The reviewer is correct in then noting that hours with < 25% of particles discarded would have an additional error in the analysis, although the magnitude is not 25% - it is the difference between what the real background concentration would be if those 25% of particles were included (and their initial concentrations were known) and the background concentration we calculate using the other 75% of particles. As multiple reviewers suggested, we tested the effect on our final flux estimates by calculating two additional possible backgrounds as a sensitivity analysis:

- a) BG1: Removing the background filter entirely: retaining all particles and if they do not exit to the West then tagging them with the background curtain mole fraction at their latitude, altitude, and time at their origin 10 days prior to the observation. No hours were removed from the analysis due to air masses beginning in the East.
- b) BG2: Retaining particles that do not exit to the West in the calculation, but still eliminating the hourly observation from the analysis entirely if less than

75% of particles exit east of 160W and below 4000 masl (same filter as original setup).

To further examine our filtering choices, we also examined the effect of the choice of eliminating hours with high variability, large vertical gradients, or CO enhancements. Each of the above background (BG) scenarios was propagated to a flux estimate calculated both including and not including this additional filter. Six scenarios were investigated in total and the results for each year shown below in the figures. The bars for the “base case” (blue) represent the final flux values shown in the manuscript. The green bars represent background condition (a) above (BG1), and the red represent the results for background condition (b) (BG2). For each color, the darker shade represents the results if the additional filter is not applied. Thus, the dark green bars represent the unique case where there is no added filter at all – all existing observations are included and there is no filter for either the particles not entering from the West or for any variability, lack of vertical mixing, or biomass burning/pollution.





Here we also report the effect of both the background choice and the filter on the number of observations retained in the annual analysis.

Over all 3 years, 50% of hours were removed due to the combination of all filters; most were eliminated due to the background restriction. We understand that these

are large numbers of points to be filtered, but we have chosen to filter them out because a proper calculation cannot be made with those observations.

Our analysis of these alternative scenarios showed that the CH₄ fluxes from the different scenarios could be higher or lower than our base case, with the mean difference over all months and scenarios of 9-20%. Unlike the other scenarios, the case of BG1 (green), where there was no removal of hourly observations or particles based on their exit longitude, almost always resulted in lower CH₄ fluxes (9%-30% lower depending on the year). We found that the footprints whose particles do not enter from the West within 10 days of the observations are stronger, meaning the observations have a larger surface influence. Enhancements in CH₄ are not significantly greater, so that the flux estimate needed to achieve those enhancements is smaller. However, without a proper background calculation during these times, it is difficult to say whether the associated enhancements are correct. The differences remain within our 1-sigma estimate of background error. The text in Section 3.3 and 3.5 addresses this sensitivity analysis now.

2. In the absence of being able to do a full quantitative study with an inverse modeling analysis and appropriate treatment of boundary conditions, the authors need to more explicitly state that the quantitative results will be subject to larger uncertainties and biases (the qualitative results presented have more weight than the fluxes and uncertainties presented).

We agree, and have now added sentences in Sections 4.6 in the text emphasizing that our uncertainty analysis only covers one of many components of the total uncertainty on the flux. We have also emphasized in our abstract and conclusion the qualitative findings of the analysis of the tower data: the small signal, the importance of background in light of this small signal, and the general agreement of a simple CO₂ flux model with the observations.

3. The prior flux maps for CH₄ need more discussion. Why is a uniform map assumed? What sources is this based on?

The spatial distribution of CH₄ sources in Alaska is not well-known. Although maps of wetland extent (and other CH₄ flux maps) do exist, they vary, and, previous studies have shown little or no correlation with observations of fluxes (Chang et al., 2015). Evaluating different models and performing an optimization was outside the scope of this work. Instead, we opted for a simple analysis to interpret the CH₄ enhancements at the tower. A follow-on study is in preparation (Miller, S., A. Michalak, R. Commane, C. E. Miller, J. B. Miller, R. Chang, S. Dinardo, J. Henderson, A. Karion, J. Lindaas, and C. Sweeney, "A multi-year estimate of methane fluxes in Alaska from CARVE atmospheric observations") that focuses on methane only and uses additional observations to perform a geostatistical inverse analysis. We now discuss our choice further in Section 3.5.

4. The method on how CH₄ fluxes were estimated is not clear. Was this not done for CO₂ because it was shown the correlation between modelled and observed CO₂ enhancement lay close to the 1:1 line (i.e. fluxes were correct?). This is a bit confusing because several times, it has also been stated that CO₂ signals are larger than predicted. In Sec 3.5, the CH₄ flux estimation method is described. Observed CH₄ enhancements are first averaged into daily values and then into monthly values. The modelled CH₄ enhancements were first simulated on hourly timescale but then averaged into daily and then monthly. Why is the flux estimation based on monthly values rather than averaging higher resolution fluxes? How is this fit done (there is no information on what method is used to determine the optimal fit)? Further in section 4.5, the fitted monthly fluxes are then used to drive daily enhancement values using the WRF-STILT footprints. The correlation analysis is then based on observed versus modelled daily enhancements (modelled values based on the fluxes derived from the monthly fit). In effect, the authors are fitting fluxes based on the monthly enhancement values, but then state that the daily modelled enhancements do not match the observations well. Can the authors firstly reorganise these two sections together and secondly, explain why the different timescales are used? What is the implication that fluxes derived based on a fit to monthly data does not lead to a good fit on daily timescales? Why is hourly not used (as was done for the CO₂ correlation analysis)? This section needs some more explanation.

We have now added equations to the text in section 3.5 explaining the CH₄ flux estimation method. Scaling fluxes to the monthly mean is equivalent to taking the average of the daily fluxes, but weighting each by the footprint influence on that day (similar to Chang et al, for their vertical profile analysis). The equations should explain the fit as well – there is no fit performed, only a simple multiplicative scaling on a monthly basis. In section 4.5, we show how the results compare with observed higher resolution observations. We have changed these to hourly in the revised paper (from daily), to be consistent with the CO₂ analysis, as this and other reviewers suggested. We have chosen to retain the organization, keeping the methods and the results in separate sections, with references to each in the text.

5. The flux uncertainties that are presented are likely to be too small. The authors are only including an uncertainty due the background term. In almost all inverse modelling analyses, studies have shown that the overwhelming uncertainties are due to model, representation and aggregation errors. In addition, uncertainties should include uncertainties in the fit. While the authors do mention the simple uncertainty analysis that was done, it should be more explicitly stated that the uncertainties presented are likely to be an underestimate of the true uncertainties for these reasons.

We have now added more explicit language and new sentences in the text in section 4.6 that the error bars on the CH₄ flux estimates represent only one of many uncertainty components, and are an underestimate of the true total uncertainties.

Minor comments: Abstract lines 19-22: These two sentences seem inconsistent. Firstly that the model does well in predicting magnitudes and distributions, and then that the signals are larger than predicted.

We agree this was confusing, and have added text to the abstract to indicate that although the model performed very well in general, the exception to this was the underestimate of late fall respiration.

P. 34874 line 4: Typo 'in' P.34876 line 16: Are these supplemental flasks to the in situ? Flasks are not discussed until later.

We fixed the typo and removed the reference to the flasks in this section, so they are only explained later on to remove confusion on this.

P. 34876 line 23: How can the authors be sure that there is no local influence at the site? A tower at 32m could still very much sample local fluxes. More discussion is needed on this.

We have removed the claim that the observations are less likely to be influenced by local sources, because we have not expressly shown this is the case here. We remove the local source influences as much as possible in our analysis by filtering the data for vertical gradients and high variability.

P. 34877 line 11: What are the three heights? Not mentioned until later.

We have included the three heights here now.

P. 34878 line 6: How long is the flushing time?

Three minutes of data is discarded for flushing, although this is conservative for the higher-flow analyzer.

P. 34878 line 9: How often is the water correction performed? Is it instrument specific?

It is instrument-specific (we have now added this information in this section), and was conducted prior to deployment for each. Because we swapped out the units several times, it was not done on a regular schedule, but the timing of the deployments is outlined earlier in this section.

P. 34878 line 17: How do the authors know that the nonlinearity has not changed during the four year period? Has it only been measured once prior to deployment?

We have added text in this section to further explain our calibration method. Two tanks deployed on site have varying mole fractions for each gas, so that a change in the slope or non-linearity would be detected from their residuals.

P. 34880 line 11: What is the size of the domain?

The domain for the WRF-STILT model is 30N to 90N and 180W to 180E (i.e. the entire globe in longitude). This is now inserted in the text.

P. 34880 line 13: Has any sensitivity analysis been done to show that 500 particles is sufficient to statistically represent the footprint? If 1000 particles were used, what is the effect? This is an issue with LPDMs in general that is never adequately discussed.

STILT is computationally more intensive than Flexpart or Hysplit, which usually are run with a larger number of particles. 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.

P. 34880 line 17: This could lead to important differences. Can the authors do a sensitivity test to show the effect of using model ground level?

We have now performed a sensitivity test to quantify the effect of running the model from different altitudes on the CH₄ flux estimate. We analyzed the difference in footprint influence and how it would affect our monthly flux estimates. We found minimal effect between runs at 35, 100, and 300 magl for the filtered observations during the months of March-September of all three years (5-9%). However, in winter months, the fluxes were decreased by 12-17% when using the 100 magl footprints and by 26-32% using the 35magl footprints. We have added a paragraph in Section 3.2 detailing these results. Although this sensitivity analysis points to larger uncertainties in transport during the winter months, we still believe that using the higher altitude is will be more accurate than the lower altitude, given the

limits of the model to appropriately account for the ridge-top location of our site. We now add the mean summer and winter differences for each year between flux estimates calculated at 35 magl and 300 agl to our uncertainty analysis.

P. 34882 line 4: To clarify, are these particles removed from the footprint calculation as well? As discussed above in the major comments, this artificially changes the footprint. For example if a plume were travelling to the south-west corner, such that 75% exit "west" and 25% exit "south", by removing the southerly particles, the plume will have narrowed by 25%, which is significant.

We do not remove the particles from the footprint calculation. The footprint is intact and used as long as at least 75% of the particles entered from the west, only the background assignment is based on fewer particles. (Now clarified in Section 3.3.) However, as we now show in Figure 1, the average footprint is changed by the elimination of footprints where > 25% of particles enter from the East.

P. 34883 line 17: What about the lowest height on the tower? Is it used for anything?
Added sentence in section 3.1 that the lowest level is not used.

P. 34885 line 1: How is this scaling performed? What statistics?
We have clarified this method here by including equations describing the method more fully. The scaling factor was equal to the ratio of observed to modeled monthly CH₄ enhancements, and was different for each month.

P. 34885 line 3: See major comment about uncertainties and being more explicit that these uncertainties are likely too small.

See response to #5 in major comments.

P. 34886 line 7: As the authors state, the magnitudes of amplitudes are difficult to compare as they are very site dependent and depend on sampling height, PBL height etc. This section doesn't add a lot because of all of these additional complexities and any comparison is quite speculative. The section would be better if shortened and condensed to say that the range of measured fluxes is xx and that this is dependent on factors such as: : :

We have shortened this text.

P. 34890 line 20: Following major comment above, this section on the correlation analysis of CH₄ enhancements is confusing.

We have added some text to clarify this section.

P. 34891 line 4: This could also be due to uncertainties in the background estimation
Yes, this is added here now.

Anonymous Referee #3

Received and published: 19 January 2016

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 available 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 modelling 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 ~20-30 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 (2nd 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.

1 **Investigating Alaskan methane and carbon dioxide fluxes**
2 **using measurements from the CARVE tower**

3

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5 **Dinardo⁴, J. M. Henderson⁵, J. Lindaas^{3,**}, J. C. Lin⁶, K. A. Luus^{7,*}, T.**
6 **Newberger^{1,2}, P. Tans², S. C. Wofsy³, S. Wolter^{1,2}, and C. E. Miller⁴**

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22

23 **Abstract**

24 Northern high-latitude carbon sources and sinks, including those resulting from degrading
25 permafrost, are thought to be sensitive to the rapidly warming climate. Because the near-
26 surface atmosphere integrates surface fluxes over large (~500-1000 km) scales, atmospheric
27 monitoring of carbon dioxide (CO₂) and methane (CH₄) mole fractions in the daytime mixed

1 layer is a promising method for detecting change in the carbon cycle throughout boreal
2 Alaska. Here we use CO₂ and CH₄ measurements from a NOAA tower 17 km north of
3 Fairbanks AK, established as part of NASA's Carbon in Arctic Reservoirs Vulnerability
4 Experiment (CARVE), to investigate regional fluxes of CO₂ and CH₄ for 2012-2014. CARVE
5 was designed to use aircraft and surface observations to better understand and quantify the
6 sensitivity of Alaskan carbon fluxes to climate variability. We use high-resolution
7 meteorological fields from the Polar Weather Research and Forecasting (WRF) model
8 coupled with the Stochastic Time-Inverted Lagrangian Transport model (hereafter, WRF-
9 STILT), along with the Polar Vegetation Photosynthesis and Respiration Model
10 (PolarVPRM), to investigate fluxes of CO₂ in boreal Alaska using the tower observations,
11 which are sensitive to large areas of central Alaska. We show that simulated
12 PolarVPRM/WRF-STILT CO₂ mole fractions show remarkably good agreement with tower
13 observations, suggesting that the WRF-STILT model represents the meteorology of the region
14 quite well, and that the PolarVPRM flux magnitudes and spatial distribution are generally
15 consistent with CO₂ mole fractions observed at the CARVE tower. One exception to this
16 good agreement is that during the fall of all three years, PolarVPRM cannot reproduce the
17 observed CO₂ respiration. Using the WRF-STILT model, we find that average CH₄ fluxes in
18 boreal Alaska are somewhat lower than flux estimates by Chang et al. (2014) over all of
19 Alaska for May-September 2012; we also find enhancements appear to persist during some
20 wintertime periods, augmenting those observed during the summer and fall. The possibility of
21 significant fall and winter CO₂ and CH₄ fluxes underscores the need for year-round in-situ
22 observations to quantify changes in boreal Alaskan annual carbon balance.

23

24 1 Introduction

25 The carbon cycle of the high northern latitudes has been the subject of study and research for
26 many decades (Harriss et al., 1992; Oechel et al., 1993; Walter et al., 2007; McGuire et al.,
27 2010; Olefeldt et al., 2013), with scientists and policy makers more recently focused on its
28 impact on global climate. This focus is in part due to the fact that global warming has
29 affected temperatures in the high northern latitudes more significantly than any other region
30 (IPCC, 2013). Higher temperatures could lead to a positive feedback of increased terrestrial
31 emissions of CO₂ and CH₄ (McGuire et al., 2009; O'Connor et al., 2010; Hayes et al., 2014;
32 Schuur et al., 2015), including a possibility of large emissions from thawing Arctic

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1 permafrost. However, the timing and magnitude of such a feedback remain uncertain (Schuur
2 et al., 2008; Schuur et al., 2009; Schuur et al., 2015), and analysis of CH₄ and CO₂
3 measurements from the Global Greenhouse Gas Reference Network (GGGRN;
4 www.esrl.noaa.gov/gmd/ccgg) do not yet show signs of enhanced Arctic to mid-latitude
5 gradients (Bruhwiler et al., 2014; CarbonTracker, 2013). Planned future studies of ecosystems
6 and carbon cycling in Arctic and boreal regions are intended to monitor changes in climate
7 and carbon fluxes (e.g. NASA's Arctic-Boreal Vulnerability Experiment (ABoVE),
8 above.nasa.gov; Next-Generation Ecosystem Experiments (NGEE) Arctic, ngee-
9 arctic.ornl.gov). To this end, quantification of current carbon fluxes from the northern high
10 latitudes, including Alaska, is a crucial piece of any effort to detect changes in the Arctic and
11 boreal carbon cycle.

12 The Carbon in Arctic Reservoirs Vulnerability Experiment (CARVE) was a 5-year NASA
13 Earth Ventures (EV-1) airborne science investigation to quantify atmospheric mole fractions
14 and surface-atmosphere fluxes of CO₂ and CH₄ and correlate these with key surface-state
15 variables for terrestrial ecosystems in Arctic and boreal Alaska. In this region, both CO₂ and
16 CH₄ fluxes are dominated by the terrestrial biosphere; CH₄ fluxes in particular are dominated
17 by wetland emissions (Kirschke et al., 2013; Bruhwiler et al., 2014). Fossil fuel emissions are
18 concentrated in urban areas and in parts of the North Slope associated with oil exploration and
19 production near Prudhoe Bay. Studies have also shown some contribution to CH₄ emissions
20 from ebullition from lakes, a source not usually included in wetland inventories (Walter et al.,
21 2007). CARVE's goal is to bridge critical gaps in our knowledge and understanding of
22 Arctic-boreal ecosystems, linkages between the hydrologic and terrestrial carbon cycles, and
23 the feedbacks from disturbances such as thawing permafrost and fires. The principal
24 components of CARVE were the intensive aircraft campaigns conducted monthly from March
25 to November for four consecutive years (2012 – 2015). The aircraft payload included in-situ
26 sensors measuring CO₂, CH₄, and carbon monoxide (CO) throughout the flights, which are
27 based out of the Fairbanks airport and cover several regions throughout Alaska (Chang et al.,
28 2014). A stationary tower-based GHG measurement site, the CARVE tower (NOAA site
29 code CRV), was established as part of the CARVE project, in order to give year-round
30 context to the intensive aircraft observations. These continuous observations from a single
31 location can verify the temporal pattern of carbon cycle models, while the aircraft
32 observations provide information on spatial accuracy.

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1 Measurements of CO₂ and CH₄ from towers in northern high latitudes have previously been
2 used to analyze emissions and trends in these regions (Sasakawa et al., 2010; Winderlich et
3 al., 2010; Worthy et al., 2015). Concentration measurements from such towers generally have
4 large regions of influence, on scales of hundreds of kilometers, in contrast to direct flux
5 measurements from eddy covariance flux tower sites, which may represent spatial scales
6 closer to tens or hundreds of meters, or chamber measurements that typically represent even
7 smaller (~1-meter) scales. In this sense, the tall tower measurements are able to integrate
8 fluxes that have been shown to be spatially heterogeneous (Olefeldt et al., 2013). Such
9 concentration or mole fraction measurements require interpretation using a model framework
10 to quantify terrestrial fluxes, because they do not measure them directly.

11 One way to infer and assess fluxes from mole fraction observations is to use a Lagrangian
12 particle dispersion model (LPDM) coupled with a meteorological model to determine the
13 influence function, or footprint, of a given observation (Lin et al., 2012). In this study, the
14 WRF-STILT modeling framework has been used to generate footprints for CARVE tower
15 observations. Henderson et al. (2015) provide details of the model configuration and
16 validation of the meteorological simulations. We assess CO₂ fluxes from the land surface of
17 Alaska by convolving surface fluxes from the PolarVPRM (Luus and Lin, 2015) with the
18 footprints and comparing the resulting modeled CO₂ enhancements with tower observations.
19 To infer CH₄ fluxes, we have convolved the footprints with a constant (in space and time)
20 flux model and an elevation-based flux model and scaled the results to monthly mean
21 observed enhancements to estimate monthly average fluxes over a wide region, using similar
22 methods as Chang et al. (2014).

23 In the following sections, we describe the CARVE tower site, its location, and region of
24 influence (Sect. 2). We then describe the measurement methods and the models used to infer
25 CO₂ and CH₄ fluxes (Sect. 3). We present the results in Sect. 4, and conclusions, including
26 future directions, in Sect. 5.

27 **2 Site overview**

28 **2.1 Site location**

29 The CARVE tower site was established in October 2011, 17 km north of Fairbanks, AK,
30 using an existing 32 m tower at the NOAA National Environmental Satellite, Data, and
31 Information Service (NESDIS) facility in Fox, AK (64.986°N, 147.598°W, ground elevation

1 611 m above sea level (asl); Fig. 1). The tower was chosen for its high elevation compared to
2 the immediate surrounding mean ground level and its relatively large region of influence, to
3 provide temporal and spatial context for CARVE aircraft measurements in interior Alaska.
4 The site was also chosen to satisfy logistical requirements, specifically that the site be easily
5 accessible year-round, and that the site be in a location that the CARVE aircraft could sample
6 over or close to during its campaigns without impacting the flight schedules or science
7 mission of each flight. NOAA/NESDIS personnel are stationed in a NESDIS office 5 km
8 from the road-accessible tower, providing technical support and high-speed Internet
9 connectivity throughout the year.

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10 The surrounding land cover (within a 20-km radius, approximately the region shown in Fig.
11 1b) is composed of deciduous and evergreen forest, shrub/scrub, some scattered areas of
12 woody wetlands, mainly south of the Chena River south of Fairbanks, and medium and low-
13 intensity developed land in and immediately around Fairbanks (population 32,000) (2011
14 USGS National Land Cover Database (NLCD) (Homer et al., 2015)). The tower is located on
15 a ridge, and measurements from the tower represent a wide region of interior Alaska,
16 however, as indicated by surface influence fields generated from the WRF-STILT modeling
17 framework (Henderson et al., 2015), which show that the tower's influence region
18 encompasses a substantial part of Alaska (Fig. 1a).

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19 3 Methods

20 3.1 Measurements

21 Three separate measurement systems for trace gases are deployed at the CRV tower site.
22 Programmable flask packages (PFPs) are used to collect air samples from the top level of the
23 tower at 32 m above ground level (agl), daily during the CARVE flight season (April –
24 October) and twice weekly during the remainder of the year (November – March).
25 Additionally, measurements of ¹⁴CH₄ are made from large-volume (~1000 L) whole-air
26 samples collected approximately biweekly, also from the 32 magl level. Lastly, continuous
27 in-situ measurements of CO₂, CH₄, and CO are made by drawing air from three heights (32
28 m, 17 m, and 5 m) through a Picarro G2401-m or G2401 Cavity Ring-Down Spectroscopic
29 (CRDS) analyzer. In addition to the measurements described above, a two-dimensional (2D)
30 sonic anemometer was deployed at the top of the tower and was operational from April 2012

1 through June 2014. Here we describe the continuous CO₂, CH₄ and CO measurements made
2 from October 2011 through the present, focusing on the calendar years 2012-2014.

3 Two different CRDS units have been deployed at the site as part of the CARVE project: SN
4 CFKBDS-2008 (model G2401-m, October 2011 – June 2013 and November 2014 – January
5 2015), and SN CFKADS-2067 (model G2401, June 2013 – October 2014 and January 2015 –
6 present). The only differences between the two units as configured at the site are the flow
7 rates (~550 standard cubic centimeters per minute (scm) for CFKBDS-2008 and ~250 scm
8 for CFKADS-2067) and their precision, defined here as the standard deviation of 30-second
9 averages, in measuring CO (1.3 ppb for CFKBDS-2008 and 4.3 ppb for CFKADS-2067).
10 Analyzer precision for CO₂ and CH₄ is the same for both analyzers (0.03 ppm and 0.2 ppb,
11 respectively).

12 The CRDS analyzer draws air through 0.635 cm (0.25 inch) outer diameter (OD) tubing
13 (Synflex 1300) with three different inlets installed at different heights above ground level:
14 31.7 m (level 3), 17.1 m (level 2), and 4.9 m (level 1). The analyzer primarily draws from the
15 highest level (level 3) for 50 minutes out of every hour, and then draws air for 5 minutes from
16 each of the other levels, operating on an hourly cycle. In our analysis, we use measurements
17 only from the top level, using measurements from level 2 to filter observations with large
18 vertical gradients (section 3.4); level 1 observations are not used. Measurements are discarded
19 for a time equivalent to three flushing volumes of the line (approximately 3 minutes) after a
20 level switch or a switch to or from a calibration tank, to allow each line to flush, because there
21 is no separate flushing of the lines during calibrations. The sample air is not dried, and a water
22 correction to the measurements is made in post-processing. The instrument-specific water
23 correction is based on a laboratory experiment conducted prior to deployment of each
24 analyzer, using methods described in Chen et al. (2013) and Rella et al. (2013). Data is
25 collected via serial communications on a Campbell Scientific CR1000 data logger along with
26 all auxiliary measurements (room temperature, line pressure, tank pressures, sonic
27 anemometer measurements of temperature and 2D winds), and averaged at 30-second
28 increments prior to remote collection via the Internet connection provided by
29 NOAA/NESDIS.

30 Two standard reference gases, calibrated against NOAA standards on the WMO scales for all
31 three gases, are each sampled every 8 hours for 5 minutes. Mole fraction measurements of
32 CO₂, CH₄, and CO are first corrected using a linear fit to either 5 or 6 NOAA reference tanks

1 from a calibration performed in the laboratory prior to each analyzer deployment, and then
2 drift-corrected using the measurements of the two tanks at the site. The average offset
3 (difference between corrected value and the actual tank value) is used for an offset drift
4 correction. The two on-site tanks are at two different mole fractions for each gas (373 and
5 409 ppm CO₂; 1818 and 2087 ppb CH₄; 177 and 284 ppb CO), so that if either the slope of
6 the correction changes, or one tank has significant drift, the measurements would show
7 increasing residuals with time. All measurements are reported here on the WMO scales for
8 each gas (CO₂ X2007, CH₄ X2004, and CO X2004, (Dlugokencky et al., 2005; Zhao and
9 Tans, 2006)). At CRV, the water correction uncertainty is estimated to be 0.1 ppm for CO₂,
10 0.5 ppb for CH₄, and 4 ppb for CO, based on analysis by Chen et al. (2013) for CO and Rella
11 et al. (2013) for CO₂ and CH₄, and is independent of other variables, including water vapor.
12 Comparisons of measurements from whole air samples in PFPs during low-variability periods
13 show differences (median ± 1-sigma) of -0.11±0.44 ppm, 0.8±1.2 ppb, and -1.3±4.5 ppb for
14 CO₂, CH₄, and CO, respectively, over the entire 3-year period. Total uncertainty
15 (reproducibility and comparability to other NOAA network sites) of hourly mole fraction
16 measurements at the site are generally <0.2 ppm, 2 ppb, and 5 ppb for CO₂, CH₄, and CO,
17 respectively (1-sigma), based on comparisons with flasks and residuals of the calibration
18 correction.

19 3.2 Polar WRF – STILT Model

20 The scientific analysis of CARVE atmospheric trace gas measurements is enabled through use
21 of the Stochastic Time-Inverted Lagrangian Transport (STILT) particle dispersion model (Lin
22 et al., 2003) coupled to the Polar variant version 3.5.1 (Wilson et al., 2011) of the Advanced
23 Research version of the Weather Research and Forecasting (WRF-ARW (Skamarock et al.,
24 2008)) numerical weather prediction model. The WRF-STILT modeling framework has been
25 used in many studies to estimate GHG emissions using airborne, surface, and tower-based
26 observations (Kort et al., 2008; Jeong et al., 2013; Chang et al., 2014; Miller et al., 2014;
27 McKain et al., 2015). Atmospheric dispersion in the LPDM is simulated by advecting tracer
28 particles by the three-dimensional gridded wind field from the WRF model, plus a turbulent
29 velocity component represented as a stochastic process (Markov chain) (Lin et al., 2003).
30 Time-averaged mass fluxes and convective mass fluxes from WRF are used in the dispersion
31 calculations (Nehrkorn et al., 2010). For each observation location (i.e. “receptor”), STILT
32 produces a two-dimensional surface influence field called a “footprint” (units of ppm / (μmol

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1 $\text{m}^{-2} \text{s}^{-1}$) that quantifies the influence of upwind surface fluxes on atmospheric concentrations
2 measured at the receptor location. The footprint field is proportional to the number of
3 particles in a surface-influenced volume (defined as the lower half of the planetary boundary
4 layer) and the time spent in that volume (Lin et al., 2003). As utilized in the current study, the
5 footprint can be multiplied by an *a priori* flux field (units of $\mu\text{mol m}^{-2} \text{s}^{-1}$) and integrated over
6 space and time to give the incremental contribution to the mole fraction (units of ppm) as
7 measured at the receptor location. The CARVE Polar WRF configuration consists of a triply
8 nested grid, with the innermost domain covering mainland Alaska on a 3.3-km grid to take
9 advantage of the improved representation at this scale of the underlying topography in this
10 region of significant orography. [The STILT model runs over the entire WRF domain \(all three](#)
11 [grids\); footprints are gridded separately from WRF in post-processing over the whole domain](#)
12 [\(30°N-90°N and 180°E-180°W\)](#). The reader is directed to Henderson et al. (2015) for more
13 detail and validation of the meteorological fields.

14 STILT footprints used for this analysis were generated every 3 hours during local nighttime
15 and hourly during local daytime, for a total of 16 footprints per day, and gridded at $0.5^\circ \times 0.5^\circ$
16 resolution. For each footprint, 500 particles were emitted from the tower location and altitude
17 above sea level and traced backwards in time for 10 days. The altitude above sea level rather
18 than ground level was used for the location of the particle emission because the elevation of
19 the model grid cell containing the tower site was significantly lower than the actual elevation
20 of the site (343 masl vs. 611 masl), despite use of the high-resolution grid (Henderson et al.,
21 2015). To reduce biases induced by differences in actual and modeled topography, we use
22 footprints generated during mid-afternoon hours (1 pm to 6 pm local Alaska standard time
23 (LST), UTC+8) only for our analysis, except where noted specifically. During these hours,
24 the lower atmosphere is generally well-mixed, and the difference between the mole fractions
25 measured at the top level (32 magl) and the middle level (17 magl) average between -0.25 and
26 0.25 ppm for CO_2 and -0.2 and 0.3 ppb for CH_4 (maximum monthly averages for the whole
27 time series), indicating good mixing and only a small influence from nearby sources that
28 would cause a near-surface gradient.

29 We also compared measurements from the top level of the tower to CARVE aircraft
30 measurements made above the tower site, generally during the months of March to October.
31 We compared aircraft measurements of CO_2 and CH_4 that were made below 2000 masl (1389
32 magl) and within 0.2 degrees in latitude and longitude of the tower, between the hours of 1

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1 pm and 6 pm LST. This allowed us to determine how well measurements made from the top
2 level of the tower represent planetary boundary layer (PBL) average mole fractions during
3 those times. Differences between the 29 aircraft observations and tower-based hourly means
4 were -0.6 ± 2.0 ppb CH₄ and 0.3 ± 0.9 ppm CO₂ (mean \pm 1-sigma) during the March-October
5 air campaign period, indicating that the hourly average mole fractions at the tower are
6 generally representative of average mole fractions in the PBL.

7 We expect, based on the measured gradients at the tower and the comparison with aircraft
8 measurements above the tower, that during local mid-afternoon periods the tower
9 measurements closely represent measurements within a well-mixed PBL, and that during
10 those times, the impact of the height difference between the modeled site elevation and the
11 real site elevation is minimized. In our flux analysis, described in the following sections, we
12 also specifically filter out hourly averages during which the absolute value of the mole
13 fraction gradient between 17 and 32 magl levels in CH₄ is larger than 2 ppb.

14 We performed a sensitivity analysis on the transport model by examining footprints generated
15 at 300 magl (the default, or 611 masl) with those generated at 100 magl and 35 magl. We
16 found that the footprint influence from March to September of all three years was increased
17 by a small amount, resulting in estimated CH₄ fluxes (Section 3.5) that were 5-9% lower.
18 The effect was greater in winter months, however, affecting our flux estimates by 12-17%
19 using the 100 magl runs and by 26-32% using the 35 magl runs (again, decreasing the fluxes
20 due to increased surface influence). These differences were calculated only based on
21 observations used in the flux analysis, i.e. filtered for large vertical gradients as described
22 above. Although CH₄ fluxes reported in Section 4.5 were estimated using the footprints from
23 the higher 300 magl altitude, reflecting the true 611 masl altitude of the observations, this
24 sensitivity analysis indicates that uncertainty in modeled transport is greater in winter months.
25 We have added (in quadrature) the mean summer and winter differences for each year
26 between the analysis at 35 and 300 magl to the flux estimates for CH₄ in Section 3.5.

27 **3.3 Calculation of background mole fractions**

28 To compare the mole fraction variability and enhancements at CRV tower to those from the
29 modeling framework, it is necessary to determine the appropriate background mole fractions
30 for both CO₂ and CH₄. We derive background mole fractions using the particle trajectories
31 from the STILT runs and a data-based Pacific basin boundary “curtain” derived from

1 NOAA/ESRL Global Monitoring Division GGGRN measurements using an approach similar
2 to the one described in Jeong et al. (2013) and Miller et al. (2014). Specifically, the boundary
3 curtain is constructed using GGGRN surface and aircraft vertical profile CO₂ and CH₄
4 observations (Sweeney et al. (2015) and www.esrl.noaa.gov/gmd/ccgg/aircraft/) to create a
5 smoothed curtain representing the Pacific boundary. The curtain is a function of time,
6 latitude, and altitude. For each STILT run, the 500 particles are traced back in time until they
7 either exit a box defined by [170°W, 130°W] and [0°N, 75°N] or remain in the box for the
8 full 10-day run. All particles are then tagged with an exit time, longitude, latitude, and
9 altitude. Any particles whose final longitude is east of 160°W with a final latitude between
10 55 and 72°N and altitude below 3000 masl are removed in order to eliminate particles that did
11 not enter Alaska from either the western boundary or from high altitudes within the 10-days
12 of the observation. This filter is necessary because air masses that contain surface influence
13 from Canada or remain in Alaska for more than 10 days would not properly be represented by
14 the Pacific boundary as background. We note that the footprint itself is not changed by this
15 choice, but the particles that do not enter from the West are not used in the background
16 calculation. However, if a given 500-particle run has more than 25% of its particles
17 eliminated due to the above constraints, no background is computed for that hour, and
18 therefore no enhancement is computed either. This choice removes 50% of the hours from the
19 analysis over all three years. If at least 75% of the particles remain, these remaining particles
20 are tagged with the mole fraction from the Pacific boundary curtain at their exit latitude,
21 altitude, and time. The mole fractions for the particles are averaged to derive the background
22 mole fraction for the corresponding tower measurement and WRF-STILT footprint.

23 To understand the sensitivity of the CH₄ analysis to this choice of filtering for influence from
24 the east, we repeated the analysis with two additional background choices: one in which all
25 particles were tagged with a mole fraction, regardless of their origin, but the background was
26 only calculated if 75% or more particles originated in the west; and one for which all particles
27 were tagged and there was no filtering based on the percentage of particles not originating in
28 the west. We found that the difference in the background values was within the one-sigma
29 uncertainty in the background itself, calculated as described below. However, we found that
30 including hourly observations for which the air mass did not enter strictly from the West
31 generally reduced the monthly methane flux estimates by 9-30%; this result is discussed
32 further in Section 3.5.

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1 Uncertainty in the background is determined similarly to Jeong et al. (2013): it is assigned the
2 quadrature sum of the standard error of the mean mole fraction (i.e. the standard deviation of
3 the particle mole fractions divided by the square root of the number of particles used) with the
4 average value of the root mean square (RMS) residuals of the empirical background curtain of
5 the particles. The RMS residuals of the curtain are calculated at every point along the curtain
6 (a function of latitude, altitude and time); they are the residuals of the curve fit that generates
7 the smoothed background curtain and the data that is used to generate the curve. Thus, it is a
8 quantification of uncertainty of the curtain itself. The background uncertainty in this work is
9 dominated by the RMS residuals of the boundary curtain component.

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10 **3.4 CO₂ Flux Model**

11 The CO₂ measurements at the CRV tower were interpreted with the assistance of biospheric
12 CO₂ flux estimates generated by the Polar Vegetation Photosynthesis and Respiration Model
13 (PolarVPRM, (Luus and Lin, 2015)). PolarVPRM captures the strong diurnal and seasonal
14 variability of CO₂ fluxes parsimoniously, according to empirical associations between
15 environmental conditions and eddy covariance measurements of CO₂, and regionally across
16 Alaska (3-hourly, 1/6 x 1/4 degrees latitude x longitude), using data products from the North
17 American Regional Reanalysis (NARR) (Mesinger et al., 2006) and the Moderate Resolution
18 Imaging Spectroradiometer (MODIS). Subnivean and growing season respiration are
19 calculated as functions of NARR soil and air temperature, respectively; snow and growing
20 seasons are differentiated using MODIS snow cover (Riggs and Hall, 2011). Photosynthesis is
21 calculated as a function of NARR air temperature, NARR shortwave radiation, water
22 availability (via MODIS), and vegetation (via the MODIS enhanced vegetation index (Solano
23 et al., 2010)). The CO₂ fluxes from PolarVPRM were convolved with footprints from
24 observations at the tower to derive model-based enhancement above background (ΔCO_2) for
25 the 3-year period from 2012-2014. These modeled CO₂ enhancements were compared to CO₂
26 enhancements observed at CRV.

27 Hourly observations used for CO₂ analysis were restricted to periods between 1 pm and 6 pm
28 LST, to minimize discrepancies between real and modeled boundary layer dynamics. In
29 addition, as described above, samples that had no background determination for more than
30 25% of the released particles were omitted. Additional filters on the data were designed to
31 restrict analysis to periods when the PBL was most likely well mixed, as determined from the
32 vertical gradient in CH₄ mole fractions between the 17 m and 32 m levels; only data for which

1 the CH₄ vertical gradient was less than 2 ppb were retained. Also, only data observations
2 with low temporal variability were retained, determined as having a standard deviation of 30-
3 second measurements in an hour below 7 ppb in CO and 3 ppb in CH₄; this filter was applied
4 to reduce influence from local sources, a concern at this site because of the proximity of
5 Fairbanks. Lastly, biomass-burning (and some large pollution) events were filtered out by
6 removing observations for which the enhancement in CO (relative to the background
7 determined using methods described in Sect. 3.3) exceeded 20 ppb. The combined effect of
8 these filters and the background filter eliminated 56% of the days analyzed – most were
9 removed by the background filter described in Sect. 3.3. The filters described above were
10 used to filter data only for the CO₂ flux-model comparison analysis described in this section
11 with the results in Section 4.4, and for the CH₄ footprint and flux analyses described in
12 Sections 3.5 with results in Sections 4.5 and 4.6.

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13 3.5 CH₄ Flux Estimation

14 In the CH₄ analysis, as for CO₂, only hourly average mole fractions between 1 pm and 6 pm
15 LST were used, with the same filters applied on the observations to limit instances of high
16 variability, large vertical gradients, and biomass burning, as outlined in Sect. 3.4. We also
17 carried out the analysis without the observational filters and did not find any difference in the
18 resulting monthly fluxes within the 1-sigma uncertainty of the background presented here,
19 with the exception of months with significant biomass burning events.

20 Chang et al. (2014) investigated the use of existing CH₄ flux models to interpret observations
21 from CARVE airborne campaigns and found that none of them performed better than the
22 assumption of a uniform flux. Hence, here we use two simple flux spatial distributions to
23 interpret the tower observations and estimate average CH₄ fluxes. The first flux map is a

24 uniform land-based flux (with oceanic flux set to zero, assuming the oceanic CH₄ flux
25 contribution is negligible (Kirschke et al., 2013)) similar to what was used in Chang et al.
26 (2014) to estimate CH₄ fluxes using aircraft observations from the 2012 CARVE campaign.

27 This model assumes a spatially constant flux over all land regions. The second flux map
28 pattern is based on elevation data from NOAA's NGDC,
29 (<http://www.ngdc.noaa.gov/mgg/topo/report/globedocumentationmanual.pdf>) (Fig. 1a). The
30 elevation map was averaged to the same spatial resolution as the footprints (0.5° x 0.5°) and
31 adjusted so that the ocean and elevations higher than 1000 masl were assumed to have zero

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1 CH₄ flux. Elevations between 0 and 1000 masl were scaled linearly from 1 to 0, with areas of
2 zero elevation (including lakes) assigned 1 and 1000 masl assigned 0. Fluxes were assumed to
3 be diurnally constant. A third map based on elevation gradients (in which highly sloped
4 regions had less flux and flatter areas had higher flux) was also tested, but the results were
5 very similar to the elevation-based map, so they are not shown here.

6 Observed CH₄ enhancements relative to the footprint background were averaged over the
7 mid-afternoon hours (1 pm – 6 pm LST) to obtain daily averages. These daily enhancements
8 were then averaged to obtain monthly average ΔCH₄ values throughout each year. However,
9 in many winter months, fewer than 6 days of observations remained after the data filtering;
10 those months were omitted from the analysis. The constant flux map and the elevation-based
11 flux map were convolved with the hourly footprints from the WRF-STILT model to obtain
12 initial values of modeled ΔCH₄. These were then averaged to create daily values (with the
13 same filters as for the observations) and then to monthly values. The monthly flux maps were
14 then scaled to the observations, so that the simulated monthly ΔCH₄ matched the
15 observations. This method is equivalent to taking the mean of the individual daily flux
16 estimates (F_D) weighted by their corresponding footprints (I_D). For the case of unit flux, the
17 daily flux estimate (F_D) is the CH₄ enhancement (ΔCH_4) divided by the footprint influence:

$$18 \quad F_D = \frac{(\Delta\text{CH}_4)_D}{I_D}$$

19 The monthly average flux (F_M), is the average of the daily fluxes weighted by each day's
20 influence, which is equivalent to dividing an average methane enhancement by the average
21 monthly influence, I_M :

$$22 \quad F_M = \frac{\sum_D F_D I_D}{\sum_D I_D} = \frac{\Delta\text{CH}_{4,M}}{I_M}$$

23 Uncertainties on monthly fluxes were determined from the background mole fraction
24 uncertainty and uncertainty based on a sensitivity analysis of influence functions calculated
25 using different heights in the model (Section 3.2). A formal transport uncertainty analysis
26 (e.g., error in PBL depth or wind speed) was not considered, but would likely increase the
27 errors shown here. Monthly background errors (Section 3.3) for CH₄ ranged from 2-7 ppb
28 (average 5 ppb), which was generally of the same order of magnitude as the CH₄

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1 enhancements. Uncertainty on the monthly enhancements was calculated as the average of
2 the uncertainty on the background for each day divided by the square root of the number of
3 independent samples during that month, approximating the standard error of the monthly
4 mean enhancement. The correlation time scale of the background for CH₄ (after a 60-day
5 smoother was subtracted to eliminate the long-term temporal correlations) was approximated
6 at 9 days, consistent with synoptic-scale variability. The number of independent realizations
7 for each month was therefore derived as the number of days in that month divided by 9. This
8 represents a 1-sigma uncertainty estimate; the fractional uncertainty on the monthly CH₄
9 enhancement was then summed in quadrature with the uncertainty based on the altitude

10 sensitivity analysis and propagated to the monthly flux estimates.
11 A sensitivity analysis to the background filter, as described in Section 3.3, showed that the
12 background mole fractions were within the 1-sigma uncertainty when calculated using the two
13 additional background options. We did find, however, that the fluxes were generally 9-30%
14 lower when the observations with the bulk of the air mass not entering from the West were
15 included. We found that in this case, the average footprint influence was greater, likely
16 because the air masses spent more time over land. With no corresponding increase in
17 observed CH₄ enhancements, this led to a lower flux estimate on those days. However, the
18 CH₄ enhancements in this case have high uncertainties because it is very hard to estimate
19 what the background for those days might be.

20 4 Results

21 4.1 Diurnal cycles

22 The diurnal cycles of CO₂ and CH₄ at the tower have been analyzed over the study period,
23 2012-2014. All analysis shown is based on hourly-averaged measurements from the top-most
24 level at 32 magl. Measurements during times when the CO mole fraction exceeded 200 ppb
25 were removed to filter out the effect of large biomass burning events. No other filters were
26 applied on the data in this portion of the analysis. The diurnal cycle of CO₂ shows an
27 amplitude (maximum – minimum CO₂) of 10 ppm in July, with a wintertime (November-
28 April) magnitude of approximately 2 ppm (Fig. 2, top panel), with similar patterns each year.
29 CH₄ diurnal cycle amplitudes also show a maximum in summer (in either July or August,
30 depending on the year) between 20 and 30 ppb. The wintertime diurnal cycle of CH₄, driven
31 by boundary layer dynamics, shows an average amplitude of 10 ppb (Fig. 2, lower panel).

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1 Shaded areas in Fig. 2 indicate the standard deviation of that month's average over all days in
2 the 3-year period, indicating significant variability in the amplitudes for both gases, and
3 especially for CH₄, where the amplitude variability (one-sigma) ranges from zero to 45 ppb.
4 The average amplitudes of the CH₄ and CO₂ diurnal cycles at the CARVE tower are
5 significantly smaller than those that have been reported at other Arctic and boreal
6 measurement sites. Worthy et al. (2015) compare diurnal cycle amplitudes of CH₄ at various
7 Arctic tower sites throughout Canada and North America, finding that summertime diurnal
8 CH₄ amplitudes at all the Arctic and boreal Canadian sites are significantly larger than at
9 CRV tower. Sasakawa et al. (2010; 2013) report larger CH₄ and CO₂ diurnal cycle
10 magnitudes as well from a network of Siberian tower sites. Winderlich et al. (2010) also
11 report similarly large diurnal cycle amplitudes in CH₄ (~200 ppb) and CO₂ (~25 ppm) from
12 the lower levels of the ZOTTO tall tower in boreal Siberia; however, at the highest level (301
13 magl) the average July 2009 diurnal cycle amplitude is significantly smaller at ~50 ppb CH₄
14 and ~5 ppm CO₂, presumably because the top of the night-time PBL is often below this tallest
15 level. This may be the case at CRV, which despite its low height above ground level, is
16 elevated above the surrounding area and does not observe high CH₄ mole fractions from
17 lower-elevation wetlands at night, as their emissions would be trapped in the shallow valleys
18 below the site. The CRV tower is surrounded by deciduous and evergreen forest. however, so
19 the CO₂ cycle is comparably larger. The diurnal cycle at the 17 magl and 5 magl levels is
20 slightly greater than at 32 magl in summer, but not significantly so (1-2 ppb larger for CH₄
21 and 1-2 ppm for CO₂ on average in July and August).

22 4.2 Seasonality of winds and influence functions

23 The mid-afternoon daily average footprints (the entire set of footprints, without applying a
24 background or other filter) from the WRF-STILT model were examined to determine the
25 influence of different regions on the measurements at the tower throughout the year. The total
26 magnitude of land-surface influence (ocean influence is not included) on the tower
27 measurements for each month of each of the three study years (2012, 2013, 2014) was
28 determined (Fig. 3), along with the total influence of several sub-regions: Canada (light blue,
29 Fig. 3), the North Slope of Alaska (defined as north of the Brooks Range, red, Fig. 3), the
30 remainder of Alaska (dark blue, Fig. 3), and Eurasia (yellow, Fig. 3). The seasonality of the
31 land surface influence is clear and consistent between all three years. Specifically, in all three
32 years, the months of May through September show significantly less land surface influence on

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1 the tower than October through April. This stems from the smaller influence of Canada, and
2 to a lesser extent, lower Alaska, on the measurements during the summer months. The
3 influence of the Eurasian continent is very small throughout the years, but so is the influence
4 of the North Slope of Alaska. This is also apparent when the mid-afternoon footprint
5 influences are aggregated over seasons and years, as shown in the 80% influence range (Fig.
6 1a), which does not include the North Slope region. The footprint influence from the subset of
7 days used in the analysis in sections 4.4 and 4.5 is also shown in Fig. 1a, showing
8 correspondingly less influence from Canada, since the main filter applied removed days for
9 which significant portions of the air mass did not enter the domain from the West. Without a
10 good method for estimating background concentrations when winds are coming from the
11 eastern sector we are substantially limiting the potential of this tower to monitor fluxes from
12 the domain east of the site. From this analysis we also conclude that measurements at the
13 CRV tower are not substantially affected by emissions north of the Brooks Range, and any
14 emissions estimates made using the tower measurements will not apply to the North Slope.

15 Daytime wind measurements from the 2D sonic anemometer (from all days when it was
16 operational) at the tower support the finding of large seasonality in the footprints (Fig. 4).
17 Winds at the tower during May-September are predominantly from the west and southwest,
18 with some frequency of winds from the east as well. However, from October to April, the
19 winds are almost exclusively from the east/northeast. These wind directions support the
20 conclusion from the model influence functions that wintertime measurements are more
21 influenced by Canadian land than in summertime, as shown in Fig. 3. In addition, winds in
22 any season do not generally come from the North, supporting the lack of influence from the
23 North Slope. Similar seasonality and lack of northern influence was found in a recent
24 analysis of data from NOAA/ESRL Aircraft Network at Poker Flat, AK (Sweeney et al.,
25 2015).

26 **4.3 Background and Relative Enhancements of CH₄, CO₂, and CO**

27 The definition of an appropriate background is a crucial aspect of analyzing the CRV tower
28 CO₂ and CH₄ measurements. We calculate the background as described in Sect. 3.3, using
29 the particle back trajectories and the empirical Pacific boundary curtain, and refer to this as
30 the footprint background. We also compare this background to the value of the same Pacific
31 curtain at 3500 masl and 65°N, i.e. the free troposphere at the latitude of the tower. For CO₂
32 (middle panel, Fig. 5) the definition of the background does not have as large an effect as it

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1 does for CH₄. For CH₄ (top panel, Fig. 5) the choice of background is crucial to any analysis
2 of the measurements, for two reasons. First, the CH₄ signal at CRV is relatively small
3 compared to the variability of the background. Second, the CH₄ background varies depending
4 on the latitudinal origin of the air mass, because of the large global latitudinal gradient in CH₄
5 (Dlugokencky et al., 2009). Comparison of the measurements with the footprint background
6 and free-tropospheric background (Fig. 5) illustrates that the footprint background varies at
7 synoptic time scales as air-mass origins change, and tracks the variability in the measurements
8 at the site. CH₄ enhancements over background are small and thus very sensitive to
9 background choice (top panel in Fig. 5). We note that despite the small signal, however, the
10 time series of CH₄ observations clearly shows both wintertime and summertime
11 enhancements, with wintertime enhancements sometimes correlated with CO enhancements
12 as well, indicating a possible anthropogenic source for these signals (see Sect. 4.6 and Table
13 1). Evidence of biomass burning events is also clear in all three species, but most easily
14 observed in the CO signals during the summers of 2012 and 2013.

15 **4.4 CO₂ Model-Observation Comparison**

16 Observations of monthly mean CO₂ relative to the background (ΔCO_2) show consistent
17 features from year to year (Fig. 6), with the sign of the enhancements showing the sign of the
18 monthly net CO₂ fluxes, or Net Ecosystem Exchange (NEE). Positive enhancements from
19 January-April indicate that respiration is occurring even during this coldest period of the year.
20 In addition, all years show the highest respiration signal in October, possibly indicative of
21 photosynthesis stopping while soil temperatures are still high enough to sustain significant
22 respiration, although some of this signal could also be due to the seasonality in vertical
23 mixing and/or winds. Although the maximum drawdown occurs in July and is of similar
24 magnitude in all years (~8 ppm), the transition from net respiration to net photosynthetic
25 uptake occurs earlier in 2014 (April) than in 2012 and 2013 (May). The timing and
26 magnitude of the ΔCO_2 observations relative to background represent a stringent test for the
27 transport and surface flux models.

28 The modeled ΔCO_2 from the convolution of WRF-STILT footprints with PolarVPRM fluxes
29 are compared to hourly averaged observed ΔCO_2 mole fractions at the tower during the mid-
30 afternoon in Figs. 6 and 7. (Note that the time series data in Fig. 6 (top) has not been filtered,
31 but the monthly averages in the lower panel and the data shown in Fig. 7 only use filtered

1 data.) Both the hourly time series and monthly average comparisons between modeled and
2 observed ΔCO_2 at the tower during mid-day hours indicate that the PolarVPRM fluxes and
3 WRF-STILT meteorology are able to reproduce the magnitude and timing of the tower CO_2
4 signal remarkably well during most seasons (Fig. 6). Hourly observations of ΔCO_2 that
5 satisfy the filtering conditions are well correlated with modeled ΔCO_2 in all three years (Fig.
6 7). The data close to the 1:1 line indicate that the magnitude of the fluxes is generally well
7 captured by the model. The correlations are strong in all three years ($R^2 = 0.61$ to 0.75),
8 indicating that the PolarVPRM CO_2 fluxes and the WRF-STILT transport model are able to
9 reproduce observed signals at the tower remarkably well with no adjustment to match the
10 data.

11 There are two exceptions apparent in otherwise the very good comparison between the model
12 and observations. First, monthly average ΔCO_2 observations compared to the model (Fig. 6,
13 lower panel) indicate that the PolarVPRM/WRF-STILT modeled NEE is slightly more
14 negative than observations in May and June in both 2012 and 2013, with an earlier spring
15 drawdown in the model than the observations suggest. However, in all of these months the
16 model results, with no uncertainty estimates, overlap with the one-sigma data uncertainty.
17 This difference is also observable in the correlations between modeled and observed ΔCO_2
18 (Fig. 7), with some data points with more negative ΔCO_2 in the model than in the
19 observations. Whether this small offset between model and observations results from
20 insufficient modeled respiration or too much modeled photosynthesis during the spring is
21 impossible to tell from CO_2 observations only. A second exception to the very good
22 agreement is that the model systematically underestimates the magnitude of the observed late
23 fall respiration flux (October to November) in all three years. This may be because model
24 respiration is calculated as a function of air temperature when per-pixel snow cover area is
25 <50%, whereas actual rates of late fall respiration are influenced by microbial activity
26 sustained in the soil, which cools more gradually than the air. Despite these differences, the
27 overall good agreement between model and observations indicates that in addition to
28 capturing the magnitude, the PolarVPRM and WRF-STILT models are likely capturing most
29 of the timing and spatial structure of the fluxes in boreal Alaska as well.

30 4.5 CH_4 Model-Observation Comparison

31 The scaled monthly CH_4 fluxes from the elevation-based and uniform (constant) flux maps
32 were convolved with the WRF-STILT footprints corresponding to the observations. The

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1 | hourly ΔCH_4 from each model was compared with the observed enhancements (Fig. 8) for
2 | each year. The elevation-based model enhancements (lower row, Fig. 8) match the data
3 | slightly better than the uniform flux model (upper row, Fig. 8) in 2012 and 2014, but not
4 | 2013. We also investigated a third spatial flux map pattern that was based on the gradient in
5 | elevation, but did not find any improvement in the correlations over the simpler elevation-
6 | based and uniform flux models. Neither model was able to achieve good correlations
7 | between the model and the observations, a conclusion that was also reached by Chang et al.
8 | (2014) when they investigated multiple different CH_4 flux models. This result is in sharp
9 | contrast to the high correlations achieved using the PolarVPRM CO_2 fluxes with the same
10 | WRF-STILT footprints, leading to the conclusion that the WRF-STILT meteorology is able to
11 | replicate observations when an accurate spatial flux map is used. Although the approximate
12 | magnitude of the CH_4 enhancements is correct because of the monthly scaling, the large
13 | spread and lack of correlation around the 1:1 line indicates that the model cannot replicate
14 | hourly variability in enhancements, because the uniform spatial and temporal (we assume
15 | constant monthly fluxes), representation is likely to be incorrect, and likely because of higher
16 | relative uncertainty in the background.

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17 | 4.6 Average Scaled CH_4 Fluxes

18 | Monthly CH_4 fluxes from the uniform flux map were averaged over the state of Alaska to
19 | obtain average fluxes for each month and for each year, and compared with the results from
20 | (Chang et al., 2014) (Fig. 9). Results from the elevation-based flux map were statistically the
21 | same as those from the uniform flux map and are not shown. CH_4 fluxes are small relative to
22 | those determined from some flux tower or chamber-based studies in arctic wetlands
23 | (Euskirchen et al., 2014; Johnston et al., 2014; Whalen and Reeburgh, 1988; Fan et al., 1992;
24 | Olefeldt et al., 2013), but very similar in magnitude to CH_4 fluxes recently reported from a
25 | black spruce forest during the snow-free seasons from 2011-2013 at a flux tower site in
26 | Fairbanks, AK (Iwata et al., 2015), as well as those reported from the Zotino Tall Tower
27 | Observatory (ZOTTO) in Siberia during the summers of 2009-2011 ($7.7 \text{ mg CH}_4 \text{ m}^{-2}\text{d}^{-1}$)
28 | (Winderlich et al., 2014). They are also smaller than fluxes estimated by Chang et al. (2014)
29 | using CARVE aircraft observations in the summer of 2012 (Fig. 9), but the two results
30 | overlap within their 1-sigma uncertainty bands. One possible reason for the lower average
31 | fluxes observed at the CARVE tower could be the region to which the CARVE tower
32 | observations are sensitive in the summer. The CARVE tower observations do not capture

1 emissions over the North Slope of Alaska, where other studies have shown large (but highly
2 variable) CH₄ emissions, at least over small areas (Euskirchen et al., 2014; Morrissey and
3 Livingston, 1992; von Fischer et al., 2010; Olefeldt et al., 2013). The signal in CH₄ at CRV
4 may also be small because of the CARVE tower's large region of influence, which integrates
5 signals from a wide variety of ecosystems that have different flux profiles, including not only
6 low-lying wetlands and forests, but also extensive upland and mountain regions.

7 The tower observations also suggest the presence of non-zero fall and wintertime fluxes.
8 Mastepanov et al. (2008) observed a burst of CH₄ emissions in high-latitude wetlands in fall,
9 and, more recently, Zona et al. (2016) reported significant natural CH₄ fluxes persisting
10 through the late fall in the North Slope of Alaska; our results support the existence of late-fall
11 (September-October) CH₄ fluxes in the boreal zone as well. Additionally, our analysis also
12 suggests the presence of CH₄ emissions in late winter (January-March) in some years, u
13 although this conclusion is uncertain and requires further investigation with a larger data set,
14 given the smaller data sample in our analysis in winter and the larger uncertainty associated
15 with modeled transport, as indicated by the sensitivity of the influence footprints to altitude
16 during winter months. To understand the role that fossil sources from nearby Fairbanks or
17 farther away might play, especially in winter, we analyze correlations between ΔCH₄ and
18 ΔCO mid-afternoon hourly enhancements. These enhancements indicate that some wintertime
19 CH₄ emissions are likely anthropogenic, with coefficients of determination (R²) generally
20 larger in the winter months and close to zero in June, July and August of all years. Not all
21 winter months show high correlations, and May 2012 also has highly correlated ΔCH₄ and
22 ΔCO (Table 1).

23 We note that total uncertainty on the quantitative flux analysis presented here has not been
24 calculated, and would come from a number of components, including transport error in the
25 model. We also note that transport uncertainty is higher in the winter months, when the
26 elevation resolution in the model introduces a larger error than during spring and summer
27 periods. Here we have calculated the uncertainty of the observed enhancements, based on the
28 background uncertainty, and an estimate of the uncertainty associated with the model's
29 representation of the tower's ridge-top location based on our sensitivity analysis; this total is
30 still likely an underestimate of the total uncertainty of the flux estimates.

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1 5 Conclusions

2 The CARVE tower, located on a ridge outside Fairbanks, is well situated to provide regional
3 year-round CO₂ and CH₄ observations that provide context to the CARVE aircraft campaign
4 measurements, which were made throughout Alaska from March to November from 2012 to
5 2015. The WRF-STILT transport model was used to determine the influence region of the
6 site and its inter-annual and seasonal variability. The model [results](#) showed significantly more
7 influence from the region east of the tower in wintertime, a pattern that [was repeated in all](#)
8 three years and was confirmed by anemometer data from the site. The model also indicated
9 that processes in the North Slope of Alaska have very little influence on the tower
10 observations. This seasonality of transport to the region has been previously documented
11 (Sweeney et al., 2015), and implies that additional long-term observing sites are required to
12 constrain Alaskan fluxes; a site in Western Alaska, for example, would be more likely to have
13 [fluxes from](#) interior Alaska in its observation footprint in the wintertime, and a site north of
14 the Brooks Range would be required to investigate fluxes from the North Slope.

15 We calculated enhancements of CO₂ and CH₄ during local mid-afternoon times by subtracting
16 a background also determined using the WRF-STILT model particle trajectories, and found
17 that the background choice is critical for CH₄, for which enhancements are very small, and of
18 the same order of magnitude as the uncertainties. CO₂ enhancements at the CARVE tower
19 site are replicated remarkably well by the WRF-STILT model when convolved with
20 PolarVPRM biogenic CO₂ fluxes (Luus and Lin, 2015), [with a few noted exceptions](#). The
21 high correlation between modeled and observed CO₂ give confidence in the STILT footprints
22 and the WRF meteorological model that was used to generate them. The signal in CO₂ is
23 larger than that for CH₄, such that the background uncertainty is not as large relative to the
24 enhancements or depletions.

25 The WRF-STILT meteorological model enables us to constrain the magnitude of mean
26 monthly CH₄ fluxes in the region of influence of the tower for all three years. Using two
27 different distribution maps of CH₄ emissions we determine that average CH₄ emissions over
28 Alaska in summer range between 3 and 8 mg CH₄ m⁻²d⁻¹, albeit with large uncertainties
29 stemming from the large uncertainty in the background. [The modeled enhancements do not](#)
30 [correlate well with observations, however, indicating that a model with a more accurate](#)
31 [spatial and temporal distribution of CH₄ fluxes is needed](#). The tower observations also
32 indicate that there are no significant differences between the three years. This simple analysis

1 provides a flux estimate range that applies as an average over a very large area of Alaska (Fig.
2 | 1a). CH₄ fluxes in this region are likely to be highly heterogeneous, but our measurements
3 show that the average flux over the entire region is relatively small. This result suggests that
4 although there may be small areas with large fluxes, there are other areas with little to no
5 | emissions, or possibly uptake by tundra (Juncher Jorgensen et al., 2015). For this reason, the
6 observations at the tower give context to other flux estimates, from flux towers or chamber
7 studies, for example, that are representative of much smaller areas and are difficult to scale to
8 the larger domain because of high spatial and temporal variability. We also observe CH₄
9 | enhancements persisting into the fall (September-October) in all three years, and the analysis
10 shows some CH₄ enhancements in winter and early spring, depending on the year, which may
11 be partially or entirely anthropogenic, based on an analysis of correlations of CH₄ with CO.
12 | These late fall and wintertime enhancements, and their large uncertainties in this analysis,
13 demonstrate the need for year-round in-situ observations in the high northern latitudes.

14 The CARVE tower site provides a continuous observation platform that will contribute to
15 future efforts to investigate the high-latitude carbon cycle and its response to warming. As a
16 long-term measurement site with a large regional coverage it will provide understanding of
17 changing emissions in interior Alaska. Our analysis of the years 2012-2014 indicates no
18 measurable change in emissions influencing this site over this period. These tower
19 observations are sensitive to changes in emissions and provide the capability to detect such
20 changes in the future. However, the location of the CARVE tower prohibits any
21 quantification or observation of processes on the North Slope, indicating that additional long-
22 term observation sites with large regional coverage are required north of the Brooks Range of
23 Alaska to detect changes in emissions in the higher northern latitudes. Future efforts will
24 combine the observations from the CARVE tower with other aircraft and ground-based
25 observations in a formal inversion framework to solve for spatially and temporally resolved
26 CH₄ and CO₂ fluxes in Alaska.

27 **Data availability**

28 All the tower observations and WRF-STILT footprints used in our analysis are publicly
29 | available on the CARVE data portal at <https://ilma.jpl.nasa.gov/portal/>. They are also
30 archived at the U.S. Oak Ridge National Laboratory Distributed Active Archive Center for
31 Biogeochemical Dynamics (ORNL DAAC, <https://daac.ornl.gov>).

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1 References

- 2 Bruhwiler, L., Dlugokencky, E., Masarie, K., Ishizawa, M., Andrews, A., Miller, J., Sweeney,
3 C., Tans, P., and Worthy, D.: CarbonTracker-CH4: an assimilation system for estimating
4 emissions of atmospheric methane, *Atmos. Chem. Phys.*, 14, 8269-8293, 10.5194/acp-14-
5 8269-2014, 2014.
- 6 CarbonTracker CT2013B: <http://www.esrl.noaa.gov/gmd/ccgg/carbontracker/>, access:
7 October 17, 2013.
- 8 Chang, R. Y. W., Miller, C. E., Dinardo, S. J., Karion, A., Sweeney, C., Daube, B. C.,
9 Henderson, J. M., Mountain, M. E., Eluszkiewicz, J., Miller, J. B., Bruhwiler, L. M. P., and
10 Wofsy, S. C.: Methane emissions from Alaska in 2012 from CARVE airborne observations,
11 *Proc. Natl. Acad. Sci. U. S. A.*, 111, 16694-16699, 10.1073/pnas.1412953111, 2014.
- 12 Chen, H., Karion, A., Rella, C. W., Winderlich, J., Gerbig, C., Filges, A., Newberger, T.,
13 Sweeney, C., and Tans, P. P.: Accurate measurements of carbon monoxide in humid air using
14 the cavity ring-down spectroscopy (CRDS) technique, *Atmos. Meas. Tech.*, 6, 1031-1040,
15 10.5194/amt-6-1031-2013, 2013.
- 16 Dlugokencky, E. J., Myers, R. C., Lang, P. M., Masarie, K. A., Crotwell, A. M., Thoning, K.
17 W., Hall, B. D., Elkins, J. W., and Steele, L. P.: Conversion of NOAA atmospheric dry air
18 CH₄ mole fractions to a gravimetrically prepared standard scale, *Journal of Geophysical*
19 *Research: Atmospheres*, 110, D18306, 10.1029/2005JD006035, 2005.
- 20 Dlugokencky, E. J., Bruhwiler, L., White, J. W. C., Emmons, L. K., Novelli, P. C., Montzka,
21 S. A., Masarie, K. A., Lang, P. M., Crotwell, A. M., Miller, J. B., and Gatti, L. V.:
22 Observational constraints on recent increases in the atmospheric CH₄ burden, *Geophys. Res.*
23 *Letts.*, 36, 10.1029/2009gl039780, 2009.
- 24 Euskirchen, E. S., Edgar, C. W., Turetsky, M. R., Waldrop, M. P., and Harden, J. W.:
25 Differential response of carbon fluxes to climate in three peatland ecosystems that vary in the
26 presence and stability of permafrost, *Journal of Geophysical Research: Biogeosciences*, 119,
27 1576-1595, 10.1002/2014JG002683, 2014.
- 28 Fan, S. M., Wofsy, S. C., Bakwin, P. S., Jacob, D. J., Anderson, S. M., Keibian, P. L.,
29 McManus, J. B., Kolb, C. E., and Fitzjarrald, D. R.: Micrometeorological measurements of
30 CH₄ and CO₂ exchange between the atmosphere and subarctic tundra, *Journal of*
31 *Geophysical Research: Atmospheres*, 97, 16627-16643, 10.1029/91JD02531, 1992.
- 32 GLOBE Task Team and others (Hastings, D. A., Paula K. Dunbar, Gerald M. Elphinstone,,
33 Mark Bootz, H. M., Hiroshi Maruyama, Hiroshi Masaharu, Peter Holland, John, Payne, N. A.
34 B., Thomas L. Logan, J.-P. Muller, Gunter Schreier, and John S., and MacDonald): The
35 Global Land One-kilometer Base Elevation (GLOBE) Digital Elevation Model, Version 1.0.,
36 edited by: National Oceanic and Atmospheric Administration, N. G. D. C., 325 Broadway,
37 Boulder, Colorado 80303, 1999.
- 38 Harriss, R. C., Sachse, G. W., Hill, G. F., Wade, L., Bartlett, K. B., Collins, J. E., Steele, L.
39 P., and Novelli, P. C.: Carbon monoxide and methane in the North American Arctic and sub-
40 arctic troposphere - July- August 1988, *J. Geophys. Res.-Atmos.*, 97, 16589-16599, 1992.
- 41 Hayes, D. J., Kicklighter, D. W., McGuire, A. D., Chen, M., Zhuang, Q., Yuan, F., Melillo, J.
42 M., and Wullschleger, S. D.: The impacts of recent permafrost thaw on land-atmosphere
43 greenhouse gas exchange, *Environmental Research Letters*, 9, 045005, 2014.

1 Henderson, J. M., Eluszkiewicz, J., Mountain, M. E., Nehrkorn, T., Chang, R. Y. W., Karion,
2 A., Miller, J. B., Sweeney, C., Steiner, N., Wofsy, S. C., and Miller, C. E.: Atmospheric
3 transport simulations in support of the Carbon in Arctic Reservoirs Vulnerability Experiment
4 (CARVE), *Atmos. Chem. Phys.*, 15, 4093-4116, 10.5194/acp-15-4093-2015, 2015.

5 Homer, C. G., Dewitz, J. A., Yang, L., Jin, S., Danielson, P., Xian, G., Coulston, J., Herold,
6 N. D., Wickham, J. D., and Megown, K.: Completion of the 2011 National Land Cover
7 Database for the conterminous United States-Representing a decade of land cover change
8 information., *Photogrammetric Engineering and Remote Sensing*, 81, 345-354, 2015.

9 IPCC: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I
10 to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change.,
11 Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 1535,
12 2013.

13 Iwata, H., Harazono, Y., Ueyama, M., Sakabe, A., Nagano, H., Kosugi, Y., Takahashi, K.,
14 and Kim, Y.: Methane exchange in a poorly-drained black spruce forest over permafrost
15 observed using the eddy covariance technique, *Agricultural and Forest Meteorology*, 214-
16 215, 157-168, <http://dx.doi.org/10.1016/j.agrformet.2015.08.252>, 2015.

17 Jeong, S., Hsu, Y.-K., Andrews, A. E., Bianco, L., Vaca, P., Wilczak, J. M., and Fischer, M.
18 L.: A multitower measurement network estimate of California's methane emissions, *Journal of*
19 *Geophysical Research: Atmospheres*, 118, 2013JD019820, 10.1002/jgrd.50854, 2013.

20 Johnston, C. E., Ewing, S. A., Harden, J. W., Varner, R. K., Wickland, K. P., Koch, J. C.,
21 Fuller, C. C., Manies, K., and Jorgenson, M. T.: Effect of permafrost thaw on CO₂ and CH₄
22 exchange in a western Alaska peatland chronosequence, *Environmental Research Letters*, 9,
23 085004, 2014.

24 Juncher Jorgensen, C., Lund Johansen, K. M., Westergaard-Nielsen, A., and Elberling, B.:
25 Net regional methane sink in High Arctic soils of northeast Greenland, *Nature Geosci.*, 8, 20-
26 23, 10.1038/ngeo2305, 2015.

27 Kirschke, S., Bousquet, P., Ciais, P., Saunoy, M., Canadell, J. G., Dlugokencky, E. J.,
28 Bergamaschi, P., Bergmann, D., Blake, D. R., Bruhwiler, L., Cameron-Smith, P., Castaldi, S.,
29 Chevallier, F., Feng, L., Fraser, A., Heimann, M., Hodson, E. L., Houweling, S., Josse, B.,
30 Fraser, P. J., Krummel, P. B., Lamarque, J.-F., Langenfelds, R. L., Le Quere, C., Naik, V.,
31 O'Doherty, S., Palmer, P. I., Pison, I., Plummer, D., Poulter, B., Prinn, R. G., Rigby, M.,
32 Ringeval, B., Santini, M., Schmidt, M., Shindell, D. T., Simpson, I. J., Spahni, R., Steele, L.
33 P., Strode, S. A., Sudo, K., Szopa, S., van der Werf, G. R., Voulgarakis, A., van Weele, M.,
34 Weiss, R. F., Williams, J. E., and Zeng, G.: Three decades of global methane sources and
35 sinks, *Nature Geosci.*, 6, 813-823, 10.1038/ngeo1955, 2013.

36 Kort, E. A., Eluszkiewicz, J., Stephens, B. B., Miller, J. B., Gerbig, C., Nehrkorn, T., Daube,
37 B. C., Kaplan, J. O., Houweling, S., and Wofsy, S. C.: Emissions of CH₄ and N₂O over the
38 United States and Canada based on a receptor-oriented modeling framework and COBRA-NA
39 atmospheric observations, *Geophys. Res. Lett.*, 35, 5, 10.1029/2008gl034031, 2008.

40 Lin, J. C., Gerbig, C., Wofsy, S. C., Andrews, A. E., Daube, B. C., Davis, K. J., and Grainger,
41 C. A.: A near-field tool for simulating the upstream influence of atmospheric observations:
42 The Stochastic Time-Inverted Lagrangian Transport (STILT) model, *Journal of Geophysical*
43 *Research: Atmospheres*, 108, 4493, 10.1029/2002JD003161, 2003.

44 Lin, J. C., Brunner, D., Gerbig, C., Stohl, A., Luhar, A. K., and Webley, P. W.: Lagrangian
45 Modeling of the Atmosphere, Vol. 200, American Geophysical Union, 349 pp., 2012.

1 Luus, K. A., and Lin, J. C.: The Polar Vegetation Photosynthesis and Respiration Model: a
2 parsimonious, satellite-data-driven model of high-latitude CO₂ exchange, *Geosci. Model*
3 *Dev.*, 8, 2655-2674, 10.5194/gmd-8-2655-2015, 2015.

4 Mastepanov, M., Sigsgaard, C., Dlugokencky, E. J., Houweling, S., Strom, L., Tamstorf, M.
5 P., and Christensen, T. R.: Large tundra methane burst during onset of freezing, *Nature*, 456,
6 628-U658, 10.1038/nature07464, 2008.

7 McGuire, A. D., Anderson, L. G., Christensen, T. R., Dallimore, S., Guo, L. D., Hayes, D. J.,
8 Heimann, M., Lorenson, T. D., Macdonald, R. W., and Roulet, N.: Sensitivity of the carbon
9 cycle in the Arctic to climate change, *Ecol. Monogr.*, 79, 523-555, 2009.

10 McGuire, A. D., Hayes, D. J., Kicklighter, D. W., Manizza, M., Zhuang, Q., Chen, M.,
11 Follows, M. J., Gurney, K. R., McClelland, J. W., Melillo, J. M., Peterson, B. J., and Prinn, R.
12 G.: An analysis of the carbon balance of the Arctic Basin from 1997 to 2006, *Tellus Ser. B-*
13 *Chem. Phys. Meteorol.*, 62, 455-474, 10.1111/j.1600-0889.2010.00497.x, 2010.

14 McKain, K., Down, A., Raciti, S. M., Budney, J., Hutyra, L. R., Floerchinger, C., Herndon, S.
15 C., Nehrkorn, T., Zahniser, M. S., Jackson, R. B., Phillips, N., and Wofsy, S. C.: Methane
16 emissions from natural gas infrastructure and use in the urban region of Boston,
17 Massachusetts, *Proceedings of the National Academy of Sciences*, 112, 1941-1946,
18 10.1073/pnas.1416261112, 2015.

19 Mesinger, F., DiMego, G., Kalnay, E., Mitchell, K., Shafran, P. C., Ebisuzaki, W., Jović, D.,
20 Woollen, J., Rogers, E., Berbery, E. H., Ek, M. B., Fan, Y., Grumbine, R., Higgins, W., Li,
21 H., Lin, Y., Manikin, G., Parrish, D., and Shi, W.: North American Regional Reanalysis,
22 *Bulletin of the American Meteorological Society*, 87, 343-360, 10.1175/BAMS-87-3-343,
23 2006.

24 Miller, S. M., Worthy, D. E. J., Michalak, A. M., Wofsy, S. C., Kort, E. A., Havice, T. C.,
25 Andrews, A. E., Dlugokencky, E. J., Kaplan, J. O., Levi, P. J., Tian, H., and Zhang, B.:
26 Observational constraints on the distribution, seasonality, and environmental predictors of
27 North American boreal methane emissions, *Glob. Biogeochem. Cycle*, 28, 146-160,
28 10.1002/2013GB004580, 2014.

29 Morrissey, L. A., and Livingston, G. P.: Methane emissions from Alaska Arctic tundra: An
30 assessment of local spatial variability, *Journal of Geophysical Research: Atmospheres*, 97,
31 16661-16670, 10.1029/92JD00063, 1992.

32 Nehrkorn, T., Eluszkiewicz, J., Wofsy, S., Lin, J., Gerbig, C., Longo, M., and Freitas, S.:
33 Coupled weather research and forecasting–stochastic time-inverted lagrangian transport
34 (WRF–STILT) model, *Meteorol Atmos Phys*, 107, 51-64, 10.1007/s00703-010-0068-x, 2010.

35 O'Connor, F. M., Boucher, O., Gedney, N., Jones, C. D., Folberth, G. A., Coppell, R.,
36 Friedlingstein, P., Collins, W. J., Chappellaz, J., Ridley, J., and Johnson, C. E.: Possible role
37 of wetlands, permafrost, and methane hydrates in the methane cycle under future climate
38 change: A review, *Rev. Geophys.*, 48, RG4005, 10.1029/2010rg000326, 2010.

39 Oechel, W. C., Hastings, S. J., Vourlitis, G., Jenkins, M., Riechers, G., and Grulke, N.: Recent
40 change of Arctic tundra ecosystems from a net carbon dioxide sink to a source, *Nature*, 361,
41 520-523, 1993.

42 Olefeldt, D., Turetsky, M. R., Crill, P. M., and McGuire, A. D.: Environmental and physical
43 controls on northern terrestrial methane emissions across permafrost zones, *Glob. Change*
44 *Biol.*, 19, 589-603, 10.1111/gcb.12071, 2013.

1 Rella, C. W., Chen, H., Andrews, A. E., Filges, A., Gerbig, C., Hatakka, J., Karion, A., Miles,
2 N. L., Richardson, S. J., Steinbacher, M., Sweeney, C., Wastine, B., and Zellweger, C.: High
3 accuracy measurements of dry mole fractions of carbon dioxide and methane in humid air,
4 *Atmos. Meas. Tech.*, 6, 837-860, 10.5194/amt-6-837-2013, 2013.

5 Riggs, G., and Hall, D.: MODIS snow and ice products, and their assessment and
6 applications, in: *Land Remote Sensing and Global Environmental Change, Remote Sensing
7 and Digital Image Processing*, edited by: Ramachandran, B., Justice, C. O., and Abrams, M.
8 J., 11, 681-707, 2011.

9 Sasakawa, M., Shimoyama, K., Machida, T., Tsuda, N., Suto, H., Arshinov, M., Davydov, D.,
10 Fofonov, A., Krasnov, O., Saeki, T., Koyama, Y., and Maksyutov, S.: Continuous
11 measurements of methane from a tower network over Siberia, *Tellus B*, 62, 403-416,
12 10.1111/j.1600-0889.2010.00494.x, 2010.

13 Sasakawa, M., Machida, T., Tsuda, N., Arshinov, M., Davydov, D., Fofonov, A., and
14 Krasnov, O.: Aircraft and tower measurements of CO₂ concentration in the planetary
15 boundary layer and the lower free troposphere over southern taiga in West Siberia: Long-term
16 records from 2002 to 2011, *J. Geophys. Res.-Atmos.*, 118, 9489-9498, 10.1002/jgrd.50755,
17 2013.

18 Schuur, E. A. G., Bockheim, J., Canadell, J. G., Euskirchen, E., Field, C. B., Goryachkin, S.
19 V., Hagemann, S., Kuhry, P., Lafleur, P. M., Lee, H., Mazhitova, G., Nelson, F. E., Rinke, A.,
20 Romanovsky, V. E., Shiklomanov, N., Tarnocai, C., Venevsky, S., Vogel, J. G., and Zimov,
21 S. A.: Vulnerability of permafrost carbon to climate change: Implications for the global
22 carbon cycle, *Bioscience*, 58, 701-714, 10.1641/b580807, 2008.

23 Schuur, E. A. G., Vogel, J. G., Crummer, K. G., Lee, H., Sickman, J. O., and Osterkamp, T.
24 E.: The effect of permafrost thaw on old carbon release and net carbon exchange from tundra,
25 *Nature*, 459, 556-559, 10.1038/nature08031, 2009.

26 Schuur, E. A. G., McGuire, A. D., Schadel, C., Grosse, G., Harden, J. W., Hayes, D. J.,
27 Hugelius, G., Koven, C. D., Kuhry, P., Lawrence, D. M., Natali, S. M., Olefeldt, D.,
28 Romanovsky, V. E., Schaefer, K., Turetsky, M. R., Treat, C. C., and Vonk, J. E.: Climate
29 change and the permafrost carbon feedback, *Nature*, 520, 171-179, 10.1038/nature14338,
30 2015.

31 Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Wang,
32 X.-Y., Wang, W., and Powers, J. G.: A Description of the Advanced Research WRF Version
33 3, Technical Note 475+STR, MMM Division, NCAR, Boulder, CO, USA, 133, 2008.

34 MODIS Vegetation Indices (MOD13) C5 Users's Guide, Terrestrial Biophysics and Remote
35 Sensing Lab, The University of Arizona: <http://www.ctahr.hawaii.edu/grem/modis-ug.pdf>,
36 access: September 7, 2010.

37 Sweeney, C., Karion, A., Wolter, S., Newberger, T., Guenther, D., Higgs, J. A., Andrews, A.
38 E., Lang, P. M., Neff, D., Dlugokencky, E., Miller, J. B., Montzka, S. A., Miller, B. R.,
39 Masarie, K. A., Biraud, S. C., Novelli, P. C., Crotwell, M., Crotwell, A. M., Thoning, K., and
40 Tans, P. P.: Seasonal climatology of CO₂ across North America from aircraft measurements
41 in the NOAA/ESRL Global Greenhouse Gas Reference Network, *Journal of Geophysical
42 Research: Atmospheres*, n/a-n/a, 10.1002/2014JD022591, 2015.

43 von Fischer, J. C., Rhew, R. C., Ames, G. M., Fosdick, B. K., and von Fischer, P. E.:
44 Vegetation height and other controls of spatial variability in methane emissions from the

1 Arctic coastal tundra at Barrow, Alaska, *Journal of Geophysical Research: Biogeosciences*,
2 115, n/a-n/a, 10.1029/2009JG001283, 2010.

3 Walter, K. M., Smith, L. C., and Chapin, F. S.: Methane bubbling from northern lakes:
4 present and future contributions to the global methane budget, *Philosophical Transactions of*
5 *the Royal Society a-Mathematical Physical and Engineering Sciences*, 365, 1657-1676,
6 10.1098/rsta.2007.2036, 2007.

7 Whalen, S. C., and Reeburgh, W. S.: A methane flux time series for tundra environments,
8 *Glob. Biogeochem. Cycle*, 2, 399-409, 10.1029/GB002i004p00399, 1988.

9 Wilson, A. B., Bromwich, D. H., and Hines, K. M.: Evaluation of Polar WRF forecasts on the
10 Arctic System Reanalysis domain: Surface and upper air analysis, *Journal of Geophysical*
11 *Research: Atmospheres*, 116, 10.1029/2010JD015013, 2011.

12 Winderlich, J., Chen, H., Gerbig, C., Seifert, T., Kolle, O., Lavric, J. V., Kaiser, C., Hofer, A.,
13 and Heimann, M.: Continuous low-maintenance CO₂/CH₄/H₂O measurements at the Zotino
14 Tall Tower Observatory (ZOTTO) in Central Siberia, *Atmos. Meas. Tech.*, 3, 1113-1128,
15 10.5194/amt-3-1113-2010, 2010.

16 Winderlich, J., Gerbig, C., Kolle, O., and Heimann, M.: Inferences from CO₂ and CH₄
17 concentration profiles at the Zotino Tall Tower Observatory (ZOTTO) on regional
18 summertime ecosystem fluxes, *Biogeosciences*, 11, 2055-2068, 10.5194/bg-11-2055-2014,
19 2014.

20 Worthy, D., Taylor, C., Dlugokencky, E. J., Chan, E., Nisbet, E. G., and Laurila, T.: AMAP
21 Assessment 2015: Methane as an Arctic climate forcer, Chapter 6: Long-term monitoring of
22 atmospheric methane., Arctic Monitoring and Assessment Programme (AMAP), Oslo,
23 Norway. <http://www.amap.no/documents/download/2499>, 61-75, 2015.

24 Zhao, C. L., and Tans, P. P.: Estimating uncertainty of the WMO mole fraction scale for
25 carbon dioxide in air, *Journal of Geophysical Research*, 111, D08S09,
26 10.1029/2005jd006003, 2006.

27 Zona, D., Gioli, B., Commane, R., Lindaas, J., Wofsy, S. C., Miller, C. E., Dinardo, S. J.,
28 Dengel, S., Sweeney, C., Karion, A., Chang, R. Y.-W., Henderson, J. M., Murphy, P. C.,
29 Goodrich, J. P., Moreaux, V., Liljedahl, A., Watts, J. D., Kimball, J. S., Lipson, D. A., and
30 Oechel, W. C.: Cold season emissions dominate the Arctic tundra methane budget,
31 *Proceedings of the National Academy of Sciences*, 113, 40-45, 10.1073/pnas.1516017113,
32 2016.

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Table 1. Coefficient of determination (R^2) between ΔCH_4 and ΔCO in each month. Months with $R^2 > 0.2$ are in bold.

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	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
2012	0.30	0.14	0.12	0.16	0.81	0.00	0.00	0.05	0.17	0.22	0.49	NA
2013	0.66	0.14	0.19	0.14	0.26	0.01	0.07	0.01	0.19	0.09	0.01	0.15
2014	0.49	0.67	0.66	0.03	0.17	0.00	0.07	0.00	0.10	0.06	0.10	0.00

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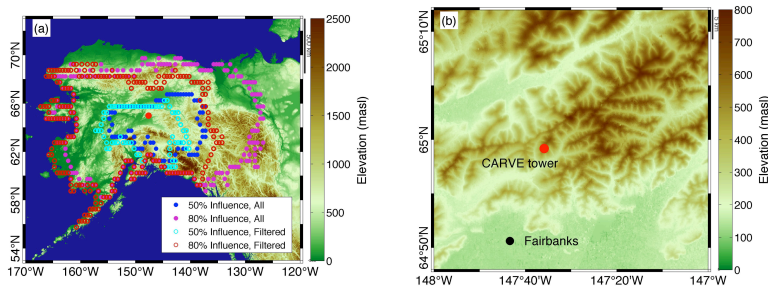
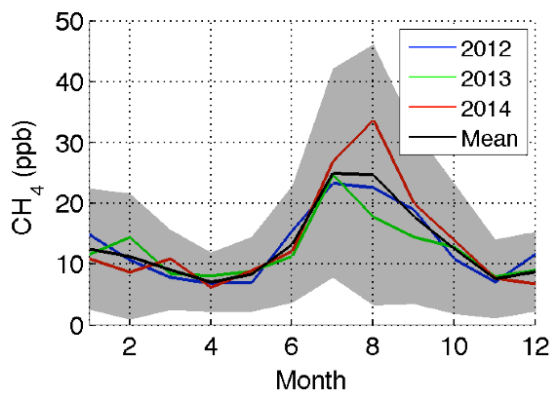
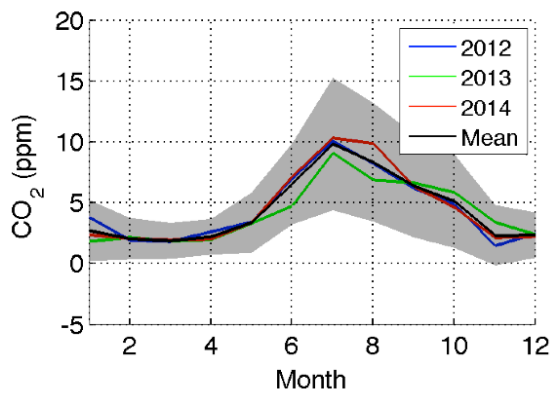


Figure 1. CO₂ and CH₄ measurements from the CARVE tower (filled red circle, both panels) have high sensitivity to the boreal forests and lowlands of interior Alaska as shown by the 50% (blue) and 80% (purple) surface influence contours for the average WRF-STILT influence functions calculated for mid-afternoon averages over the period 2012-2014 (a). The same influence contours (cyan and red open circles) are shown for the subset of footprints used in the flux analysis after filtering. Note the elevation scale differs between the panels. Elevation data in (a) is from NOAA's National Geophysical Data Center [Global Land One-kilometer Base Elevation \(GLOBE\) Database](http://www.ngdc.noaa.gov/mgg/topo/report/globedocumentationmanual.pdf) (<http://www.ngdc.noaa.gov/mgg/topo/report/globedocumentationmanual.pdf>, (GLOBE Task Team and others (Hastings et al., 1999)). High-resolution elevation data in (b) is from ASTER GDEM, a product of METI and NASA.

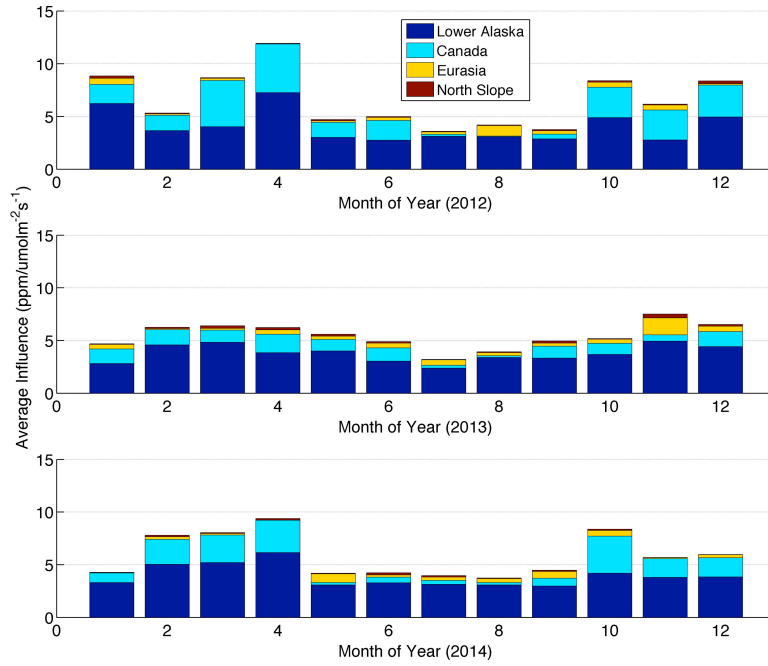
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Figure 2. Mean monthly diurnal cycle amplitude of hourly averaged CO₂ (top) and CH₄ (bottom). The average over three full calendar years (2012-2014) is shown in black with the gray shading indicating one standard deviation of each month's average. The average diurnal cycles for each individual year are indicated by the blue (2012), green (2013) and red (2014) solid lines.

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3 Figure 3. Magnitude of land surface influence on the tower measurements, in ppm/(μmol m⁻²
4 s⁻¹), for average monthly mid-afternoon footprints from the WRF-STILT model, for 2012
5 (top), 2013 (middle), and 2014 (bottom). Colors, as indicated in the figure legend, show the
6 average monthly surface influence of Lower Alaska (defined as any part of Alaska south of
7 the Brooks Range, i.e. not part of the North Slope), Canada, Eurasia, and the North Slope of
8 Alaska.

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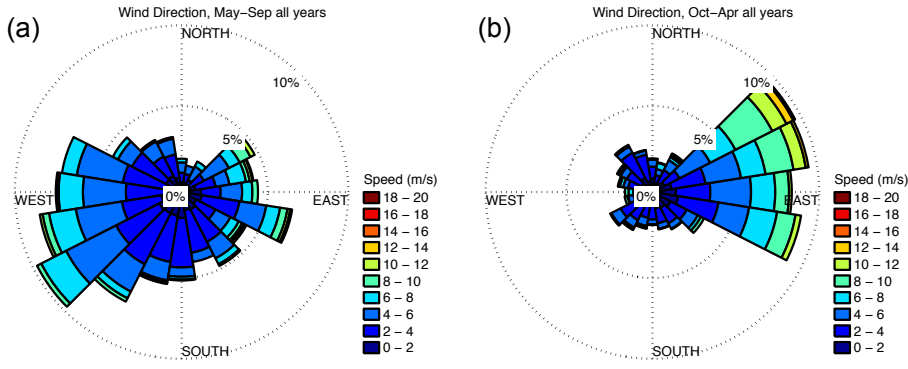
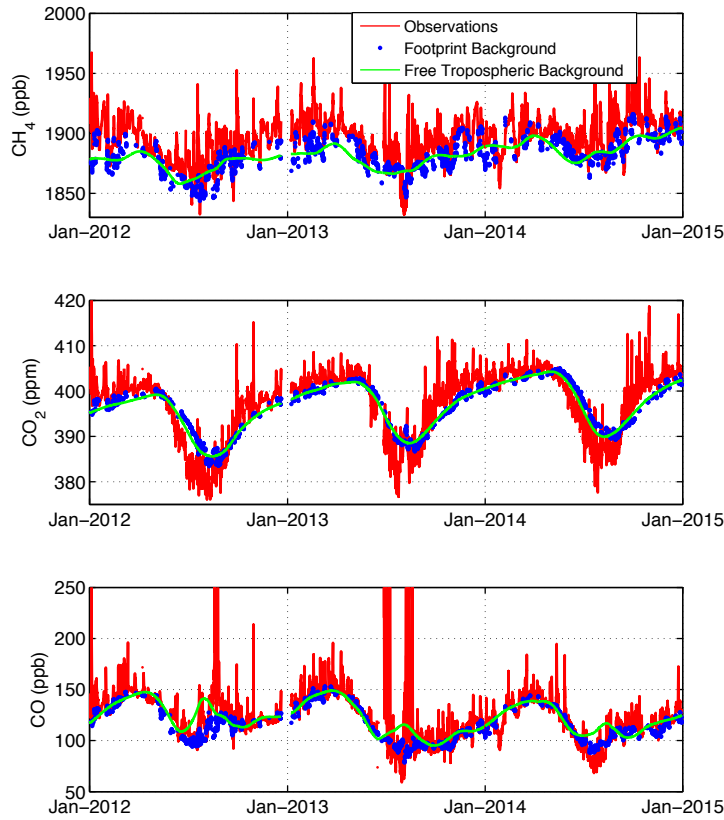


Figure 4. Wind roses for the tower averaged over (a) May-September and (b) October-April for all three years during mid-afternoon hours, from the 2D sonic anemometer at the 32 magl level of the tower.

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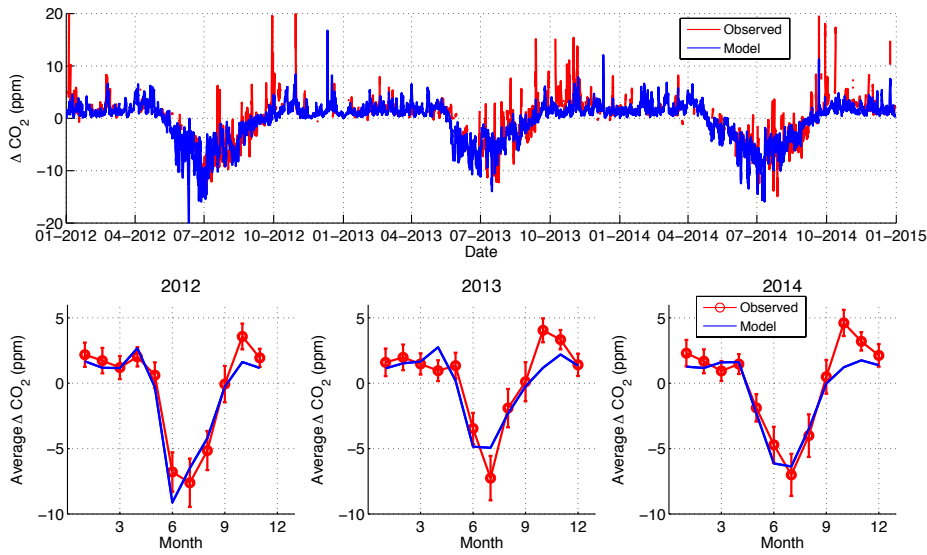


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3 Figure 5. Time series of hourly average observed mole fractions (red) and background mole
4 fractions (blue), 1 pm to 6 pm local standard time (LST) only for CH₄ (top), CO₂ (center), and
5 CO (lower) at the CARVE tower. Observations are indicated by solid red lines, while the
6 background mole fractions used for this analysis are shown in blue dots, and are derived using
7 the particle trajectories from the STILT model and an empirical Pacific boundary curtain,
8 described in the text. Gaps in the blue dots appear when the background could not be
9 calculated using the model because the air masses did not enter the domain from the West (as
10 described in Sect. 3.3). The green line represents the value of the same Pacific boundary
11 curtain at the site latitude (65°N) at 3500 meters above sea level, i.e. the free troposphere. The
12 vertical scale for CO has been truncated.

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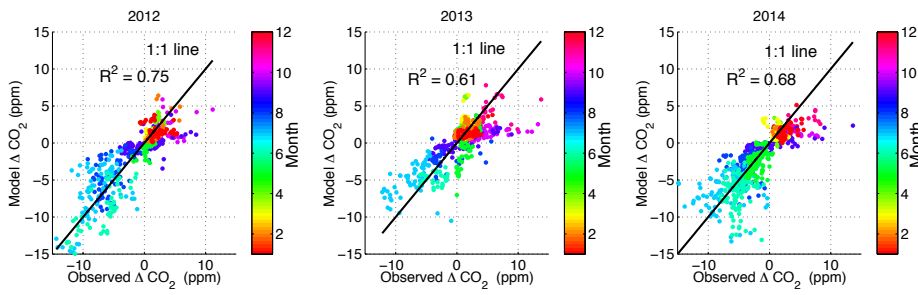


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3 Figure 6. Top: ΔCO_2 observed (i.e. observations minus background, red), along with the
4 modeled ΔCO_2 convolution (blue). Bottom: Monthly average comparisons between the model
5 (blue) and observations (red line and circles) for each year. Error bars on the observations
6 represent the average background uncertainty.

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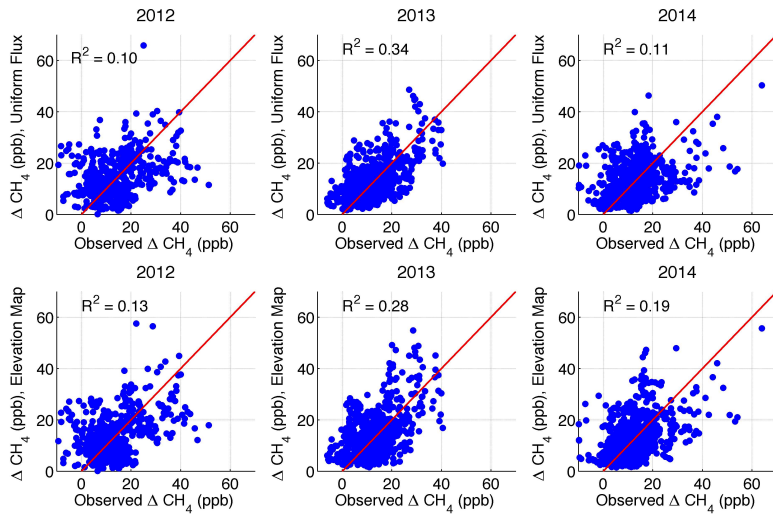
3 Figure 7. Correlation between observed and modeled ΔCO_2 , for 2012 (left), 2013 (center),
4 and 2014 (right), colored by month. The coefficient of determination, R^2 , is indicated in each
5 plot, along with the one-to-one line. Data points represent hourly averages between 1 pm and
6 6 pm LST and filtered according to criteria described in the text.

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3 Figure 8. Optimized model ΔCH_4 (ppb) for the scaled uniform flux (top row) and scaled
 4 elevation-based flux map (bottom row), for 2012 (left), 2013 (center), 2014 (right), all plotted
 5 against observed ΔCH_4 . Fluxes were scaled to match monthly average observed ΔCH_4 with
 6 monthly scaling factors. The coefficient of determination, R^2 , is indicated in each plot, along
 7 with the one-to-one line. Data points represent hourly averages between 1 pm and 6 pm LST
 8 and filtered according to criteria described in the text.

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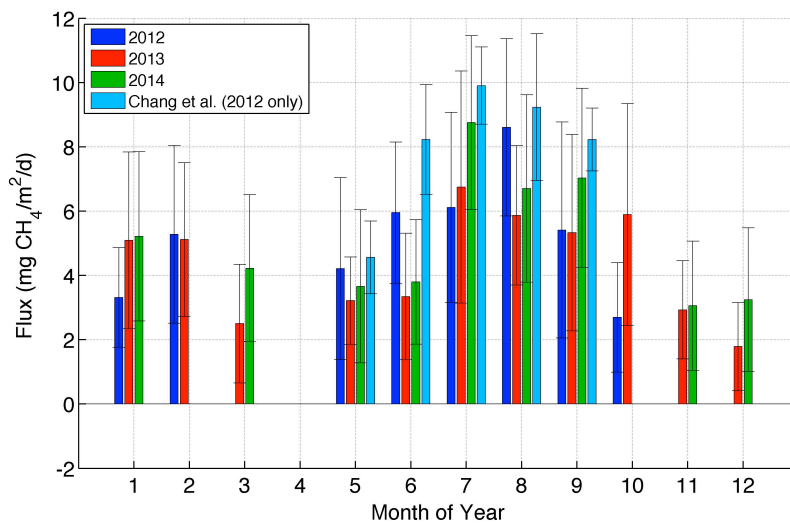
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4 Figure 9. Average Alaska monthly CH₄ fluxes for 2012 (blue), 2013 (red), and 2014 (green),
5 estimated based on a uniform Alaska-wide flux scaled to monthly mean observations at the
6 CRV tower. Light blue bars indicate monthly fluxes derived from 2012 CARVE aircraft
7 observations, with error bars representing the 68% confidence interval (CI), (calculated by
8 dividing the 95% CI by 1.96), from Chang et al. (2014). Error bars on the tower-derived
9 fluxes are based on propagating background uncertainty (1-sigma, Sect. 3.3) and uncertainty
10 derived from a sensitivity analysis on the altitude of the model runs (Sect. 3.2) only. Months
11 for which fluxes were based on six or fewer days were eliminated from the analysis.

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