Atmospheric
Chemistry
and Physics

Supplement of

Estimating regional-scale methane flux and budgets using CARVE aircraft measurements over Alaska

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S1 STILT footprint sensitivities

The STILT simulations assume that particles in the lower 50% of the boundary layer are directly influenced by the surface. A previous sensitivity study found that varying this percentage from 10–100% did not change the results significantly, although a lower value resulted in fewer particles actually influenced by the surface and an increase in noise (Gerbig et al., 2003). It should be noted that this sensitivity study was conducted over continental North America using different meteorological drivers and that these results may not be valid over our study domain, especially in the spring and fall. Nevertheless, it suggests that this assumption is not the major contributor to uncertainties in our results.

S2 Individual CH_4 profiles

The following Tables include the regional CH_4 flux estimations of Hartery et al. (2017) in tabulated form. Please refer to the main text for specific details such as the domain over which these estimates are valid and the measurements and particle transport modelling from which they were derived. Observer information is listed for all those profiles for which the estimate was determined to be unsuitable for analysis in three different messages. The first, "Large variability in FT CH_4," refers to an observed problem in constraining the background value of CH_4 in the free troposphere which is used to calculate the mixed layer enhancement of CH_4 (and subsequently the regional CH_4 flux). Since our method involves integrating observed profiles of mole fractions of CH_4 over the entire mixed layer, a large uncertainty in the background will result in a large uncertainty in CH_4 flux and therefore these profiles were withheld from further analysis.

The second observer message is, "Difficulty estimating BLH." This message is reported when the refractivity method for determining the mixed layer height significantly differed from observations of the profiles of water vapour, potential temperature and other trace gasses. It also appears when the boundary layer height predicted from modelling simulations differed from observations.

The third message which appears throughout these tables reports that "BL influence anomalously low." This message was used to indicate when the average integrated footprint influence throughout the mixed layer was less than 5% of the campaign average ($\bar{I} \sim 1$ ppm nmol^{-1} m^{-2} s^{-1}). When footprint influence is so low it is indicative that the air mass we measured was not predominantly of local origin and therefore not useful in deriving regional CH_4 estimates. This also results in overestimates of CH_4 flux as a consequence of our estimation method (see main text). The resulting profiles kept for analysis have been marked in the final column of each table by "incl."
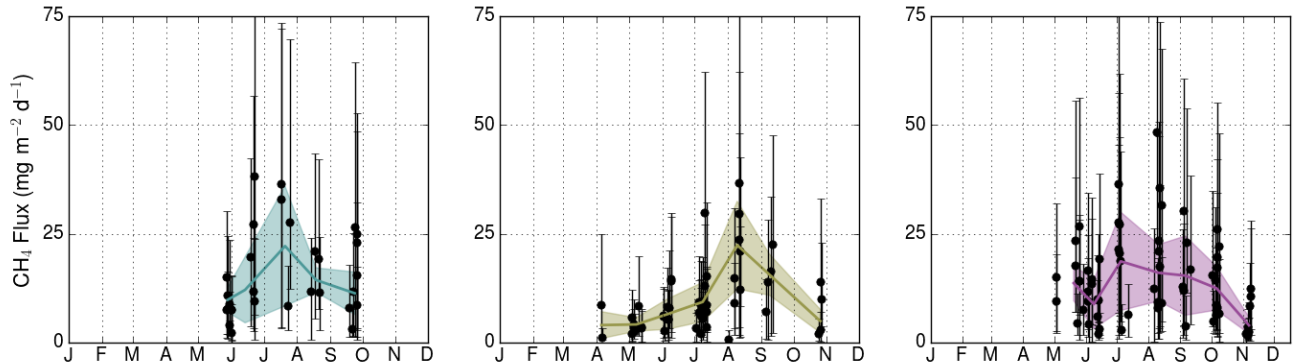
Figure S1: Individual estimates of CH_4 flux that were used in the analysis of the main text are shown overlaying the monthly average CH_4 fluxes. Error bars reflect uncertainties in individual flux estimates derived from Monte Carlo/bootstrap estimation.

As there can be considerable monthly variability among CH_4 flux estimates within a given month, we plot individual net flux estimates over the monthly averages. As such, Fig. S1 is a useful summary of Tables S1–S3; *n.b.*, only profiles included in the analysis are plotted. To highlight the fact that overall, the individual CH_4 flux estimates were normally distributed about the monthly mean CH_4 flux estimates, the residuals of

each individual estimate were plotted in a histogram in Fig. S2. The shape of this distribution gives us confidence that our monthly averaging is representative of the underlying seasonal trend.
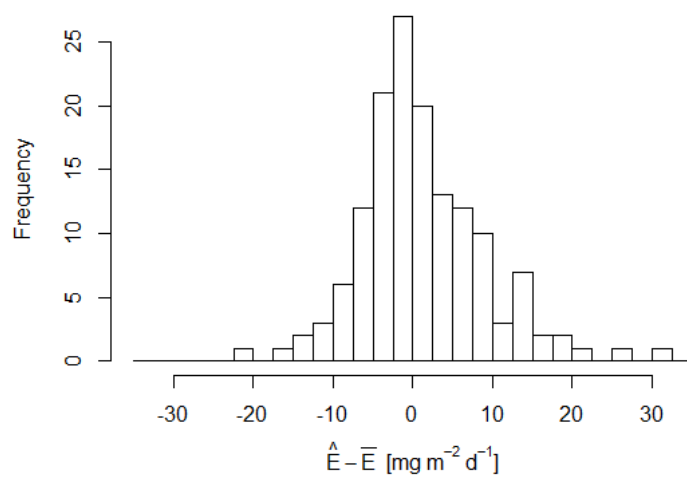
Figure S2: Residuals of individual CH_4 flux estimates with respect to monthly averages are shown in the histogram.

Table S1: CARVE 2012 regionally averaged CH$_4$ Fluxes are presented.

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
20120523	1	0.8	Large variability in FT CH$_4$	-
20120527	1	5.0	Difficulty estimating BLH	-
20120527	2	7.4		incl.
20120527	3	15.0		incl.
20120528	1	10.9		incl.
20120528	2	7.8		incl.
20120530	1	8.9		incl.
20120530	2	4.1		incl.
20120530	3	4.6	Large variability in FT CH$_4$	-
20120601	1	7.6		incl.
20120601	2	7.5		incl.
20120601	3	2.2		incl.
20120618	1	1.8	Large variability in FT CH$_4$	-
20120618	2	49.0	Large variability in FT CH$_4$	-
20120618	3	8.3	Large variability in FT CH$_4$	-
20120619	1	2.7	Large variability in FT CH$_4$	-
20120619	2	19.6		incl.
...	...	...	...	...

Table S1: (*cont.*)

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20120619	3	9.1	BL influence anomalously low, Large variability in FT CH$_4$	-
20120621	1	27.1		incl.
20120621	2	11.7		incl.
20120621	3	3.9	Difficulty estimating BLH	-
20120622	1	-6.2	Large variability in FT CH$_4$	-
20120622	2	43.1	Difficulty estimating BLH, BL influence anomalously low	-
20120622	3	38.1		incl.
20120622	4	9.5		incl.
20120624	1	-3.4	Large variability in FT CH$_4$	-
20120624	2	25.7	Difficulty estimating BLH, BL influence anomalously low	-
20120717	1	32.8		incl.
20120717	2	31.0	Difficulty estimating BLH	-
20120717	3	36.3		incl.
20120722	1	-7.9	Large variability in FT CH$_4$	-
20120722	2	7.3	Large variability in FT CH$_4$	-
20120722	3	-1.4	Large variability in FT CH$_4$	-
...	...	...	...	...

Table S1: (*cont.*)

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20120724	1	8.3		incl.
20120724	2	3.6	Large variability in FT CH$_4$	-
20120725	1	3.7	Large variability in FT CH$_4$	-
20120725	2	11.8	Large variability in FT CH$_4$	-
20120725	3	27.5		incl.
20120814	1	15.5	Large variability in FT CH$_4$	-
20120814	2	11.7		incl.
20120818	1	20.9		incl.
20120819	1	-19.0	BL influence anomalously low, Large variability in FT CH$_4$	-
20120819	2	8.2	Large variability in FT CH$_4$	-
20120819	3	-4.0	Difficulty estimating BLH	-
20120820	1	3.5	Large variability in FT CH$_4$	-
20120821	1	19.1		incl.
20120821	2	19.2	Difficulty estimating BLH	-
20120822	1	11.4		incl.
20120823	1	18.5	Large variability in FT CH$_4$	-
...	...	...	...	...

Table S1: *(cont.)*

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20120919	1	7.9		incl.
20120921	1	2.6	Difficulty estimating BLH	-
20120921	2	3.2		incl.
20120922	1	11.7		incl.
20120923	1	-77.2	Difficulty estimating BLH, BL influence anomalously low	-
20120924	1	26.4		incl.
20120924	2	53.3	Large variability in FT CH$_4$	-
20120926	1	8.6		incl.
20120926	2	24.8		incl.
20120926	3	15.4		incl.
20120926	4	22.9		incl.

Table S2: CARVE 2013 regionally averaged CH$_4$ Fluxes are presented.

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
20130402	1	2.0	Large variability in FT CH$_4$	-
20130403	1	0.6	Large variability in FT CH$_4$	-
20130404	1	18.4	Large variability in FT CH$_4$	-
20130404	2	83.8	BL influence anomalously low, Large variability in FT CH$_4$	-
20130404	3	28.4	Large variability in FT CH$_4$	-
20130405	1	8.7		incl.
20130406	1	4.2	Difficulty estimating BLH	-
20130406	2	2.9	Large variability in FT CH$_4$	-
20130406	3	1.0		incl.
20130502	1	10.9	Difficulty estimating BLH	-
20130502	2	4.2	Difficulty estimating BLH	-
20130504	1	2.1		incl.
20130504	2	5.7		incl.
20130506	1	3.4		incl.
20130506	2	4.2		incl.
20130507	1	2.6		incl.
20130507	2	1.0	Large variability in FT CH$_4$	-
...	...	...	...	...

Table S2: (*cont.*)

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20130507	3	-4.5	Large variability in FT CH$_4$	-
20130508	1	2.2	Difficulty estimating BLH	-
20130509	1	-9.6	Large variability in FT CH$_4$	-
20130509	2	-0.4	Large variability in FT CH$_4$	-
20130509	3	16.0	Large variability in FT CH$_4$	-
20130510	1	8.4		incl.
20130510	2	8.1	Large variability in FT CH$_4$	-
20130513	1	3.5		incl.
20130513	2	3.4		incl.
20130602	1	6.1		incl.
20130602	2	6.0		incl.
20130603	1	2.8		incl.
20130603	2	2.1	Large variability in FT CH$_4$	-
20130606	1	8.1		incl.
20130606	2	0.2	Difficulty estimating BLH	-
20130607	1	5.2		incl.
...	...	...	...	...

Table S2: (*cont.*)

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20130607	2	5.3		incl.
20130607	3	0.3	Difficulty estimating BLH	-
20130608	1	8.0		incl.
20130609	1	14.1		incl.
20130609	2	14.5		incl.
20130611	1	3.4	Difficulty estimating BLH	-
20130611	2	8.8	BL influence anomalously low, Large variability in FT CH$_4$	-
20130611	3	-86.6	Difficulty estimating BLH, BL influence anomalously low	-
20130703	1	3.2		incl.
20130703	2	-3.1	Difficulty estimating BLH	-
20130704	1	3.0	Difficulty estimating BLH	-
20130705	1	9.3		incl.
20130705	2	6.8		incl.
20130705	3	0.6	Large variability in FT CH$_4$	-
20130707	1	8.5		incl.
20130707	2	-3.2	Large variability in FT CH$_4$	-
...	...	...	...	...

Table S2: (*cont.*)

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20130707	3	2.0		incl.
20130707	4	7.2		incl.
20130707	5	6.2		incl.
20130709	1	9.4		incl.
20130709	2	5.5		incl.
20130709	3	8.7		incl.
20130711	1	15.8	Difficulty estimating BLH	-
20130711	2	29.9		incl.
20130711	3	13.0		incl.
20130712	1	3.6		incl.
20130712	2	8.7		incl.
20130712	3	7.2		incl.
20130712	4	15.1		incl.
20130802	1	0.7		incl.
20130802	2	6.0	Difficulty estimating BLH	-
20130803	1	6.1	Difficulty estimating BLH, BL influence anomalously low	-
...	...	...	...	...

Table S2: (*cont.*)

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20130803	2	13.3	Large variability in FT CH$_4$	-
20130803	3	-0.5	Large variability in FT CH$_4$	-
20130804	1	5.3	Large variability in FT CH$_4$	-
20130807	1	-14.7	BL influence anomalously low, Large variability in FT CH$_4$	-
20130807	2	9.0		incl.
20130807	3	14.9		incl.
20130811	1	9.8	BL influence anomalously low, Large variability in FT CH$_4$	-
20130811	2	25.2	Large variability in FT CH$_4$	-
20130811	3	16.8	Large variability in FT CH$_4$	-
20130812	1	23.7		incl.
20130812	2	36.7		incl.
20130812	3	29.6		incl.
20130813	1	41.9	Large variability in FT CH$_4$	-
20130813	2	20.8		incl.
20130813	3	12.2		incl.
20130905	1	-0.4	Large variability in FT CH$_4$	-
...	...	...	...	...

Table S2: (*cont.*)

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20130906	1	3.3	Large variability in FT CH$_4$	-
20130906	2	6.9		incl.
20130906	3	11.7	Difficulty estimating BLH	-
20130907	1	13.9		incl.
20130910	1	11.4	Difficulty estimating BLH	-
20130910	2	16.4		incl.
20130912	1	22.5		incl.
20131024	1	47.0	Large variability in FT CH$_4$	-
20131024	2	14.3	Large variability in FT CH$_4$	-
20131025	1	2.0		incl.
20131026	1	13.8		incl.
20131026	2	2.8		incl.
20131027	1	10.0		incl.
20131027	2	7.2	Difficulty estimating BLH, BL influence anomalously low	-

Table S3: CARVE 2014 regionally averaged CH$_4$ Fluxes are presented.

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
20140503	1	15.1		incl.
20140503	2	-7.8	Large variability in FT CH$_4$	-
20140503	3	9.4		incl.
20140522	1	23.3		incl.
20140522	2	17.7		incl.
20140523	1	35.8	Large variability in FT CH$_4$	-
20140524	1	12.0	Difficulty estimating BLH	-
20140524	2	4.4		incl.
20140526	1	-0.7	Difficulty estimating BLH	-
20140526	2	14.0		incl.
20140526	3	14.2		incl.
20140526	4	26.7		incl.
20140529	1	7.5		incl.
20140603	1	11.7		incl.
20140603	2	9.5		incl.
20140603	3	16.5		incl.
20140604	1	4.3		incl.
...	...	...	...	...

Table S3: (*cont.*)

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20140607	1	13.4		incl.
20140607	2	14.5		incl.
20140607	3	0.2	Large variability in FT CH$_4$	-
20140611	1	2.0	Difficulty estimating BLH	-
20140611	2	2.6	Difficulty estimating BLH	-
20140611	3	1.8	Large variability in FT CH$_4$	-
20140612	1	5.9		incl.
20140612	2	9.8		incl.
20140613	1	3.4	Difficulty estimating BLH	-
20140613	2	1.9		incl.
20140613	3	9.6		incl.
20140614	1	-1.2	Large variability in FT CH$_4$	-
20140614	2	19.2		incl.
20140614	3	3.1		incl.
20140703	1	3.5	Difficulty estimating BLH	-
20140703	2	36.3		incl.
...	...	...	...	...

Table S3: (*cont.*)

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20140703	3	27.6		incl.
20140703	4	21.3		incl.
20140704	1	27.0		incl.
20140704	2	20.5		incl.
20140705	1	5.0	Large variability in FT CH$_4$	-
20140705	2	18.8		incl.
20140705	3	-1.2	Difficulty estimating BLH	-
20140706	1	2.9		incl.
20140706	2	3.1	Difficulty estimating BLH	-
20140706	3	12.9	Difficulty estimating BLH	-
20140710	1	0.3	Large variability in FT CH$_4$	-
20140711	1	1.1	Large variability in FT CH$_4$	-
20140712	1	6.4		incl.
20140712	2	6.5	Difficulty estimating BLH	-
20140806	1	14.6	Difficulty estimating BLH	-
20140806	2	12.4		incl.
...	...	...	...	...

Table S3: (*cont.*)

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20140809	1	48.3		incl.
20140809	2	24.6	Difficulty estimating BLH	-
20140810	1	1.0	Difficulty estimating BLH	-
20140810	2	9.3		incl.
20140810	3	8.0		incl.
20140811	1	20.9		incl.
20140811	2	23.5		incl.
20140812	1	47.7	Difficulty estimating BLH, BL influence anomalously low	-
20140812	2	17.4		incl.
20140812	3	35.4		incl.
20140813	1	29.8	Large variability in FT CH$_4$	-
20140813	2	42.3	Large variability in FT CH$_4$	-
20140814	1	7.6	Difficulty estimating BLH	-
20140814	2	9.1		incl.
20140814	3	31.6		incl.
20140903	1	1.6	Large variability in FT CH$_4$	-
...	...	...	...	...

Table S3: (*cont.*)

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20140903	2	12.8		incl.
20140904	1	52.5	Large variability in FT CH$_4$	-
20140904	2	30.1		incl.
20140904	3	12.0		incl.
20140905	1	0.7	Large variability in FT CH$_4$	-
20140905	2	-3.0	Large variability in FT CH$_4$	-
20140906	1	9.8	Difficulty estimating BLH, BL influence anomalously low	-
20140906	2	3.7		incl.
20140906	3	22.7	Large variability in FT CH$_4$	-
20140907	1	23.0		incl.
20140907	2	0.6	Large variability in FT CH$_4$	-
20140907	3	453.4	Difficulty estimating BLH, BL influence anomalously low	-
20140909	1	3.7	Difficulty estimating BLH, BL influence anomalously low	-
20140911	1	5.5	Difficulty estimating BLH	-
20140911	2	6.2	Difficulty estimating BLH	-
20140911	3	3.0	Difficulty estimating BLH	-
...	...	...	...	...

Table S3: (*cont.*)

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20140911	4	16.8		incl.
20140911	5	12.2	Difficulty estimating BLH	-
20141002	1	15.4		incl.
20141002	2	11.0	Large variability in FT CH$_4$	-
20141003	1	4.8		incl.
20141003	2	7.7	Difficulty estimating BLH	-
20141005	1	3.3	Large variability in FT CH$_4$	-
20141005	2	14.3		incl.
20141006	1	6.4		incl.
20141006	2	8.5		incl.
20141006	3	8.2	Large variability in FT CH$_4$	-
20141007	1	19.7		incl.
20141007	2	26.1		incl.
20141007	3	17.1		incl.
20141007	4	8.0		incl.
20141009	1	6.5		incl.
...	...	...	...	...

Table S3: (*cont.*)

YYYYMMDD	Profile #	Flux (mg m^{-2} d^{-1})	Observer Information	Final Analysis
...	...	...	...	...
20141009	2	22.1		incl.
20141104	1	2.0		incl.
20141106	1	1.6		incl.
20141106	2	3.6		incl.
20141106	3	2.6		incl.
20141107	1	2.4		incl.
20141107	2	-4.4	BL influence anomalously low, Large variability in FT CH$_4$	-
20141107	3	31.6	Difficulty estimating BLH, BL influence anomalously low	-
20141108	1	10.0	Difficulty estimating BLH, BL influence anomalously low	-
20141108	2	8.4		incl.
20141109	1	12.3		incl.
20141109	2	10.6		incl.

S3 Methodological uncertainties from bootstrapping / Monte Carlo analysis

Table S4: Methodological 95% confidence intervals as described in Sect. 3.7

Parameter	Mean	Minimum	Maximum
	(mg m^{-2} d^{-1})	(mg m^{-2} d^{-1})	(mg m^{-2} d^{-1})
Observations	0.7	0.05	3.7
Model initial conditions	1.2	0.05	8
h for integration	0.9	0.05	4
Free tropospheric [CH$_4$]	3.7	0.2	14
All of the above	5.2	0.6	15

S4 Monthly fluxes and uncertainties

As discussed in the main text, the monthly average CH$_4$ fluxes shown in Fig. 4 are calculated by averaging the individual estimates of CH$_4$ flux into monthly bins. The averaging is adjusted by weighting each individual estimate by the column integrated total surface influence, such that estimates which have a larger degree of spatial coverage are weighted more heavily. In the following tables, the monthly average fluxes are tabulated for reference. In adjacent columns, uncertainties in the monthly average are calculated in four different ways: *weighted average uncertainty*, the 95% C.I. for each individual estimate is averaged for each month and weighted by the column integrated total surface influence; *average uncertainty*, the 95% C.I. for each individual estimate is averaged for each month with no weighting; *weighted standard deviation*, the standard deviation of all CH$_4$ flux estimates within a given month is calculated, weighting each residual by the footprint and the square of the 95% C.I.; and *standard deviation*, the normal standard deviation is calculated from the residuals of the weighted monthly mean. To be more explicit about the weighted standard deviation, for each month the set of residuals $(\hat{E}_i - \overline{E})$ is weighted by:

$$w_i = \frac{\Delta I}{\text{C.I.}_i^2} \tag{1}$$

where ΔI is the column integrated surface influence and C.I.$_i$ is the 95% C.I. of the ith CH$_4$ flux estimate. The weighted standard deviation is then calculated according to:

$$\sigma = \sqrt{\frac{N \sum_i w_i (\hat{E}_i - \overline{E})^2.}{(N-1) \sum_i w_i}} \tag{2}$$

where N is the number of profiles for a given month. At the end of each table, the integrated May–September budgets are presented as well as the propagated uncertainty using each method listed above.

Table S5: CARVE 2012 monthly averaged CH_4 fluxes and uncertainties (mg m^{-2} d^{-1}) are presented.

Month	CH_4 Flux	Uncertainty (weighted average)	Uncertainty (average)	Standard Deviation (weighted)	Standard Deviation
1	–	–	–	–	–
2	–	–	–	–	–
3	–	–	–	–	–
4	–	–	–	–	–
5	9.8	3.1	4.4	2.9	3.5
6	12.1	3.5	4.4	7.6	11.8
7	22.2	8.4	11.3	13.8	11.5
8	14.3	4.9	6.1	2.8	4.5
9	11.3	3.9	4.8	4.9	9.0
10	–	–	–	–	–
11	–	–	–	–	–
12	–	–	–	–	–
Budgets & Uncertainty (Tg):	2.2	0.4	0.5	0.5	0.6

Table S6: CARVE 2013 monthly averaged CH_4 fluxes and uncertainties (mg m^{-2} d^{-1}) are presented.

Month	CH_4 Flux	Uncertainty (weighted average)	Uncertainty (average)	Standard Deviation (weighted)	Standard Deviation
1	–	–	–	–	–
2	–	–	–	–	–
3	–	–	–	–	–
4	4.0	4.5	5.2	3.1	3.9
5	4.2	2.2	2.0	1.6	1.9
6	6.5	2.1	2.7	3.5	4.0
7	9.3	3.9	3.6	4.5	6.3
8	22.4	5.0	4.5	10.1	11.6
9	15.6	2.6	2.4	4.7	5.6
10	5.0	3.3	4.7	2.7	5.4
11	–	–	–	–	–
12	–	–	–	–	–
Budgets & Uncertainty (Tg):	1.9	0.2	0.2	0.4	0.5

Table S7: CARVE 2014 monthly averaged CH_4 fluxes and uncertainties (mg m^{-2} d^{-1}) are presented.

Month	CH_4 Flux	Uncertainty (weighted average)	Uncertainty (average)	Standard Deviation (weighted)	Standard Deviation
1	–	–	–	–	–
2	–	–	–	–	–
3	–	–	–	–	–
4	–	–	–	–	–
5	13.7	6.3	7.6	4.1	6.9
6	8.8	3.6	4.1	5.6	5.3
7	18.7	7.7	8.0	11.3	10.4
8	16.0	4.7	9.1	6.5	13.8
9	15.4	6.5	7.3	9.1	8.5
10	12.5	5.8	6.3	5.0	6.9
11	4.0	1.9	3.7	1.9	4.3
12	–	–	–	–	–
Budgets & Uncertainty (Tg):	2.3	0.4	0.5	0.6	0.7

S5   Constraining the CH_4 background

In order to estimate the regional CH_4 flux from the aircraft observations presented in the main text, it is necessary to first estimate the natural background of CH_4 in the atmosphere. To achieve this, the CARVE flight patterns were planned to include flying up to 5–6 km a.g.l. so as to sample air well above the planetary boundary layer and any local influence. For each of the resulting profiles of measured CH_4 mixing ratios we make the assumption that the average mixing ratio observed in the layer from $500 - 1\,500$ m above the top of the boundary layer is representative of the background CH_4 mixing ratio in the boundary layer. To assess the accuracy of this claim, we compare these averages to CH_4 backgrounds observed in the Alaskan boundary layer from two ground stations. These comparisons are shown in Figure 1 and contrast the background CH_4 we estimate from free tropospheric observations, the background CH_4 observed at Barrow, Alaska (Dlugogencky, 2016), and the background CH_4 derived from particle simulations and observations at the CARVE tower near Fairbanks, Alaska (Karion et al., 2016). These two sites have the advantage that they are situated in the two ecosystems of interest to the current study and have coverage across the entire campaign (with the exception of the Barrow tower in 2012). Overall we can clearly observe that the backgrounds we estimate from free tropospheric observations are certainly within the observations of both ground stations. It is useful to highlight that a latitudinal gradient in CH_4 has been observed across the ESRL network, and as our flights cover a wide latitudinal range, it is not surprising that monthly variability (shown as the shaded area around the CARVE aircraft observations) covers the difference between Fairbanks and Barrow.
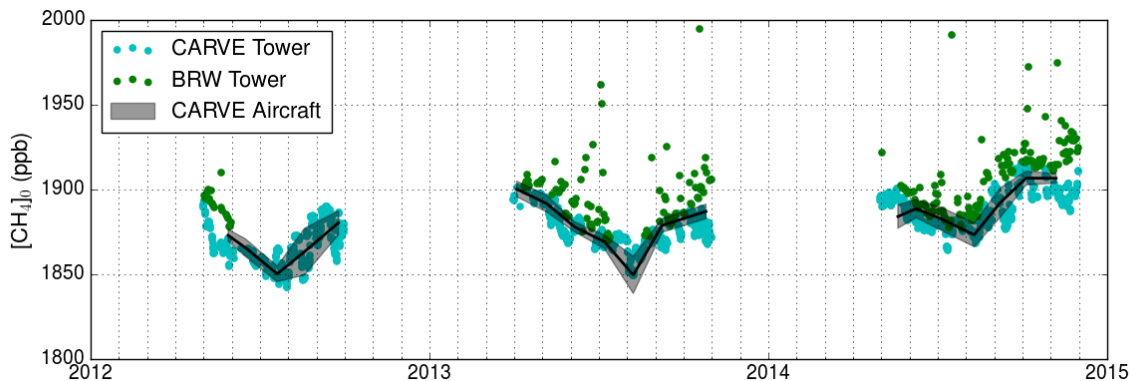

Figure S3: Comparison of monthly average free tropospheric mixing ratios of CH_4 observed during the CARVE campaign to boundary layer background CH_4 observed at Barrow, Alaska (Dlugogencky, 2016) and derived from observations at the CARVE Tower near Fairbanks Alaska (Karion et al., 2016).

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