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# Methane emissions from Alaska in 2012 from CARVE airborne observations

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We determined methane (CH<sub>4</sub>) emissions from Alaska, USA using airborne measurements from the Carbon Arctic Reservoirs Vulnerability Experiment (CARVE). Atmospheric sampling was conducted between May and September 2012, and analyzed using a customized version of the Polar Weather Research and Forecast model linked to a Lagrangian particle dispersion model (Stochastic Time-Inverted Lagrangian Transport Model). We estimated growing season CH<sub>4</sub> fluxes of  $8 \pm 2 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$  averaged over all of Alaska, corresponding to fluxes from wetlands of  $56^{+22}_{-13} \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$  if we assumed that wetlands are the only source from the land surface (all uncertainties are 95% confidence intervals from a bootstrapping analysis). Fluxes roughly doubled from May to July, then decreased gradually in August and September. Integrated emissions totaled  $2.1 \pm 0.5 \text{ Tg CH}_4$  for Alaska from May to September 2012, close to the average (2.3, range 0.7–6 Tg CH<sub>4</sub>) predicted by various land surface models and inversion analyses for the growing season. Methane emissions from boreal Alaska were larger than from the North Slope; the monthly regional flux estimates show no evidence of enhanced emissions during early spring or late fall, although these bursts may be more localized in time and space than can be detected by our analysis. These results provide an important baseline to which future studies can be compared.

Methane | Alaska | Tundra | Arctic | Boreal

## Significance Statement

Alaska emitted  $2.1 \pm 0.5 \text{ Tg CH}_4$  during the 2012 growing season, an unexceptional amount despite widespread permafrost thaw and other evidence of climate change in the region. Our results are based on more than 30 airborne measurement flights conducted by the Carbon in Arctic Reservoirs Vulnerability Experiment from May to September 2012 over Alaska. Methane emissions peaked in summer and remained high in the fall. Emissions from boreal regions were notably larger than from North Slope tundra. This is the first regional study of methane emissions from Arctic and boreal regions over a growing season. Our estimates reinforce and refine global models, and they provide an important baseline against which to measure future changes associated with climate change.

## Introduction

Recent studies have raised concerns about an increase in methane (CH<sub>4</sub>) emissions from Arctic regions as temperatures warm [1, 2, 3]. Carbon stocks in polar regions are estimated to be as large as 1700 Pg of organic carbon [4], preserved by cold, wet conditions that inhibit decomposition. Over the last 20 years, temperatures have increased more rapidly at these latitudes than the rest of the world [5]; continuation of this trend will lead to permafrost warming and thawing [6], potentially releasing vast quantities of carbon dioxide (CO<sub>2</sub>) and CH<sub>4</sub> to the atmosphere [7, 8, 9, 10]. A recent synthe-

sis of carbon emissions predicted by permafrost models reported releases in the range of  $120 \pm 85 \text{ Pg C}$  by 2100 [11]. Large uncertainties are likewise associated with estimates of CH<sub>4</sub> emissions ( $12\text{--}90 \text{ Tg CH}_4 \text{ yr}^{-1}$ ) [12]. The potential for large increases in CH<sub>4</sub> emissions are a particular concern since CH<sub>4</sub> strongly impacts both atmospheric chemistry and climate [13]. Estimates of the impact of permafrost carbon emissions on future global temperatures range from  $\sim 0.1\text{--}0.2^\circ\text{C}$  [14] to  $0.3 \pm 0.2^\circ\text{C}$  [11] by 2100, with increased carbon emissions expected to continue after 2100 [11].

Recent global inversion studies find no evidence for increasing CH<sub>4</sub> emissions from these regions in the last 10 years [15, 16], despite warming, as indicated by earlier studies [17, 18, 19] and some biogeochemical models [14]. Surface flux observations in the pan-Arctic during 1996–2000 have ranged widely and measurement locations have changed, making it difficult to detect any trend over those years [20], cf. [21].

The present paper derives estimates of CH<sub>4</sub> surface fluxes in Alaska from May to September 2012, based on an extensive program of regional-scale airborne measurements of atmospheric CH<sub>4</sub>, the Carbon in Arctic Reservoirs Vulnerability Experiment (CARVE). We quantify the monthly mean CH<sub>4</sub> emissions from Alaska during the growing season, providing a snapshot of the interactions between climate and the vast reservoir of preserved organic matter in the Arctic.

## Methods

**Measurements.** Measurements were made on board a NASA C-23B aircraft (N430NA) during the last two weeks of each month between May and September 2012. Flights were based in Fairbanks, Alaska, USA and ranged from  $60.21\text{--}71.56^\circ\text{N}$  and  $164.5\text{--}143.6^\circ\text{W}$ , covering three major regions: 1) the North Slope, which included transits to Barrow and Deadhorse on the northern coast; 2) the Lower Yukon region following the course of the Yukon river south and west of Fairbanks, including the Yukon Delta National Wildlife Refuge (which includes the Yukon and Kuskokwim deltas) and the Innoko National

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Wildlife Refuge; and 3) the Upper Yukon region which included the Yukon Flats National Wildlife Refuge (grey points in Fig. 1). Each flight lasted 4–10 hours, with the majority of sampling occurring below 200 m above ground level (agl).  
 100 One or more vertical profiles reaching a maximum of 5500 m above sea level (asl) were flown during each flight, with the maximum height determined by weather conditions. In total, 200 flight hours were flown over 31 flight days.

Two independent cavity ringdown spectrometers measured *in situ* greenhouse gas mole fractions every ~2.5 s with two separate on board calibration standards for each unit. The first spectrometer measured CO<sub>2</sub>, CH<sub>4</sub> and H<sub>2</sub>O (Picarro, G1301-m) directly from the inlet. This sensor sampled one of the two calibration gas cylinders every 30 min and is similar to the instrument described by Karion et al. [22]. For the second instrument, ambient air first passed through a Nafion dryer followed by a dry ice trap which effectively lowered the dewpoint to approximately 195 K, before being sampled by the spectrometer. This sensor reported CO<sub>2</sub>, CH<sub>4</sub> and carbon monoxide (CO) mixing ratios (Picarro, G2401-m) and sampled both its calibration cylinders every 30 min. The time series used in our analysis merge the CH<sub>4</sub> data from these two instruments, enabling us to fill in gaps when an instrument was calibrating or malfunctioning. Further discussion on the comparison of these two instruments can be found in the SI. Other relevant measurements made on board include ozone (O<sub>3</sub>) mixing ratios (2B Technologies, model 205), dewpoint temperature (Edgetech, Vigilant), outside air temperature (Harco, 100366-18), pressure (Paroscientific, 745-15A) and location using a global positioning unit (Crossbow, NAV420).  
 125

**Model description.** Aircraft measurements were aggregated horizontally every 5 km and vertically in 50 m intervals below 1 km asl and 100 m intervals for measurements above 1 km, giving ~23,000 data points. Each of these points at (x,y,z,t) was treated as a receptor for the Stochastic Time-Inverted Lagrangian Transport (STILT) model [23], which traces the trajectory of the air parcel at each receptor location backward in time over the preceding 10 days and quantifies in space and time where upstream surface fluxes influenced the measured concentrations. Particles are advected by the large-scale (i.e. explicitly resolved) wind field, as simulated by the Advanced Research version of the Weather Research and Forecasting (WRF) model (v3.4.1) [24] on a 3.3 km grid in the innermost domain over Alaska, plus stochastic motions to simulate turbulence. To improve prediction of the meteorological fields in the Arctic, basic options from the Polar variant of WRF [25, 26, 27] were implemented. A two-dimensional influence field (“footprint”) is available for each particle every 3 h over its 10 day travel period, representing the response of the receptor to a unit emission of tracer at each grid square (converted unit of ppb / (mg m<sup>-2</sup> d<sup>-1</sup>)). The footprints used in this analysis were on a 0.5° × 0.5° grid. Further details of both the WRF and STILT models can be found in Henderson et al. [28]. Figure S1 shows the sum of all footprints for the vertical profiles (see below) used in the analysis.  
 150

**CH<sub>4</sub> fluxes derived from column analysis.** Our primary analysis focuses on applying the WRF-STILT framework to the partial column integrals of CH<sub>4</sub> mole fractions measured during vertical profiles, subtracting the background value for air flowing in from outside the study region (the State of Alaska). This “column enhancement” represents the mass loading of the atmosphere from the ground to the top of the residual layer (the maximum height influenced by surface emissions during transit from the boundary) due to emissions in the region. The advantage of this approach is that results are only dependent  
 160

on the large scale simulation of the vertical structure of the atmosphere, reducing our reliance on the detailed structure of the boundary and residual layers, fine scale variations of emissions at the surface, and turbulent transport elements in the lower atmosphere.

Atmospheric column enhancements have been used in previous studies of CO<sub>2</sub> in the Amazon [29, 30], based on the concept that this quantity measures the total amount of trace gas added to the atmosphere during the transit of an air mass over the land. Similar to Chou et al. [29], we used the CH<sub>4</sub> mole fraction measured at the top of the residual layer height as our background value. The top of the residual layer is effectively equivalent to the bottom of the free troposphere and was identified by comparing the vertical profiles of CH<sub>4</sub>, CO<sub>2</sub>, CO, O<sub>3</sub> and water vapor (*P*<sub>H<sub>2</sub>O</sub>). For each vertical profile, the height at which the slope changes sign for each chemical compound was compared and used to determine the residual layer height for that profile. The height at which Alaskan land ceased to influence the column was also assessed using WRF-STILT and contributed to the identification of the residual layer height when there were discrepancies between different chemical compounds. The dashed purple line in Fig. 2 shows the top of the residual layer for a sample profile. Vertical profiles over Alaska from the NOAA measurements on board the Alaska Coast Guard flights [22] during this same period were consistent with the inferred background concentrations.  
 185

Column enhancements below the residual layer height (*E*<sub>CH<sub>4</sub>,obs</sub>) were calculated by block averaging the observed CH<sub>4</sub> mole fraction ([CH<sub>4</sub>]) from each vertical profile into 250 m altitude bins, subtracting the concentration at the top of the residual layer ([CH<sub>4</sub>](*h*)) and then integrating the density-weighted concentration enhancements:

$$E_{CH_4,obs} = \int_0^h ([CH_4](z) - [CH_4](h)) \times \frac{P_{air}(z) - P_{H_2O}(z)}{RT(z)} dz,$$

where *P*<sub>air</sub>, *T* and *R* are the ambient pressure, temperature and universal gas constant, respectively. The column enhancement is illustrated by the black hatch in Fig. 2A. A similar calculation is used to determine the column enhancement from WRF-STILT assuming a unit flux from land *E*<sub>CH<sub>4</sub>,unit</sub>. The mean surface flux associated with each profile (*F*<sub>CH<sub>4</sub>,VP<sub>i</sub></sub>) is then calculated as  $\overline{F_{CH_4,VP_i}} = E_{CH_4,obs} / E_{CH_4,unit}$ . The overall mean was calculated by averaging the  $\overline{F_{CH_4,VP_i}}$  for all vertical profiles weighted by their corresponding footprints. Monthly means were calculated in a similar manner but using only profiles from that month. A comparison of surface influences between profiles can be seen in Fig. S2. The red hatch in Fig. 2A shows the modeled column enhancement calculated from the mean monthly surface flux determined from the bootstrapping analysis described below. The mean emission for a given region ( $\overline{F_{CH_4,A}}$ , where A is the region of interest) is determined by weighing  $\overline{F_{CH_4,VP_i}}$  for every vertical profile by the portion of the corresponding footprint influence in that region (*I*<sub>A,VP<sub>i</sub></sub>), such that

$$\overline{F_{CH_4,A}} = \frac{\sum_i \overline{F_{CH_4,VP_i}} \times I_{A,VP_i}}{\sum_i I_{A,VP_i}}.$$

To determine the uncertainties in the derived fluxes, observed parameters used in the calculation (measured mole fraction, pressure, temperature, water vapor) were bootstrapped by randomly sampling 1000 times with replacement at each 250 m altitude bin. The residual layer height, which also determines the background concentration, was also sampled 1000 times assuming a uniform probability of the true residual layer height being ± 500 m of the determined height. A second

195 method of determining the uncertainty compared the calcu-  
 lated mean flux with  $F_{CH_4,VP_i}$  for each vertical profile. Figure  
 S2 in the SI shows this comparison with the mean monthly  
 fluxes. Results are similar for the overall mean. The aver-  
 age uncertainty from this method lies within the uncertainty  
 200 determined from our bootstrapping analysis.

Of the 50 vertical profiles from the 2012 campaign, 30 were  
 well-suited for deriving  $CH_4$  flux from the land surface in  
 Alaska (locations shown in black points in Fig. 1 and times  
 given in Table S1). Profiles were rejected due to a) influences  
 205 by biomass burning (increase in CO of at least 40 ppb within  
 the residual layer) (four profiles); b) significant land influences  
 (>30%) from outside the CARVE study region, usually from  
 Siberia (10 profiles); or c) undefined residual layer, either be-  
 cause the maximum height of the aircraft was too low or the  
 210 atmospheric structure was too complex for this simple analysis  
 (six profiles).

**Land elevation categories derived from ecoregions.** The  
 United States Geological Survey and Environmental Protec-  
 tion Agency identifies 20 Level III ecoregions in Alaska [31].  
 215 For the purposes of our  $CH_4$  surface-atmosphere flux calcu-  
 lations, these 20 ecoregions were grouped into four categories  
 based on elevation: Highlands (plateaus and uplands); Low-  
 lands (plains, lowlands and flats); the North Slope (Arctic  
 coastal plain and Arctic foothills); and Mountains (ranges and  
 220 mountains) (colored regions in Fig. 1, complete list in SI). This  
 grouping was used because  $CH_4$  fluxes depend on water table  
 depth and elevation [32, 33] and the atmospheric data in this  
 study cannot resolve all 20 ecoregions. The ecoregions were  
 gridded to  $0.5^\circ \times 0.5^\circ$  to match the resolution of the STILT  
 225 footprints.

## Results and Discussion

**Results of the column analysis.** The black circle in Fig. 3A  
 shows the overall mean  $CH_4$  flux estimates from Alaska if we  
 adopt a uniform emission rate for all land surfaces during each  
 230 month:  $8 \pm 2 \text{ mg } CH_4 \text{ m}^{-2} \text{ d}^{-1}$ , where the uncertainty is the  
 95% confidence interval from the bootstrapping analysis de-  
 scribed above. This baseline assessment does not reflect ac-  
 tual emissions at the surface, but it is determined independent  
 of any assumed surface map and is the most robust number  
 235 derived from our calculations. Flux estimates were also de-  
 termined if the Mountains category was assumed to not con-  
 tribute to  $CH_4$  emissions, which increases the flux from other  
 land types by  $\sim 25\%$  to  $10 \pm 2 \text{ mg } CH_4 \text{ m}^{-2} \text{ d}^{-1}$  (red triangle  
 in Fig. 3A). Uncertainties in Fig. 3 show the 95% confi-  
 240 dence interval derived from the bootstrapping analysis. These  
 flux estimates represent all land emission processes: biogenic,  
 anthropogenic, and geologic/thermogenic (including possible  
 thermogenic seeps arising from thawing permafrost [3]), but  
 exclude emissions from biomass fires and any ocean processes.  
 245 These fluxes correspond to an overall emission of  $2.1 \pm 0.5 \text{ Tg}$   
 $CH_4$  from May–September, 2012.

Mean fluxes for the entire study period were derived for the  
 three broad land categories (Highlands, North Slope and Low-  
 lands) as shown in Fig. 3A. The  $CH_4$  flux from the Lowlands  
 250 are consistently greater than from the Highlands, and both  
 of these regions emit significantly more  $CH_4$  than the North  
 Slope ( $p < 0.001$  in a paired t-test). This result is consistent  
 with the Lowlands being wetter than the Highlands and the  
 North Slope being cooler, with a thinner active soil layer, than  
 255 the other regions.

The seasonality of  $CH_4$  fluxes derived over the entire state  
 is shown in Fig. 3B and exhibits an increase in emissions from  
 May to July followed by a gradual decrease until September.

The overall range is only  $5 \text{ mg } CH_4 \text{ m}^{-2} \text{ d}^{-1}$ , which is weaker  
 260 than the  $14\text{--}80 \text{ mg } CH_4 \text{ m}^{-2} \text{ d}^{-1}$  difference that can be ob-  
 served over a season at ground sites [7, 34]. The  $CH_4$  column  
 enhancements sampled by the CARVE aircraft are influenced  
 by emissions from land types heterogeneous in elevation, soil  
 moisture, and organic substrate, as well as diverse seasonal  
 characteristics. (Even at altitudes below 200 m agl, footprints  
 can span a distance of  $> 500 \text{ km}$ .) The large sampling area  
 for each profile tends to dampen seasonal signals that may be  
 observed at individual ground sites with more coherent sea-  
 sonality.

The seasonal variation observed in our study is generally  
 consistent with other regions in North America [7, 34] and  
 with northern wetland emissions diagnosed from global in-  
 version studies [15, 16, 17]. We observe neither the pattern  
 observed at Zackenberg, Greenland, with high spikes in  $CH_4$   
 270 fluxes during the spring thaw and fall freeze up [35], nor as  
 predicted for the Yukon River Valley [36]. Sampling began be-  
 fore the spring thaw, so widespread bursts at that time should  
 have been seen, but it is possible that we did not sample late  
 enough in the season to capture  $CH_4$  bursts in the fall, or that  
 275 these bursts are more localized in time and space than can be  
 detected by our flight program.

**$CH_4$  fluxes estimated from  $CH_4$ : $O_3$  covariance.** We developed  
 a second independent method to estimate  $CH_4$  fluxes using the  
 observed covariance of  $CH_4$  and  $O_3$  in the lowest 1500 m of  
 the atmosphere. These flux estimates are independent of the  
 WRF-STILT footprints, and use the collected data merged  
 at 5 s, resulting in  $\sim 40,000$  data points rather than just the  
 vertical profiles. This method heavily weights the particular  
 flight tracks, and involves many simplifying assumptions; it is  
 290 included to check the order of magnitude of the estimates cal-  
 culated from the vertical profile analysis. Altitudes closest to  
 the surface can be treated as a constant flux layer, where con-  
 centration changes of a chemical compound are dominated by  
 surface exchange with little influence from atmospheric flux  
 divergence. Near the surface in the Arctic,  $O_3$  loss is domi-  
 nated by dry deposition and *in situ* chemistry can be ne-  
 glected [37, 38]. Similar to the column analysis, influences  
 from biomass burning were removed by excluding data when  
 absolute CO mole fractions exceeded 150 ppb [39]. At the scale  
 of our measurements, we can assume that  $O_3$  is effectively lost  
 through dry deposition from the same surfaces that emit  $CH_4$ ,  
 and we can use similarity theory to independently determine  
 $CH_4$  flux:  $F_{CH_4} = F_{O_3} \times (\Delta CH_4 / \Delta O_3)$ , where  $F_x$  is the flux  
 of compound  $x$ . Ozone flux is computed from the deposition  
 305 velocity ( $v_D$ ) as  $F_{O_3} = -v_D \times [O_3]_{500}$ , where  $[O_3]_{500}$  is the  
 average  $O_3$  mole fraction in the lowest 500 m agl. Figure 4  
 shows  $O_3$  and  $CH_4$  mole fraction deviations from 10 minute  
 means in the lowest 1500 m agl for June (see Fig. S3 for other  
 months). The slope of the line ( $\Delta O_3 / \Delta CH_4$ ) is determined us-  
 ing standard major axis regression [40] and is used to calculate  
 $F_{CH_4}$ , shown in the red circles of Fig. 5. We used a constant  
 $O_3$   $v_D = -0.3 \pm 0.1 \text{ cm s}^{-1}$ , as determined by Henderson et  
 al. [28] which is consistent with measurements reported over  
 fens, Scots pine forests and tundra [41, 42, 43]. Using this  $v_D$   
 with WRF-STILT footprints results in the modeled  $O_3$  shown  
 in the red triangles in Fig. 2B, reasonably consistent with  
 observations.

The domain-wide average  $F_{CH_4}$  from this method is esti-  
 mated to be  $15 \pm 5 \text{ mg } CH_4 \text{ m}^{-2} \text{ d}^{-1}$  for May–September 2012,  
 where the uncertainty reflects the range of  $O_3$   $v_D$  in the liter-  
 ature and the calculated  $v_D$  ( $-0.3 \pm 0.1 \text{ cm s}^{-1}$ ) [28]. Ozone  
 $v_D$  is expected to vary seasonally [43] since it is dependent  
 on the reactivity of  $O_3$  with leaves. Applying the seasonally-  
 varying  $v_D$  determined by Henderson et al. [28] (0.13, 0.28,

0.44, 0.35, 0.34 cm s<sup>-1</sup> for May–September 2012, respectively, with ~33% uncertainty) results in the estimated  $F_{CH_4}$  shown in the black triangles of Fig. 5 (mean=16±5 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>). The resulting seasonal cycle is not dissimilar to that calculated using the column analysis in Fig. 3 although the peak of the emissions is later. Overall, the CH<sub>4</sub> flux estimated from its covariance with O<sub>3</sub> is remarkably close to the mean value determined from all of Alaska if mountains were excluded (10 ± 2 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>), which is most comparable since we seldom flew near the surface in mountainous terrain. The general agreement between these two independent estimates of CH<sub>4</sub> fluxes increases our confidence in the overall analysis.

**Comparison with other flux observations.** Our regional flux estimates integrate over wet and dry areas uniformly, giving a more objective regional flux than upscaling from chambers or towers which are typically deployed in areas expected to be significant CH<sub>4</sub> sources. To compare our estimates with these other studies that are sensitive to smaller spatial scales, a distribution map [44] was used to infer the emission rate for wetlands, effectively restricting the areal extent from which CH<sub>4</sub> was emitted and assuming that other CH<sub>4</sub> sources are negligibly small. Resulting emissions are seven times higher than the overall regional mean (56<sup>+22</sup><sub>-13</sub> mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) and follow a similar seasonal pattern. This value is similar to CH<sub>4</sub> fluxes measured via airborne eddy covariance during the Arctic Boundary Layer Experiment which took place over the Yukon-Kuskokwim River Delta in southwest Alaska 28 July to 9 August, 1988 (51<sup>+34</sup><sub>-26</sub> mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> [45]).

Flux measurements determined from static chambers in Alaska range from 0–300 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> (compiled by Olefeldt et al. [46]), with a median over 90 studies of 49 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>, and eddy-covariance and gradient tower measurements in tundra regions range from 3–80 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>, with a median over 13 studies of 34 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> (see Table S1). A recent aircraft study over northern Sweden determined CH<sub>4</sub> fluxes equivalent to 29 ± 12 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> for a flight in July 2012 over extensive wetland areas [47]. Our values are consistent with these previous measurements once the sampling differences are taken into account.

**Comparison with models and inversion studies.** Our integrated CH<sub>4</sub> emission estimate of 2.1 ± 0.5 Tg CH<sub>4</sub> over May–September, 2012 falls within the 0.7–6 Tg CH<sub>4</sub> range of emissions estimated from an ensemble of ten different global bottom-up models for the same region and months (Table 1). Our findings are also consistent with the 1.5±0.2 Tg CH<sub>4</sub> estimated by Carbon Tracker-CH<sub>4</sub> [16] and the 1.3±0.3 Tg CH<sub>4</sub> estimated by TM5-4DVAR when biomass burning is excluded [15] for May–September. Our mean is very close to the mean of all the comparable values in Table 1 (2.1 vs. 2.3 Tg CH<sub>4</sub>). Uncertainties in Table 1 are 2σ of the emissions from the averaging period. The global inversion study by Chen and Prinn [17] estimates an annual emission of 2±1 Tg CH<sub>4</sub> from Alaska if 17% of North American wetlands are assumed to be in Alaska, as stated in their source map [48]. Our value can be used as a lower-bound for total emissions in 2012, and if we assume that 50% of annual CH<sub>4</sub> emissions occurs between October and April, as reported for a site in Greenland [35], then the upper-bound for emissions in 2012 would be 4 ± 1 Tg CH<sub>4</sub>. A

reasonable annual estimate for 2012 is the mean of these two bounds, 3 ± 1 Tg CH<sub>4</sub>, and is consistent with assuming that emissions for the months of October and November are similar to August and September and that emissions in the remaining months are near zero.

Our results are lower than emissions reported in a recent study of the Yukon River Valley [36], which gave an annual emission of 4.01 Tg CH<sub>4</sub> yr<sup>-1</sup> for this region alone, which comprises 30% of Alaska. Likewise, the annual emissions from Alaskan thermogenic seeps have been reported to be 1.5–2 Tg CH<sub>4</sub> yr<sup>-1</sup> [3]. This value would comprise at least 50–67% of the total annual Alaskan emissions. Both of these estimates seem to be higher than can be accommodated by our observations.

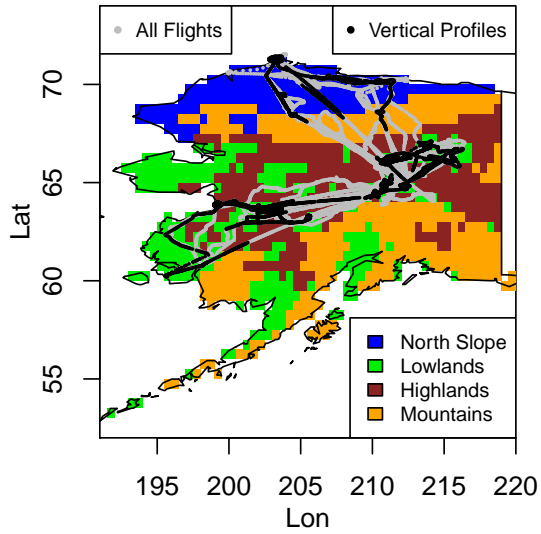
## Summary and Conclusions

CARVE is the first study to make frequent and sustained airborne measurements of CH<sub>4</sub> over large areas of Arctic and boreal Alaska throughout the growing season. We derived emissions of 2.1 ± 0.5 Tg CH<sub>4</sub> from Alaska during May to September 2012, and we found that the Lowland and Highland regions consistently emitted CH<sub>4</sub> at higher rates than the North Slope. A modest seasonal cycle was observed over all regions, with fluxes roughly doubling from May to July, then decreasing gradually in August and September. Stronger seasonality was likely not observed because the atmosphere integrates over heterogeneous land-types with asynchronous seasonal cycles. Analysis of CARVE 2013 measurements is under way, with 2014 measurements currently taking place. Comparing the results from these additional years with their different environmental forcing may allow the factors affecting emissions at a regional-scale to be determined.

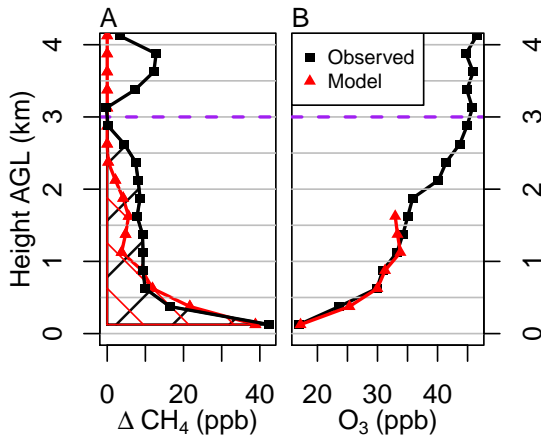
The total estimated CH<sub>4</sub> emitted from the region (2.1 ± 0.5 Tg CH<sub>4</sub> over May–September 2012) is quite small compared to the global emissions of 550 Tg CH<sub>4</sub> yr<sup>-1</sup> [21] (<0.5%), despite the recent warming of permafrost areas in Alaska. Since this is the first top-down regional study of Alaska based on observations, we cannot directly assess whether emissions have increased in response to climatic shifts. However, our results are consistent with fluxes obtained in recent global top-down inversion studies, which reported a lack of recent trends in CH<sub>4</sub> emission in the Arctic [15, 16, 18, 19]. Our work and these studies together indicate that CH<sub>4</sub> emissions from Arctic tundra regions have not contributed significantly to increasing levels of global CH<sub>4</sub> observed during the last decade. Our work during the growing season of 2012 in Alaska provide the baseline against which possible future increases in Arctic boreal and tundra CH<sub>4</sub> emissions can be assessed.

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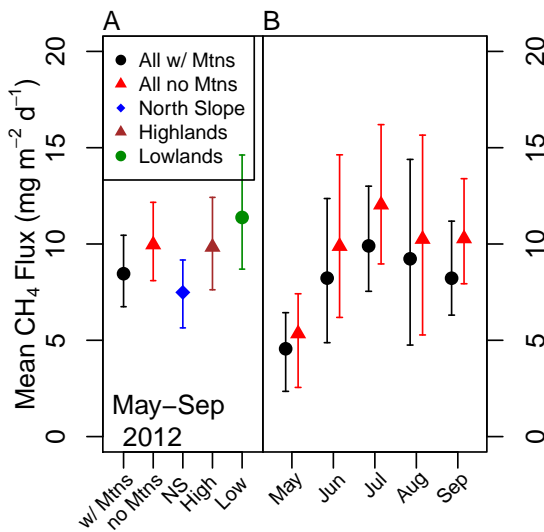
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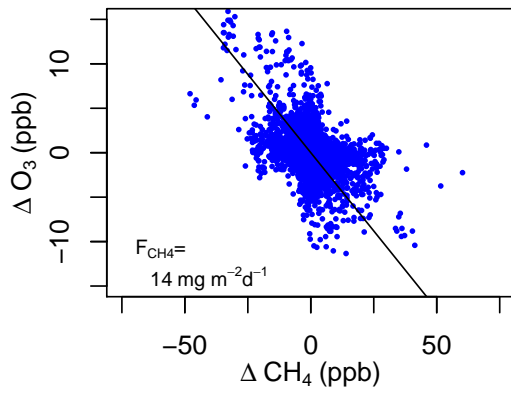
**Fig. 1.** Location of flight tracks (grey) and vertical profiles (black) during CARVE 2012. Background colours are elevation categories based on US EPA Level III ecoregions.



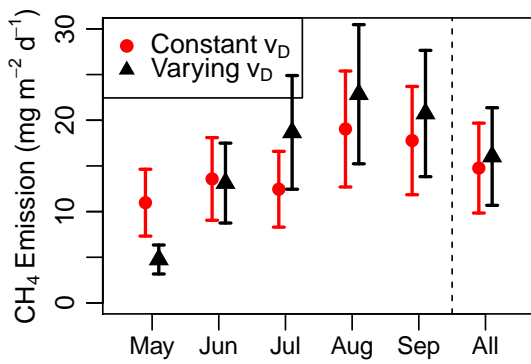
**Fig. 2.** Sample CH<sub>4</sub> vertical profile used for column analysis and corresponding O<sub>3</sub> profile from 22 September 2012. Dashed purple line is the identified top of the residual layer and hatched areas are used to determine the column enhancement.



**Fig. 3.** Estimated mean CH<sub>4</sub> fluxes from the column analysis for (A) the entire study period (May–September 2012) and (B) by month.



**Fig. 4.** Covariance of O<sub>3</sub> and CH<sub>4</sub> below 1500 m agl for June. See SI for other months.



**Fig. 5.** Estimated methane fluxes from the O<sub>3</sub>:CH<sub>4</sub> analysis assuming a constant and seasonally-varying O<sub>3</sub> v<sub>D</sub>, red and black points, respectively. Error bars reflect the uncertainty in the O<sub>3</sub> v<sub>D</sub> (~33%).

**Table 1.** Methane emissions from various models for the region 55–75°N, 141–169°W for May–Sep of the given years, except TEM which was run for all of Alaska and the given value is the annual emission

Lead author	Model	Emissions (Tg)	Averaging Period	Ref
Land surface models				
Melton	DLEM	0.8±0.2	1993–2004	[49]
Melton	LPJ-Bern	1.2±0.3	1993–2004	[49]
Melton	LPJ-WHyMe	6±1	1993–2004	[49]
Melton	LPJ-WSL	0.9±0.2	1993–2004	[49]
Melton	ORCHIDEE	1.0±0.4	1993–2004	[49]
Melton	SDGVM	0.7±0.2	1993–2004	[49]
Riley	CLM4Me	5±2	2001–2010	[50]
Zhu		2.6±0.1	2000–2009	[51]
Zhuang	TEM	3 (annual)	1980–1996	[32]
Matthews		4.34		[52]
Inverse models				
Bergamaschi	TM5-4DVAR	1.3±0.3	2001–2010	[15]
Bruhwyler	CT-methane	1.5±0.2	2000–2009	[16]
Chen	MATCH	2±1	1996–2001	[17]
<b>This study</b>		2.1±0.5	2012	



# Supplemental Information for “Methane emissions from Alaska in 2012 from CARVE airborne observations”

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September 29, 2014

5

## S1 Comparison of the two spectrometers

The water vapor correction for the G1301 model was calibrated in the laboratory before deployment. The water vapor levels throughout the study ranged from 0.013–1.8%, resulting in a correction for CH<sub>4</sub> and CO<sub>2</sub> of 0.013–1.9% and 0.016–  
10 2.3%, respectively, with average corrections of 16 ppb for CH<sub>4</sub> and 3.8 ppm for CO<sub>2</sub>. No water vapor correction was applied to measurements from the G2401 model because the sample is dried in that system and water vapor levels were less than 0.001%. For the entire 2012 study, the difference between the two instruments was on average  $0.7 \pm 2.7$  ppb for CH<sub>4</sub> and  $0.3 \pm 0.4$  ppm for CO<sub>2</sub>,  
15 where the uncertainty is the standard deviation. This gives us confidence in the water vapor correction, since the differences between the instruments are not correlated with water vapor and the water vapor correction is much greater. This result is consistent with previous studies in the literature detailing the water vapor correction for Picarro cavity ring down systems [1, 2]. The merged  
20 time series used in this study was based on the measurements from the G2401 and missing measurement points (e.g. due to calibration) were filled in by the G1301 offset by the mean difference between the two instruments for each flight.

## S2 Elevation categories based on ecoregions

The 20 Level III ecoregions defined by the United States Environmental Protection Agency [3] were grouped into four elevation categories: North Slope (Arctic Coastal Plain, Arctic Foothills); Highlands (Interior Forested Lowlands and Uplands, Interior Highlands and Klondike Plateau, Copper Plateau); Lowlands (Subarctic Coastal Plain, Seward Peninsula, Bristol Bay Nushagak Lowlands,

Aleutian Islands, Interior Bottomlands, Yukon Flats, Cook Inlet, Coastal West-  
ern Hemlock Sitka Spruce Forests); and Mountains (Brooks Range / Richardson  
Mountains, Ogilvie Mountains, Alaska Range, Wrangell and St. Elias Moun-  
tains, Ahklun and Kilbuck Mountains, Alaska Peninsula Mountains, Pacific  
Coastal Mountains).

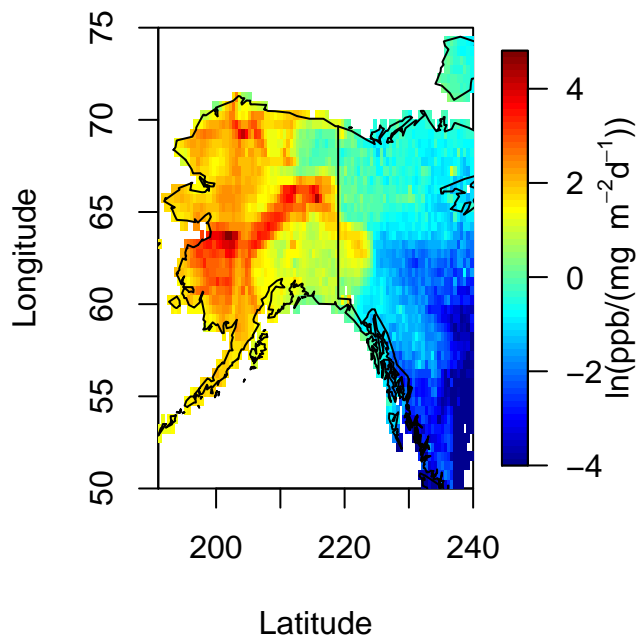


Figure S1: Surface influence of 30 vertical profiles used in this analysis.

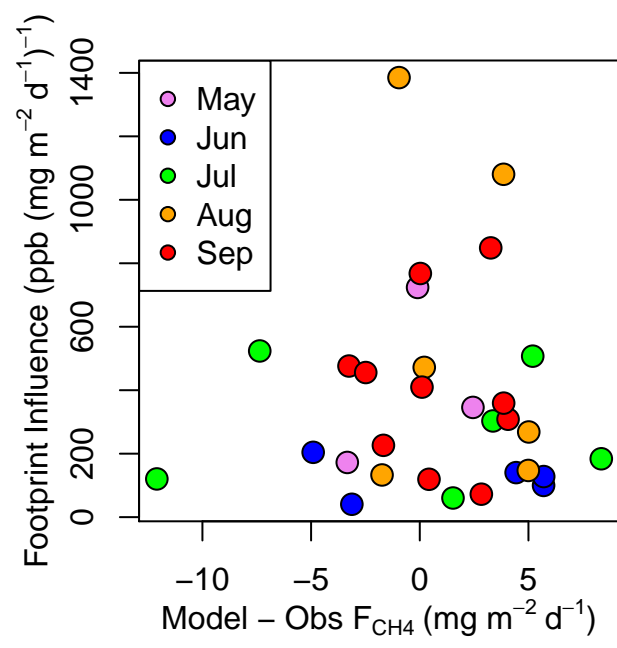


Figure S2: Difference between mean monthly flux and  $\overline{F_{\text{CH}_4, \text{VP}_i}}$  and corresponding footprint influence used in the weighted-average.

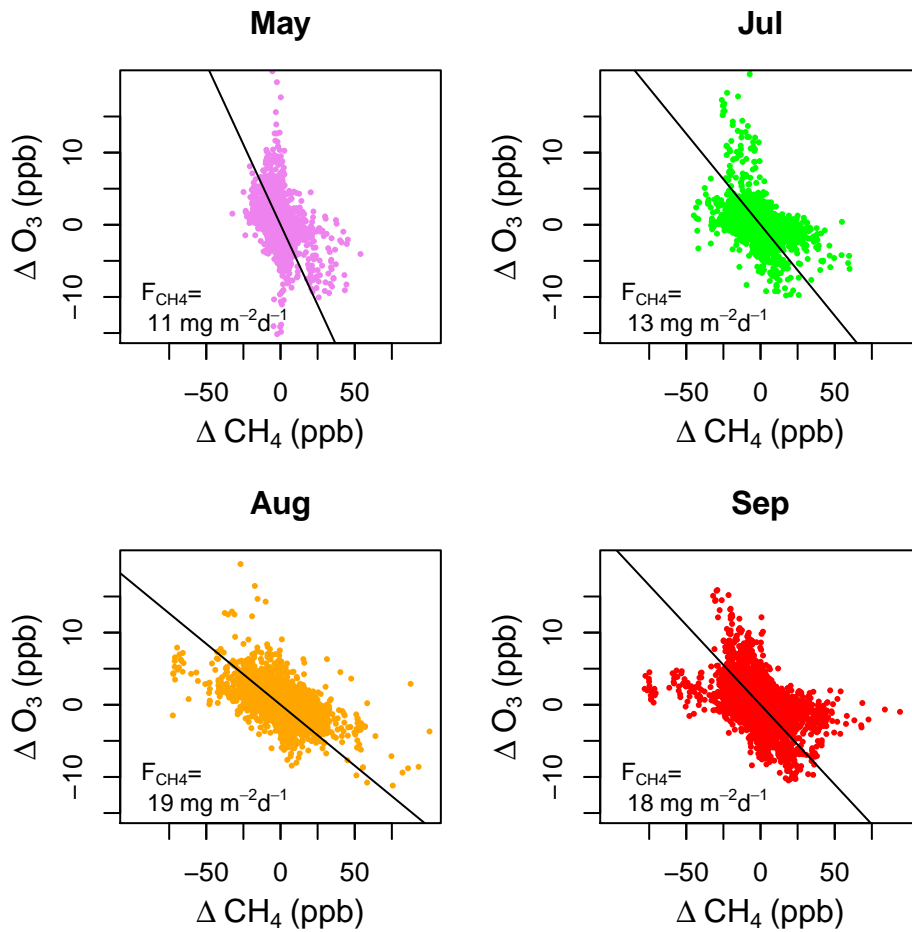


Figure S3: Covariance of  $\text{O}_3$  and  $\text{CH}_4$  below 1500 m agl for each month and corresponding  $\text{CH}_4$  flux.

Table S1: Details of vertical profiles used in analysis (dates in days since Jan 1)

Date	Start Date (UTC)	End Date (UTC)	Residual Layer Height (m)
20120523	143.838	143.878	4000
20120527	147.932	147.970	2300
20120601	152.866	152.950	3000
20120621	172.875	172.981	1900
20120622	173.765	173.824	1600
20120622	173.973	173.987	3000
20120624	175.882	175.895	5000
20120624	175.896	175.910	2500
20120717	198.775	198.871	3000
20120717	199.004	199.057	3200
20120722	203.881	203.905	1800
20120722	203.905	204.051	1800
20120725	206.857	206.895	1300
20120725	206.925	207.058	1200
20120814	226.762	226.784	2100
20120814	226.785	226.855	3200
20120819	231.984	232.028	3100
20120821	233.772	233.876	2400
20120822	234.771	234.960	1800
20120823	236.106	236.150	2900
20120919	262.855	262.868	4000
20120919	262.869	262.898	2100
20120921	264.913	265.027	3800
20120922	265.865	265.914	3000
20120922	265.919	265.991	3000
20120924	267.859	268.086	2000
20120924	268.086	268.170	2600
20120926	270.034	270.045	2500
20121001	275.013	275.025	2250
20121001	275.100	275.199	2300

Table S2: Methane emissions from tower measurements in tundra regions

Location	Lat	Lon	Land Type	Year	Flux (mg/m <sup>2</sup> /d)	Reference
Yukon Delta	61.09	-162	Tundra and Lake	1988	25	[4]
Happy Valley	69.17	-148.85	Wet Tundra	1995	80.2	[5]
Kuparuk Bay	69.51	-148.23	Wet Tundra	1996	3.3	[5]
Zackenberg	74.5	-21	Fen	1997	86.5	[6]
Barrow	71.32	-156.62	Wet Tundra	1999	68.7	[5]
Barrow	71.32	-156.62	Wet Tundra	2000	29.9	[5]
Barrow	71.32	-156.62	Wet Tundra	2001	34.3	[5]
Siberia	72.37	-126.5	Wet Tundra	2006	18.7	[7]
Barrow	71.28	-156.6	Wet Tundra	2007	24.6	[8]
Greenland	74.47	-20.57	Fen	2008	78.7	[9]
Greenland	74.47	-20.57	Fen	2009	52	[9]
Barrow	71.28	-156.6	Wet Tundra	2009	32	[10]
Barrow	71.28	-156.6	Wet Tundra	2011	37.3	[11]

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