

Using atmospheric trace gas vertical profiles to evaluate model fluxes: a case-study of Arctic-CAP observations and GEOS simulations for the ABoVE domain

5 Colm Sweeney¹, Abhishek Chatterjee^{2,3}, Sonja Wolter^{4,1}, Kathryn McKain^{4,1}, Robert Bogue^{5*}, Tim Steve Conley⁶, Newberger^{4,1}, Lei Hu^{4,1}, Lesley Ott³, Benjamin Poulter³, Luke Schiferl⁶, Brad Weir^{2,3}, Zhen Zhang⁷, Charles E. Miller⁵

¹NOAA Global Monitoring Laboratory, Boulder CO, USA

²Universities Space Research Association, Columbia MD, USA

³NASA Goddard Space Flight Center, Greenbelt MD, USA

10 ⁴CIRES, University of Colorado, Boulder CO, USA

⁵Jet Propulsion Laboratory, California Institute of Technology, Pasadena CA, USA

⁶Scientific Aviation, Boulder CO, USA

⁷LDEO, Columbia University, New York, NY, USA

⁷University of Maryland, College Park, MD, USA

15 * now at McGill University, Montreal, QC, Canada

Correspondence to: Colm Sweeney (colm.sweeney@noaa.gov)

Abstract. Accurate estimates of carbon-climate feedbacks require an independent means for evaluating surface flux models at regional scales. The altitude-integrated enhancement (AIE) derived from the Arctic Carbon Atmospheric Profiles (Arctic-CAP) project demonstrate the utility of this bulk quantity for surface flux model evaluation. This bulk quantity leverages background mole fraction values from the middle free troposphere, is agnostic to uncertainties in boundary layer height, and can be derived from model estimates of mole fractions and vertical gradients. To demonstrate the utility of the bulk quantity, six airborne profiling surveys of atmospheric carbon dioxide (CO₂), methane (CH₄) and carbon monoxide (CO) throughout Alaska and northwestern Canada between April and November 2017 were completed as part of NASA's Arctic-Boreal Vulnerability Experiment (ABoVE). The Arctic-CAP sampling strategy involved acquiring vertical profiles of CO₂, CH₄ and CO from the surface to 5 km altitude at 25 sites around the ABoVE domain on a 4- to 6-week time interval. All Arctic-CAP measurements were compared to a global simulation using the Goddard Earth Observing System (GEOS) modeling system. Comparisons of the AIE bulk quantity from aircraft observations and GEOS simulations of atmospheric CO₂, CH₄ and CO highlight the fidelity of the modeled surface fluxes. The model-data comparison over the ABoVE domain reveals that while current state-of-the-art models and flux estimates are able to capture broadscale spatial and temporal patterns in near-surface CO₂ and CH₄ concentrations, more work is needed to resolve fine-scale flux features that are captured in CO observations.

35 1. Introduction

There are many uncertainties to predicting the impact of increased emissions of CO₂ and CH₄ in the atmosphere. Carbon-climate feedbacks (Arora et al., 2020) are among the most uncertain climate

40 feedbacks. Without a better understanding of how changes in temperature, CO₂ itself, water and nutrients are magnifying or reducing the impact of increased emissions of greenhouse gases (GHG), it will be difficult to use climate models to accurately predict climate change. This uncertainty not only stems from a poor mechanistic understanding of how the biosphere will respond at the smallest scales but also how changes in the landscape drive changes in local environments.

45 The Arctic, in particular, is a region where carbon-climate feedbacks are critical to understand given the vast quantities of carbon sequestered in the permafrost soils of the northern high latitudes (Hugelius et al., 2014). Rapid changes in temperature have led to concerns about the potential for significant carbon emissions due to changes in ecosystems, permafrost and large-scale disturbances like fires (Schuur et al., 2015; McGuire et al., 2018; Turetsky et al., 2020). Our understanding of the magnitude and behavior of the carbon system response to these changes is rudimentary (Koven et al., 2015). For instance, release of carbon from the permafrost pool could result in increased emissions of CH₄ from anaerobic degradation; increased emissions of CO₂ from aerobic degradation; increased uptake of carbon due to new availability of nutrients and above-ground ecosystem growth; or an increase in mobilization of carbon through runoff. Alternatively, increases in disturbances such as fires may significantly impact below-ground carbon storage, uptake of CO₂ and emissions of CH₄, CO, and CO₂.

50 Limitations in our understanding of the accuracy of modeled fluxes of CO₂, CO and CH₄ have resulted in large uncertainties in the magnitude of Arctic carbon-climate feedbacks (e.g., Koven et al., 2011; Schneider von Deimling et al., 2012; Schaefer et al., 2014; Lawrence et al., 2015; Schuur et al., 2015). The lack of observations from which to build and evaluate models of the biosphere is a significant source of the problem and leads to both enhanced uncertainty and reduced fidelity in our model simulations. In general, land- and ocean-atmosphere fluxes from climate models are most commonly

60 evaluated using flux measurements made with eddy covariance or flux chamber techniques (Sasai et al., 2007). While flux measurements of these types are widely available over many ecosystem types, they represent the impact of limited spatial domains that are rarely more than a 1000 m radius around a given site (Schmid, 2002; Gockede et al., 2005) and may be significantly smaller depending on topography, wind direction, boundary layer stability, and measurement approach. Land surface inhomogeneities

65 within these small footprints (Baldocchi et al., 2005) and regional-scale (100-1000 km scales) variability of these ecosystems can lead to significant biases when eddy covariance measurements are scaled up to represent large areas (e.g. Mekonnen et al., 2016). This is especially true in the Arctic where microtopography can result in fluxes varying by orders of magnitude on a scale of 1-100 meters (Johnston et al., 2014).

70 An alternative to the “bottom-up” evaluation approach, which relies on the eddy covariance measurements, is the “top-down” approach, which makes use of atmospheric measurements of species like CO₂, CH₄ and CO and modeled atmospheric transport patterns to infer the surface fluxes needed to reproduce observed atmospheric concentrations (Pickett-Heaps et al., 2011; Miller et al., 2016; Thompson et al., 2017 are examples in the Arctic) over large regional scales. In a data-limited region,

75 this inverse approach generally takes a forward-flux model, or a set of observations that are likely correlated with the flux, as a prior or first guess. The inverse approach then estimates the flux by scaling the prior. While the inverse approach results in a flux estimate that meets the constraint of the trace gas measurements and modeled transport, the variability in surface flux from these analyses cannot be directly attributed to mechanisms such as temperature changes, CO₂ fertilization, nutrient enrichment

80 and water stress and, therefore do not have any predictive capabilities. Also, inverse methods are influenced by errors in atmospheric transport and assumptions about error covariances, which are difficult to characterize (Gourdji et al., 2012; Lauvaux et al., 2012; Mueller et al., 2018; Chatterjee and Michalak, 2013).

In this study, a hybrid approach is taken to evaluate and benchmark the accuracy of current state-of-the-art, bottom-up, land-surface flux models using a bulk quantity calculated from atmospheric vertical profiles of trace gas mole fractions. The goal is to present an approach to evaluate land-surface flux models that capture complex carbon cycle dynamics over the northern high-latitudes. NASA's Goddard Earth Observing System (GEOS) general circulation model (GCM) is used with a combination of surface flux components for CO₂, CH₄ and CO to create 4D atmospheric fields; these fields are subsequently evaluated using the altitude-integrated enhancements (AIE) calculated from profiles collected during the Arctic Carbon Atmospheric Profiles (Arctic-CAP) airborne campaign. Both the Arctic-CAP project and the GEOS model runs for the domain are part of NASA's Arctic Boreal Vulnerability Experiment (ABoVE, www.above.nasa.gov), a decade-long research program focused on evaluating the vulnerability and resiliency of the Arctic tundra and boreal ecosystems in western North America (Miller et al., 2019). One of the primary objectives of the ABoVE program is to better understand the major processes driving observed trends in Arctic carbon cycle dynamics, in order to understand how the ecosystem is responding to environmental changes and to characterize the impact of climate feedbacks on greenhouse gas emissions. ABoVE has taken two approaches to better understand critical ecosystem processes vulnerable to change. The first is through ground-based surveys and monitoring sites in representative regions of the ABoVE domain. These multi-year studies provide a backbone for intensive investigations, such as airborne deployments. The Arctic-CAP campaign discussed here was one such airborne deployment that was conducted during the spring-summer-fall of 2017 (Section 2.1). The subsequent analysis described here illustrates how improvements in surface models develop through ground-based surveys, and monitoring sites can be evaluated and tested over larger spatial scales using aircraft profiles (Section 3). This study uses the bulk quantify from Arctic-CAP aircraft profiles to directly evaluate the terrestrial surface flux models of CO₂, CH₄ and CO. For the sake of demonstration, we rely on one transport model and one flux scenario for each tracer (i.e., CO₂, CH₄ and CO) to show the utility of the three carbon species to diagnose and identify deficiencies in the land flux models. Ongoing and future studies will build upon the results discussed here and further diagnose transport and flux patterns from multiple models based on additional aircraft and ground-based observations throughout the ABoVE domain. This approach demonstrates the value of aircraft profiles.

2. Methods

2.1. Arctic-CAP Flight Planning and Sampling Strategy

115 Arctic-CAP was designed to measure vertical profiles of atmospheric CO₂, CH₄ and CO mole fraction to capture the spatial and temporal variability of carbon cycle dynamics (Sweeney et al., 2015; Parazoo et al., 2016) across the ABoVE domain. Six campaigns were performed during 2017: late April – early May, June, July, August, September, and late-October – early November. Arctic-CAP flights surveyed

the ABoVE Study Area and were organized around an Alaskan circuit and a Canadian circuit (Fig. 1).
120 The Alaskan circuit covered a region where aircraft measurements were previously made during 2012-
2015 by the Carbon in Arctic Reservoirs Vulnerability Experiment (CARVE; Miller et al., 2012), which
included the Alaskan Boreal Interior, Brooks Range Tundra and the Alaskan Tundra ecoregions. The
Arctic-CAP Alaska circuit was primarily west of Fairbanks, Alaska, and included Galena, Bethel,
Unalakleet, Nome and Kotzebue. The northern section of the circuit overflowed Utqiagvik (formerly
125 Barrow), Atqasuk, Deadhorse and the Toolik Lake Research Station – all North Slope tundra sites with
long-term measurements of atmospheric CO₂ and CH₄. The Arctic-CAP Canadian circuit focused on
flying over sites in and around the Inuvik and Yellowknife areas in the Canadian Arctic. In the Inuvik
region, the aircraft overflowed the Trail Valley Creek and Havipak Creek research sites, and the Daring
Lake and Scotty Creek flux tower sites were overflowed on the way to and from the Yellowknife area.
130 The Canadian Circuit expands upon the ecoregions covered in the CARVE missions to include the
Boreal Cordillera, Taiga Plain, Taiga Shield and the Southern Arctic Tundra ecoregions.
Approximately 25 vertical profiles were acquired during each campaign (Fig 2). The majority of each
flight day was spent in the well-mixed boundary layer with 2-4 vertical profiles up to altitudes of 5000
m above sea level (masl). Using missed approaches to get as near to the ground as possible, profiles
135 diagnosed temporal changes in the boundary layer and residual layers above where surface fluxes may
have recently (< 3 days) influenced that atmospheric column. During the 2017 season, Arctic-CAP
flights were complemented by additional vertical profiles collected in the ABoVE domain by the
ASCENDS (Active Sensing of CO₂ Emissions over Nights, Days, & Seasons, [https://www-
air.larc.nasa.gov/cgi-bin/ArcView/ascends.2017?MERGE=1](https://www-air.larc.nasa.gov/cgi-bin/ArcView/ascends.2017?MERGE=1)) and ATom (Atmospheric Tomography,
140 Wofsy et al., 2018) campaigns and the NOAA Carbon Cycle Aircraft Program (Karion et al., 2013;
Sweeney et al., 2015). The focus of this study will be on the CO₂, CH₄ and CO data acquired during
Arctic-CAP and, in particular, utilizing the profiles acquired during each flight to separate signals from
near-field surface fluxes from large-scale deviations in a way that is agnostic to model errors due to
inaccurate vertical transport.

145

2.2. Aircraft and Payload

Arctic-CAP flights were performed with a Mooney Ovation 3 aircraft (tail number N617DH, Scientific
Aviation). The Mooney operated at a cruise speed of 170 kts and reached profile altitudes of 5 km
(17,000 feet) on each flight, with most legs lasting 4-5 hours and covering an average distance of ~1350
150 km. The average ascent and descent rates were limited to ~100 m/min to minimize hysteresis in the
temperature and relative humidity measurements. The basic research payload flown on all six research
missions included continuous in-situ CO₂, CH₄, CO, H₂O, temperature and horizontal winds. The in-situ
measurements (Sweeney and McKain, 2019) followed the methodology described in Karion et al.
(2013), and wind measurements followed the protocol outlined in Conley et al. (2014). During Arctic-
155 CAP, *in situ* measurements of CO₂, CH₄ and CO were made every ~2.4 s and aggregated to 10 s
averages for comparison to the GEOS 4D fields (latitude, longitude, altitude and time). Sampling at the
10 s resolution reduces the spatial representativeness error between the model grid cell and the aircraft
observations.

160 Programmable flask packages (PFPs; Sweeney et al., 2015) provided an independent check of the
calibration scale of the continuous *in situ* CO₂, CH₄ and CO measurements, as well as samples for more
than 50 different species including N₂O, SF₆, and a variety of hydrocarbons, halocarbons and isotopes
of carbon (Sweeney et al., 2020). Carbonyl sulfide measured in the flask samples can be used as a tracer
of gross primary productivity (GPP) (Montzka et al., 2007), while ethane, propane and the C-13 isotope
165 of CH₄ provide another constraint on the source of the CH₄ emissions. Each flight sampled a single 12-
flask package providing a total of ~84 flasks per research mission to better understand the factors
controlling local fluxes of CO₂, CH₄ and CO and the long-range transport of these species from low
latitudes.

2.3. GEOS Earth System Model & Atmospheric CO₂, CO and CH₄ Modelling

170 The GEOS (Rienecker et al., 2011; Molod et al., 2015) model is a complex yet flexible modeling
system that describes the behavior of the land and atmosphere on a variety of spatial (~12.5-100 km)
and temporal (hourly to decadal) scales. GEOS includes both an atmospheric General Circulation
Model (GCM) and data assimilation system that have been used to produce the widely-used Modern-
Era Retrospective Analysis for Research and Applications (MERRA) (Rienecker et al., 2011) and
MERRA-2 (Bosilovich et al., 2015; Gelaro et al., 2017). The GEOS Forward Processing (GEOS FP)
175 system produces atmospheric analyses and 10-day forecasts in near real-time, which are used to provide
forecasting support to NASA field campaigns and satellite instrument teams (e.g. Strode et al., 2018).
GEOS has also been used extensively to study atmospheric carbon species (e.g. Allen et al., 2012; Ott et
al., 2015; Weir et al., 2021).

180 The GEOS setup utilized in this work simulates CO₂, CO and CH₄ simultaneously at nominal 0.5°
horizontal resolution, 72 vertical layers (up to ~0.1 hPa) with trace gas output saved every 3-hours. For
CO₂, the surface fluxes consist of five different components from a Low-order Flux Inversion (LoFI)
package (Weir et al., 2021): 1) net ecosystem exchange (NEE) from the Carnegie Ames Stanford
Approach - Global Fire Emissions Database (CASA-GFED) mode with a parametric adjustment applied
to match the atmospheric growth rate (Weir et al., 2021), 2) anthropogenic biofuel burning emissions,
185 i.e., harvested wood product (Van Der Werf et al., 2003), 3) biomass burning emissions derived from
the fire radiative power based Quick Fire Emissions Dataset (QFED; Darmenov and Da Silva, 2015) 4)
fossil fuel emissions from the Open-source Data Inventory for Anthropogenic CO₂ (ODIAC; Oda and
Maksyutov, 2011), and 5) ocean exchange fluxes based on *in situ* measurements of the partial pressure
of CO₂ in sea-water from the Takahashi et al. (2009) dataset but adding back the inter-annual variability
190 and applying a mean ocean partial pressure of CO₂ growth rate of 1.5 μatm/yr at each point every year.
For CO, the emissions include biomass burning emissions from QFED, and climatologies of fossil fuel
and biofuel emissions and VOC fields (Duncan et al., 2007; Ott et al., 2010). Finally, the CH₄ flux
collection consists of five components: 1) wetland emissions from the process-based ecosystem model
LPJ-*wsf* (Lund-Potsdam-Jena model, WSL version - Poulter et al., 2011), 2) biomass burning emissions
195 from the QFED, 3) industrial and fossil fuel emissions from the Emissions Database for Global
Atmospheric Research (EDGAR v4.3.2, Janssens-Maenhout et al., 2017; Crippa et al., 2018), 4)
agricultural emissions from EDGAR v4.3.2 and 5) anthropogenic biofuel burning emissions from
EDGAR v4.3.2. Note that since the EDGAR v4.3.2 emissions record ends in 2012, the same set of
values from 2012 were used for the year 2017. As shown later, this is not a bad assumption considering

200 that for the majority of the ABoVE domain, the most critical CH₄ emissions are from the wetlands
sector. On the other hand, care was taken to use a version of the LPJ-*ws/* model that includes a state-of-
the-art hydrology subroutine (TOPMODEL) to determine wetland area and its inter- and intra-annual
dynamics (Zhang et al., 2016), a permafrost and dynamic snow model (Wania et al., 2009) with explicit
205 the CH₄ emissions. Table 1 provides a summary of the flux components, their specifications and
associated references.

2.4. AIE calculation

210 As will be explained in the following results section the surface fluxes of CO₂, CH₄ and CO in GEOS
are compared to aircraft observations by first subtracting the average daily free tropospheric value
(>3000 m for CO₂ and CH₄ and >4000 m for CO, X_{FT}) from each measurement below 3000 m and
comparing that to the altitude integrated sum

$$\Delta X = \int_{z=ground}^{z=3000} ((X - X_{FT})/n_{BL}) n dz \quad \text{Eq. 1}$$

215 where ΔX is altitude-integrated sum of the mole fraction of species X minus X_{FT} divided by the n_{BL}
where $n_{BL} = \int_{z=ground}^{z=3000} n dz$ and n is the atmospheric number density. It is assumed that the mole
fraction of each trace gas species measured at the lowest point in each profile is constant to the ground
level. Ground level altitude is taken from USGS (USGS, 2017). Thus, the AIE is equivalent to average
enhancement in the boundary layer after accounting for altitude changes in number density. As will be
220 explained in the results section, the 3000 m was picked as cutoff for CO₂ and CH₄ because of the low
variability of these tracers above that altitude level where as the cutoff point for CO was chosen to be
4000 m.

Table 1. Components of fluxes for simulation of atmospheric concentrations of CO₂, CO and CH₄ in GEOS. Flux components that are the primary drivers of observed signals within our study domain are distinguished with italics.

225

Flux type	Used in simulation of	Inventory / Process-based model name	Reference
Fossil fuel	CO ₂	ODIAC	Oda and Maksyutov, 2011
Biofuel	CO ₂	CASA-GFED3	Van Der Werf et al., 2003
<i>NEE</i>	CO ₂	LoFI CASA	Weir et al., 2021
Ocean	CO ₂	LoFI Takahashi	Weir et al., 2021
Biomass Burning / Fires	CO ₂ , CO,	QFED	Darmenov and Da Silva, 2015
Fossil fuels & biofuels	CO	Climatology	Duncan et al., 2007
VOC	CO	GMI climatology	Duncan et al., 2007
<i>Wetlands</i>	CH ₄	LPJ-wsl	Poulter et al., 2011, Zhang et al.,
Agriculture and waste	CH ₄	EDGAR v4.3.2	Crippa et al., 2018
Biofuels	CH ₄	EDGAR v4.3.2	Crippa et al., 2018
Industrial and fossil fuel	CH ₄	EDGAR v4.3.2	Crippa et al., 2018

Table 2. GEOS CO₂ Flux Estimates (PgC yr⁻¹) for 2017. Flux emissions are specified for (a) the natural land sink component, which includes the sum of NEE and biomass burning, and (b) all anthropogenic source components, which include fossil fuel and biofuel burning.

ABoVE Domain		pan-Arctic (>48 N)		Global	
Land Sink	Fuel Sources	Land Sink	Fuel Sources	Land Sink	Fuel Sources
-0.32	0.11	-1.84	1.37	-3.28	11.08

230 We have assessed global and pan-Arctic budgets and compared against existing studies (Tables 2 and 3) and estimates to establish the fidelity of the model fluxes for large-scale assessments. CO₂ flux estimates indicate that the ABoVE domain is a 0.32 PgC sink for our study year, 2017. This represents ~17% of the calculated pan-Arctic terrestrial carbon sink, which is consistent with the fraction of the land area > 48N represented by the ABoVE domain (~16%). Perhaps more significantly, the 1.84 PgC

235 pan-Arctic sink represents 56% of the global sink for 2017. We attribute this large uptake to the vast boreal forests > 48 N, particularly in Siberia (Sasakawa et al., 2013), where the contemporary Arctic tundra is thought to be nearly carbon neutral with uncertainties allowing for a small to moderate sink or a small source (McGuire et al., 2016). These findings are also consistent with Wunch et al. (2013) who used GOSAT satellite data and TCCON ground-based column measurements to determine that

240 interannual variability in Northern Hemisphere CO₂ uptake was dominated by changes in the boreal forest. More recent studies, such as Welp et al. (2016) and Commane et al. (2017) have also used atmospheric inversions to highlight that >90% of the carbon sink in the northern high latitudes reside in the boreal forests. Our simple forward model simulations and the Arctic-CAP data provide a unique opportunity to assess the validity of these previous findings over the ABoVE domain. Sub-regional flux

245 estimates within the ABoVE domain are part of ongoing investigations and will be captured in future studies.

Table 3. GEOS CH₄ Flux Estimates (TgCH₄ yr⁻¹) for 2017. CH₄ flux emissions are specified for (a) the wetland component, and (b) all source components, which include wetlands, industrial and fossil fuel, agriculture and waste, biomass burning, biofuel burning and other natural emissions.

ABoVE Domain		pan-Arctic (>48 N)		Global	
Wetland	All Sources	Wetland	All Sources	Wetland	All Sources
9.01	11.64	21.74	52.03	187.39	536.01

250 Examination of the specified CH₄ flux estimates for the ABoVE domain (Table 3) reveal a remarkable
 result: 78% of the emissions, 9.01 TgCH₄ yr⁻¹, come from wetlands. Furthermore, ABoVE wetlands
 emissions account for 41% of pan-Arctic CH₄ wetland emissions. Both results suggest a
 disproportionately large contribution of North American wetlands to the regional CH₄ budget. Placing
 this in a larger context, the 52 TgCH₄ yr⁻¹ from all pan-Arctic emissions account for only about 10% of
 255 the global emissions. Our pan-Arctic CH₄ emissions estimate of 52 TgCH₄ yr⁻¹ is only 60% of the 82-84
 TgCH₄ yr⁻¹ determined by Thompson et al. (2017) for latitudes > 50N and the period 2005-2013. The
 reasons for this large discrepancy are unclear, particularly since the Thompson et al. (2017) study
 derived their estimate from an inversion of atmospheric CH₄ observations; previously, such top-down
 estimates have tended to be lower than most forward model emissions estimates. Subtracting the 11
 260 TgCH₄ yr⁻¹ we estimate for the ABoVE domain from our pan-Arctic value leaves 41 TgCH₄ yr⁻¹ for the
 remainder of the pan-Arctic. Future work with additional observations and model simulations will help
 us understand how specific sectors in the ABoVE domain can better capture the complexity of pan-
 Arctic CH₄ emissions. Our overall model value of 536 TgCH₄ yr⁻¹ for global CH₄ emissions in 2017
 falls just outside the range of annual emissions estimates for the decade 2008-2017 (Saunois et al.,
 265 2019). This discrepancy is primarily due to the fact that we are looking at different time periods and,
 unlike Saunois et al. (2019), we do not extrapolate the EDGARv4.3.2 dataset using the extended FAO-
 CH₄ emissions and/or British Petroleum statistical review of fossil fuel production and consumption
 (see Equation 1 in Saunois et al., 2019); instead, we adopt a much simpler approach of repeating the
 EDGARv4.3.2 from 2012 for the year 2017. Contrary to the emissions from the coal, oil and gas sector,
 270 our wetland methane flux emissions are obtained from the LPJ-*wsl* model (Table 1). LPJ-*wsl* is one of
 the prognostic models that provide wetland emission estimates to the global methane budget (Table 2 in
 Saunois et al., 2019). It is not surprising then that our global wetland CH₄ emission estimates for 2017
 are in line with both the bottom-up (100-183 TgCH₄ yr⁻¹) and top-down (155-217 Tg CH₄ yr⁻¹)
 estimates used in the global methane budget estimate.

275 3. Results and Discussion

3.1. Analysis of Profiles

Table 4. Arctic-CAP 2017 campaign summary

Campaign	Start (DOY)	End (DOY)
Apr/May	116	124
June	157	170

July	190	202
August	229	242
September	251	271
Oct/Nov	291	310

280 Vertical profiles of CO₂, CH₄ and CO were acquired during 56 flights over the six Arctic-CAP
 campaigns from late April (day of year (DOY) 116) through early November (DOY 310) 2017 (Table
 4). Figure 4 presents the composite vertical profile data for each campaign. The monthly composite
 CO₂, CH₄ and CO vertical profiles capture the expected variations in the seasonal cycle. The composite
 profiles also show more variability in the boundary layer (altitudes < 3000 masl) within each month and
 across months than in the free troposphere for CO₂ and CH₄ (altitudes > 3000 masl). Unlike CO₂ and
 CH₄, CO variability in the free troposphere is significantly greater in July and October than the
 285 boundary layer showing either long-range transport of CO or CO injected high (>3000 masl) into the
 troposphere by local wildfires.

A clearer picture of the vertical gradients between the free troposphere and the boundary layer can be
 seen by subtracting free tropospheric means from measurements below 3000 masl. The CO₂ gradients
 between the measurements below 3000 masl and average daily free troposphere values show a
 290 drawdown in the boundary layer for most of the profiles starting in June and lasting until the end of the
 September campaign (Fig. 5). The drawdown signal in CO₂ over the Northern Alaska Tundra (often
 referred to as the “North Slope”) was most pronounced in mid-July and continued through the
 September campaign. The CO₂ drawdown in the more southerly regions of the Boreal Cordillera and
 Alaskan Boreal Interior peaked in August. By the October campaign many regions were showing
 295 significant enhancements in the boundary layer CO₂ mole fraction relative to the free troposphere. On
 the other hand, for both CH₄ and CO, significant enhancements were observed from June through early
 November. Methane enhancements over the Northern Alaska Tundra were observed from July onward,
 consistent with patterns observed at the long-term surface monitoring station in Utqiagvik (Sweeney et
 al., 2016). Similarly, boundary layer CO₂ and CH₄ are both most enhanced in September and October
 300 on the North Alaska Tundra. Due to the high variability in CO above 3000 masl during July and
 October (Fig. 4), it is more difficult to use this approach to derive CO enhancements from surface
 fluxes. To avoid the impact of fire-based CO that has been injected into the free troposphere, the mean
 background value is taken from measurements above 4000 masl. This analysis shows that Canadian
 Taiga and Alaskan Boreal Interior are the predominant sources of boundary layer CO emissions
 305 reflecting fires in these regions at that time. It should be noted that large enhancement values for CO₂,
 CH₄ and CO were observed with the Alaskan Boreal Interior, which were the result of samples taken in
 the early morning (10:00 local time) before the boundary layer had fully developed (typically around
 11:00-12:00 local time). This trapping of night-time emissions results in significant surface
 enhancements that quickly taper off with altitude. These measurements were typically taken during the
 310 first profile out of Fairbanks where the majority of the Arctic-CAP flights originated.

3.2. Model Data Comparisons

Aircraft profiles that measure the gradient from the boundary layer into the free troposphere are
 particularly useful for evaluating atmospheric models and for separating errors and uncertainties related
 to atmospheric vertical transport and surface flux model simulations. This is demonstrated by

315 comparing surface flux models for CO₂, CH₄, and CO using a single GCM to evaluate the land surface
flux model.

3.2.1. Point by Point Comparison

In the GEOS model run used for these comparisons, an effort was made to match the global
atmospheric burdens of CO₂, CH₄ and CO; however, given the uncertainties in the sources and sinks of
320 these trace gases and in the representation of long-range and local atmospheric transport, it is not
uncommon to have mean offsets between the observed and the modeled mole fractions. To evaluate
surface fluxes in the ABoVE domain, it is important to consider both the impact of regional-scale fluxes
and long-range transport processes that control the mole fractions of CO₂, CH₄ and CO throughout the
ABoVE domain. A time series comparison of the modeled and the observed CO₂, CH₄ and CO mole
325 fractions (Fig. 6) suggests that gross features of the seasonal cycles are matched, although some
significant differences require detailed analysis by considering different elements of each vertical
profile.

3.2.2. Free Troposphere Comparisons

As demonstrated from the analysis of the boundary layer enhancements (Fig. 6) observed during Arctic-
330 CAP, it is useful to subtract the average free tropospheric mole fraction from each profile to better
understand the local influences within a particular profile. Differences in the mean free tropospheric
values, however, can be a valuable indicator of how large-scale biases in the model influence point-to-
point comparisons.

In the case of CO₂, the mean daily CO₂ mole fraction in the observed free troposphere is increasing
335 faster than modeled values over the course of six research missions. The largest offset exceeds a mean
value of ~2 ppm (observed – modeled) during the September campaign (Fig. 7). Based on the available
model runs, it is difficult to diagnose what causes this offset, although a few hypotheses can be put
forward. Given the decreasing latitudinal gradient for CO₂ in the free troposphere at this time of year,
the offset could be explained by sluggish meridional transport in the model. Alternatively, exaggerated
340 biological uptake in the model in regions outside the study area could be pulling down the CO₂ in
modeled free troposphere more rapidly than the drawdown observed over the ABoVE domain.
Likewise, measured CH₄ increases faster than modeled CH₄ over the course of the campaign. Given the
decreasing meridional gradient for CH₄ that exists during the summer months, sluggish transport could
explain the difference between model and observations. Alternatively, modeled June-July-August
345 emissions of CH₄ in areas contained by the ABoVE domain could be underestimated, leading to slower
increase in modeled free tropospheric CH₄.

Finally, the difference between modeled and observed mole fractions of CO in the free troposphere is
mainly driven by inaccuracies in the modeled CO from fire plumes both within and outside the ABoVE
domain. Figures 4, 6 and 7 show observations of large CO enhancements above 4000 masl during the
350 July, August and October/November campaigns. Local fires were likely responsible for the large
excursions in the free tropospheric CO between different profiles. Accurately simulating the injection
height of fire plumes is challenging (Freitas et al., 2007; Strode et al., 2018). The GEOS model

distributes biomass burning emissions throughout the planetary boundary layer (PBL) to represent injection above the surface layer, but this method can result in underestimated local emissions for fire plumes detraining in the free troposphere. In regions remote to the ABoVE domain, emissions can be mixed and lofted by large-scale weather systems, which may explain why the model performs better in simulating long-range CO plume transport than it does in capturing the CO enhancements from local fires. The observation-model mismatch is likely compounded by the inability of the model to accurately simulate the subgrid-scale vertical mixing necessary for capturing vertical profiles for local sources.

360 3.2.3. Boundary Layer Comparisons

Accurately modeling boundary layer mole fractions of CO₂, CH₄ and CO depends on the correct representation of two key factors. First, there is a need to accurately model the local surface-atmosphere flux, and second, there is a need to correctly model the physical evolution of the PBL, as well as horizontal transport and vertical mixing out of the PBL into the free troposphere. GCMs have limited horizontal and vertical resolution and require parameterizations to predict both the rate of change and the absolute value of the PBL height over the course of the day. Errors in PBL mixing directly impact the tracer mole fraction estimate. Overestimation of the PBL height causes an artificial dilution of the impact of surface flux. Conversely, underestimation of the PBL height results in amplification of the impact of a surface flux on the simulated PBL mole fraction. Additionally, GCMs typically simulate large-scale horizontal gradients more accurately than PBL height unless there are large topographic changes that occur on horizontal scales less than the model resolution (for GEOS, 0.5 degree). This is because such large-scale patterns are generally well-constrained by the millions of *in situ* and satellite observations incorporated into meteorological analyses while PBL mixing is represented by highly simplified parameterizations

375 The three carbon species that we investigate in this study provide different diagnostic information about the model transport and flux specifications. In the case of a gas like CO that often comes from a specific point source in the Arctic, accurate placement of the emissions, both in the horizontal and the vertical, and the modeled wind direction are critical factors. The ABoVE domain is made up of large expanses of forest and tundra in which CO₂ fluxes are more uniformly distributed, making the transport accuracy of individual plumes a less critical factor for simulating CO₂. Accurately estimating CH₄ mole fractions may be more sensitive to horizontal transport in the PBL if CH₄ emissions are dominated by specific features such as lakes or wetlands, or anthropogenic point sources from oil and gas production such as those observed on the North Slope (Floerchinger et al., 2019). However, we observed consistent PBL CH₄ enhancements throughout each campaign (Fig. 5), suggesting a spatial homogeneity in CH₄ emissions rather than emissions from specific point sources.

385 3.3. Altitude-integrated Enhancements (AIEs)

While individual mole fraction measurements are challenging to reproduce given errors in both modeled surface fluxes and transport, the vertical profile provides a unique opportunity for removing significant uncertainties in transport in order to better assess the surface flux model of a specific long-lived tracer. Assuming that horizontal transport is a relatively small source of bias and the upper part of the free troposphere (>3000 masl) is largely unaffected by local processes, it is possible to use the information

in the vertical profile to reduce the effects of vertical transport. This can be estimated by vertically integrating the net change in the PBL due to a surface flux from the surface to a specific altitude that is well above the boundary layer. For this study, almost all the enhancements for CO₂ and CH₄ were
395 observed below 3000 masl.

By subtracting the average free tropospheric (FT) values in both the model and the measurements and averaging the resulting enhancements or depletions for each profile mapped on equal altitude bins from surface to 3000 masl (Eq. 1), we quantify a total enhancement (AIE) resulting from the surface flux (Fig. 8). The resulting measured and modeled AIE show good correlations for CO₂ and CH₄ but the CO
400 correlations are not as promising.

The average measured enhancement in CO₂ and CH₄ below 3000 masl is correlated with the forward model such that more than 50% and 36%, respectively, of the observed variability is captured by the model (Fig. 8). The average CO enhancements in the lower 3000 masl is captured by the model with lesser accuracy – in fact, the model only captures 26% of the observed variability along with a
405 significant bias throughout the growing season.

3.3.1. CO₂ AIE

To understand the true value of the aircraft profile in evaluating the ability of the surface flux model to reproduce observed fluxes over large regional expanses, it is useful to rigorously compare the differences between modeled and observed near-surface enhancements. The enhancements of CO₂
410 below 3000 masl shown in Fig. 8 for both data and the GEOS model are well correlated. As expected, during April/May we see very little change in the AIEs below 3000 masl, while June and July and August show significant drawdown, followed by enhancements in September and October/November (Fig. 6 and 8). The modeled AIEs in the lower 3000 masl reproduce the observations suggesting that the surface flux of CO₂ throughout most of the ABoVE domain is accurately modeled by GEOS.

415 Despite the overall agreement indicated by aggregated statistics, a closer look shows significant differences in observed and modeled CO₂ enhancements for many individual flight days (Fig. 9). Inspection of individual profiles (Fig. 10) reveal that in some cases the model is not capturing near-ground stratification observed in the river valleys of the interior parts of the ABoVE domain. This is not surprising given that the observations have a much higher vertical resolution than the model's vertical
420 resolution, which is ~100m in the PBL. Consequently, the observed mole fraction values are much higher than the model estimates because the model is not able to capture the stratification. However, the overall modeled vertical gradients in CO₂ match the observations suggesting that the large-scale vertical transport of emissions is accurately simulated above ~1000 masl. As an example, the set of profiles from July 10 (Fig. 10) demonstrates that, although infrequent, high PBL heights and emissions from
425 fires (as indicated by large (>400 ppb) enhancements in CO) add some uncertainty to the AIE values. Both of these factors impact the mean free tropospheric correction and altitude of integration that we have chosen to accurately capture the total CO₂ enhancement from the surface fluxes.

3.3.2. CH₄ AIE

430 Although the correlation between the observed and modeled AIEs of CH₄ is significant, they are not as good as they are for CO₂. In particular, we see some clear biases in the seasonality where the enhancements in the early part of the season are underestimated by the model while the enhancements in the later part of the season are overestimated. This is demonstrated both by the comparisons of AIEs (Fig. 8) and of mole fraction enhancements below 3000 masl (Fig. 9) where the mean difference (observed – modeled) switches from positive to negative over the course of the study period. The Arctic-CAP profile observations provide a critical point of comparison to which future surface flux models of CH₄ can be compared, helping to identify areas where process improvements are needed.

3.3.3. CO AIE

The comparison of observed and modeled AIEs of CO is less useful because some of the critical assumptions made for this comparison are designed to shed light on surface processes affecting CO₂ and CH₄. The biggest limitation in the CO simulation for interpreting vertical profile observations appears to be in the accuracy of the vertical distribution of CO emissions. While the model shows an increase in mole fractions during the July and October/November campaigns, the extreme mole fractions in the observations are twice that of the model (Fig. 6). A good example of how the model and the observed mole fractions are different can be seen on July 10, 2017 (Fig. 10) during a flight up the Mackenzie River in the Northwest Territories of Canada. Here, large enhancements of CO (>400 ppb) are observed at altitudes between 3000 and 5000 masl while CH₄ and CO₂ boundary layer enhancements are observed below 3000 masl in most of the profiles measured that day. The ~100 ppb CO/ppm CO₂ ratio and the large CO enhancement not only support the idea that a fire is the source but that the fire is nearby (<100 km). Both the magnitude and altitude of the CO enhancement point to a few critical limitations in the model that were less important for CO₂ and CH₄. First, most GCMs, including GEOS, do not take into account the massive heat source that fires provide to correctly model the injection of fire emissions above the boundary layer. Second, the fire radiative power observations used to estimate emissions can be obscured by thick clouds or aerosols resulting in the emissions estimates missing some fire hotspots. Third, the heterogenous nature of fires as a surface source of CO means that any inaccuracies in horizontal transport or location of the fire will play a large role in the ability of the model to accurately reproduce the observations. Fourth, the lack of diurnal cycle in biomass burning emissions from the emission database (QFED; Table 1) may result in ‘temporal aggregation errors’, whereby the model simulations may miss the high emission values that coincide with the daytime aircraft observations.

460 3.3.4. Model-data mismatch over ecoregions

The bulk quantity AIE can be used to evaluate surface flux models with aircraft profiles at the regional-scale (Fig. 11). For most regions and times of year, the difference in CO₂ AIEs is not statistically significant; however, there are certain regions such as the Northern Tundra of Alaska, where the modeled CO₂ AIEs are significantly different and amplify a pattern that is observed over other regions. In early spring, the model slightly overestimates observed boundary layer enhancements but a month

later the model underestimates drawdown. Figures 6 and 11 suggest that the peak in early-summer model drawdown in CO₂ is preceding the observed CO₂ drawdown. The difference between observed and modeled enhancements change sign again during the July flight in Northern Tundra Alaska with an underestimation of the drawdown. Similar patterns can be observed in the Canadian Boreal Cordillera, suggesting that the timing of the summertime drawdown is too early in the model in this region. Over the same period, however, comparisons over the Western Alaska Tundra depict opposite patterns (although far more subtle). While the offsets in the fall months are smaller, there is the suggestion that the enhancements in the Southern Arctic and Canadian Taiga ecoregions are both underestimated in the model. For CH₄, the seasonal bias (underestimation in the spring and overestimation between July-September) in the AIEs between observations and models stands out as the most significant feature. The notable exceptions are again the Northern Tundra of Alaska and Canadian Boreal Cordillera, where CH₄ AIEs in July and at the end of October are significantly underestimated. For reasons explained earlier, the CO comparison is less informative. However, if one were to analyze data from the month of September, which had no significant influence from fires in the free troposphere, it would suggest that the model continues to underestimate the impact of CO emissions across all regions.

3.3.5. Separating local, region and global vertical gradients

By extracting enhancements below 3000 masl from the observations and the model, we have largely separated two major sources of biases and uncertainty in a model-data comparison – vertical transport and offsets in background mole fraction. However, it should be acknowledged that gradients between the boundary layer and free troposphere are not controlled exclusively by local fluxes and that in the Arctic, in particular, vertical gradients can be controlled by non-local influences. To explore the impact of long-range transport, Parazoo et al. (2016) performed three simulations to better understand the drivers of the vertical gradient over Alaska and found that 48% of the amplitude (April/May-July/August) in the seasonal vertical gradient was driven by local fluxes from Alaska while the rest was driven by fluxes from the rest of the Arctic (11%) and low latitude (<60N, 41%). For CO₂, the impact of long-range transport to the vertical gradient is complicated by the difference in timing of the initial drawdown in the spring and the uptick in the fall at low latitudes verses that of high latitudes. The earlier drawdown of CO₂ at low latitudes and the transport of that air via the free troposphere to the Arctic significantly reduces the negative vertical gradient in the Arctic. At the same time, the early uptick of CO₂ mole fraction in the Arctic relative to the low latitudes enhances the positive vertical gradient in the early fall (Parazoo et al., 2016).

To account for the background vertical gradient in CH₄ entering the contiguous US, Baier et al. (2020) and Lan et al. (2019) subtracted 12-15 ppt from the vertical gradient to account for a preexisting gradient in CH₄ coming onto the continent. Analysis of the background gradient suggests that this preexisting vertical gradient is a combination of upstream emissions and wind shear, which separates the origin of the boundary layer air from that of the free troposphere. Large meridional gradients in CH₄, such as those observed in the mid-latitudes, will drive depletion of the free troposphere relative to that of the boundary layer over the Arctic. Similarly, CO vertical gradients will also be affected by non-local fluxes and wind shear between the boundary layer and the free troposphere. In the case of CO and CH₄, there is also likely to be a vertical gradient that is influenced by the oxidation of these molecules.

However, given the relatively long residence time of these molecules and the low sampling altitude in the free troposphere (between 3000 and 5000 masl) of this experiment, this effect is small.

From this perspective, the preexisting vertical gradient outside the domain of interest illustrates the importance of the model accuracy in non-local fluxes and the importance of long-range transport in the analysis. One approach ensuring a better boundary conditions is to use a global inversion (e.g. CarbonTracker (Peters et al., 2007)) to initialize the local region where the prognostic flux model is then run to simulate local fields as is done to initialize regional Lagrangian inversion models (e.g. Hu et al., 2019).

3.3.6. AIEs as a tool for benchmarking fluxes

This comparison of AIEs from Arctic-CAP and GEOS demonstrates one of the many values of the aircraft profiles as a metric for evaluating model performance. In a similar vein, Stephens et al. (2007) used the vertical gradient to evaluate the model performance, which pointed out significant errors both from the surface flux models and the vertical transport in the Transcom 3 inversions (Gurney et al., 2002; 2004). The AIE approach has also been used extensively in the Amazon and Arctic as a means of optimizing fluxes in an inversion framework. Zhou et al. (2002), Miller et al. (2007) and Gatti et al. (2010; 2014) have all used some form of AIE from aircraft profiles to estimate surface fluxes of CO₂ and CH₄ in the Amazon basin. Similarly, Zhang et al. (2014), Hartery et al. (2018) and Commane et al. (2017) use the AIE to produce a set of optimized fluxes CH₄ and CO₂ in the Alaska region. This approach to quantifying regional fluxes has significant advantages over other approaches because it is less dependent on an accurate simulation of vertical transport and boundary layer height as noted point out in section 3.2.3. However, even in this instance, there is a need to calculate the average influence of the boundary layer enhancements and this can change dramatically depending on the accuracy of the modelled boundary layer height relative to the integration height of the AIE. In the comparison between observed and modelled AIE presented in this study the focus is on benchmarking a given model's ability to reproduce the AIE in different regions and seasons to objectively quantify how this model might do as conditions change, as is expected with changing climate. From this perspective the need for an accurate simulation of vertical transport largely disappears because the near-field fluxes are just being evaluated, not being computed but just evaluated. The obvious caveat to this approach is that changing climate will bring with it different covariations in temperature, water, radiation and nutrient availability that cannot be reproduced over this time and space domain. While this approach does not replace model benchmarking using eddy covariance measurements, it provides an important view of how modelled processes reproduce observations over scales of 1-3 days and 10-100s of kms.

4. Conclusions

The Arctic-CAP campaign was composed of six different research missions from April to November 2017. It sampled CO₂, CH₄ and CO vertical profiles from the surface to 5000 masl across the ABoVE domain in Alaska and Northwestern Canada, covering six major Arctic ecoregions. Arctic-CAP airborne surveys included large Tundra and Boreal ecosystems that are the likely sources of large

changes in the seasonal cycle of CO₂ and have been the subject of great speculation about future emissions of CH₄.

545 Arctic-CAP's CO₂, CH₄ and CO profiles provide an excellent basis for evaluating the surface flux
models used within state-of-the-art atmospheric transport models, and thus, are an important tool for
understanding carbon cycle feedbacks. Comparisons of Arctic-CAP CO₂, CH₄ and CO observations
against GEOS model show the following main results. For CO₂, the flux model (land/ocean biosphere
and fossil fuel) reproduces seasonal and regional depletions and enhancements observed by aircraft
550 profiles after adjusting for small systematic offsets. For CH₄, the model simulations agree reasonably
well with the observed vertical profiles, but the model underestimates CH₄ enhancements in the spring
and overestimates it in the fall. Modeled North Slope CH₄ is underestimated throughout the
measurement period pointing to deficiencies in the wetland flux specifications over this ecoregion. For
CO, the comparison between modeled and observed values were confounded by large biomass burning
555 enhancements in the free troposphere that were not captured in the model. Despite these minor
shortcomings, the forward model estimates for CO₂ and CH₄ represent a marked improvement in
model-data differences compared to those done previously for CARVE (Chang et al., 2014; Commane
et al., 2017). Results and the flux budgets demonstrate that model representation of CO₂ and CH₄ for
northern high-latitude ecosystems have advanced significantly since the state-of-the-science survey by
560 Fisher et al. (2014). Inversions of the Arctic-CAP data using these fluxes as the prior estimate should
further refine the flux estimates and the budget for the ABoVE domain. We note that our comparisons
used only GEOS forward-model values and slightly different model-data mismatches may be obtained
by using a different transport model.
This study highlights the value of collocated airborne CO₂, CH₄ and CO vertical profiles for quantifying
565 model strengths and weaknesses and for benchmarking fluxes over larger spatial and temporal scales
than is offered by EC comparisons. Such evaluation information is essential to improve model
characterization of both surface-atmosphere fluxes and to improve our confidence in the accuracy of
projections of future conditions. We strongly recommend regular, systematic CO₂, CH₄ and CO vertical
profile observations across the Arctic as an important and cost-effective method to monitor the Arctic
570 for abrupt transformations or potential tipping points in the permafrost-carbon system.

5. Data Availability

Arctic-CAP *in situ* data can be found at <https://doi.org/10.3334/ORNLDAAAC/1658>

6. Sample Availability

N/A

575 7. Video supplement

N/A

8. Supplement link

N/A

9. Team List

580 N/A

10. Author contributions

CS, KM, CM, AC did experimental design. CS, TN, SW, KM carried out experiment. CS, AC, CM, KM, RB, SW, LS, LH helped with manuscript.

11. Competing interests

585 The authors declare that they have no conflict of interest.

12. Disclaimer

N/A

13. Special issue statement

N/A

590 14. Acknowledgements

This research was supported by the NASA Terrestrial Ecology Program award #NNX17AC61A, “Airborne Seasonal Survey of CO₂ and CH₄ Across ABOVE Domain”, as part of the Arctic-Boreal Vulnerability Experiment (ABOVE). A portion of the research presented in this paper was performed at the Jet Propulsion Laboratory, California Institute of Technology, under contract with the National Aeronautics and Space Administration. GEOS model runs and the work of AC was supported by funding from the NASA ROSES-2016 Grant/Cooperative Agreement NNX17AD69A. We thank the Thomas Lauvaux and 2 anonymous reviewers for their hard work in reviewing this manuscript.

15. References

- Allen, M., Erickson, D., Kendall, W., Fu, J., Ott, L., and Pawson, S.: The influence of internal model variability in GEOS-5 on interhemispheric CO₂ exchange, *Journal of Geophysical Research-Atmospheres*, 117, 10.1029/2011jd017059, 2012.
- 600 Arora, V. K., Katavouta, A., Williams, R. G., Jones, C. D., Brovkin, V., Friedlingstein, P., Schwinger, J., Bopp, L., Boucher, O., Cadule, P., Chamberlain, M. A., Christian, J. R., Delire, C., Fisher, R. A., Hajima, T., Ilyina, T., Joetzjer, E., Kawamiya, M., Koven, C. D., Krasting, J. P., Law, R. M., Lawrence, D. M., Lenton, A., Lindsay, K., Pongratz, J., Raddatz, T., Séférian, R., Tachiiri, K., Tjiputra, J. F., Wiltshire, A., Wu, T., and Ziehn, T.: Carbon-concentration and carbon-climate feedbacks in CMIP6 models and their comparison to CMIP5 models, *Biogeosciences*, 17, 4173-4222, 10.5194/bg-17-4173-2020, 2020.
- 605 Baier, B. C., Sweeney, C., Choi, Y., Davis, K. J., DiGangi, J. P., Feng, S., Fried, A., Halliday, H., Higgs, J., Lauvaux, T., Miller, B. R., Montzka, S. A., Newberger, T., Nowak, J. B., Patra, P., Richter, D., Walega, J., and Weibring, P.: Multispecies Assessment of Factors Influencing Regional CO₂ and CH₄ Enhancements During the Winter 2017 ACT-America Campaign, *Journal of Geophysical Research-Atmospheres*, 125, 10.1029/2019jd031339, 2020.
- 610 Baldocchi, D. D., Krebs, T., and Leclerc, M. Y.: "Wet/dry Daisyworld": a conceptual tool for quantifying the spatial scaling of heterogeneous landscapes and its impact on the subgrid variability of energy fluxes, *Tellus Ser. B-Chem. Phys. Meteorol.*, 57, 175-188, 10.1111/j.1600-0889.2005.00149.x, 2005.
- Bosilovich, M. G., Chern, J.-D., Mocko, D., Robertson, F. R., and da Silva, A. M.: Evaluating Observation Influence on Regional Water Budgets in Reanalyses, *J. Clim.*, 28, 3631-3649, 10.1175/jcli-d-14-00623.1, 2015.
- 615 Chang, R. Y. W., Miller, C. E., Dinardo, S. J., Karion, A., Sweeney, C., Daube, B. C., Henderson, J. M., Mountain, M. E., Eluszkiewicz, J., Miller, J. B., Bruhwiler, L. M. P., and Wofsy, S. C.: Methane emissions from Alaska in 2012 from CARVE airborne observations, *Proc. Natl. Acad. Sci. U. S. A.*, 111, 16694-16699, 10.1073/pnas.1412953111, 2014.
- Chou, W. W., Wofsy, S. C., Harriss, R. C., Lin, J. C., Gerbig, C., and Sachse, G. W.: Net fluxes of CO₂ in Amazonia derived from aircraft observations, *Journal of Geophysical Research: Atmospheres*, 107, ACH 4-1-ACH 4-15, <https://doi.org/10.1029/2001JD001295>, 2002.
- 620 Commane, R., Lindaas, J., Benmergui, J., Luus, K. A., Chang, R. Y. W., Daube, B. C., Euskirchen, E. S., Henderson, J. M., Karion, A., Miller, J. B., Miller, S. M., Parazoo, N. C., Randerson, J. T., Sweeney, C., Tans, P., Thoning, K., Veraverbeke, S., Miller, C. E., and Wofsy, S. C.: Carbon dioxide sources from Alaska driven by increasing early winter respiration from Arctic tundra, *Proc. Natl. Acad. Sci. U. S. A.*, 114, 5361-5366, 10.1073/pnas.1618567114, 2017.
- 625 Conley, S. A., Faloon, I. C., Lenschow, D. H., Karion, A., and Sweeney, C.: A Low-Cost System for Measuring Horizontal Winds from Single-Engine Aircraft, *J. Atmos. Ocean. Technol.*, 31, 1312-1320, 10.1175/jtech-d-13-00143.1, 2014.
- Crippa, M., Guizzardi, D., Muntean, M., Schaaf, E., Dentener, F., van Aardenne, J. A., Monni, S., Doering, U., Olivier, J. G. J., Pagliari, V., and Janssens-Maenhout, G.: Gridded emissions of air pollutants for the period 1970–2012 within EDGAR v4.3.2, *Earth Syst. Sci. Data*, 10, 1987-2013, 10.5194/essd-10-1987-2018, 2018.
- 630 Darmenov, A. S., and Da Silva, A. M.: The Quick Fire Emissions Dataset (QFED): Documentation of versions 2.1, 2.2 and 2.4., *NASA Technical Report Series on Global Modeling and Data Assimilation*, 38, 2015.
- Duncan, B. N., Strahan, S. E., Yoshida, Y., Steenrod, S. D., and Livesey, N.: Model study of the cross-tropopause transport of biomass burning pollution, *Atmos. Chem. Phys.*, 7, 3713-3736, 10.5194/acp-7-3713-2007, 2007.
- 635 Fisher, J. B., Sikka, M., Oechel, W. C., Huntzinger, D. N., Melton, J. R., Koven, C. D., Ahlstrom, A., Arain, M. A., Baker, I., Chen, J. M., Ciais, P., Davidson, C., Dietze, M., El-Masri, B., Hayes, D., Huntingford, C., Jain, A. K., Levy, P. E., Lomas, M. R., Poulter, B., Price, D., Sahoo, A. K., Schaefer, K., Tian, H., Tomelleri, E., Verbeeck, H., Viovy, N., Wania, R., Zeng, N., and Miller, C. E.: Carbon cycle uncertainty in the Alaskan Arctic, *Biogeosciences*, 11, 4271-4288, 10.5194/bg-11-4271-2014, 2014.
- Floerchinger, C., McKain, K., Bonin, T., Peischl, J., Biraud, S. C., Miller, C., Ryerson, T. B., Wofsy, S. C., and Sweeney, C.: Methane emissions from oil and gas production on the North Slope of Alaska, *Atmospheric Environment*, 218, 10.1016/j.atmosenv.2019.116985, 2019.
- 640 Freitas, S. R., Longo, K. M., Chatfield, R., Latham, D., Silva Dias, M. A. F., Andreae, M. O., Prins, E., Santos, J. C., Gielow, R., and Carvalho, J. A., Jr.: Including the sub-grid scale plume rise of vegetation fires in low resolution atmospheric transport models, *Atmospheric Chemistry and Physics*, 7, 3385-3398, 10.5194/acp-7-3385-2007, 2007.

- 645 Gatti, L. V., Miller, J. B., D'Amelio, M. T. S., Martinewski, A., Basso, L. S., Gloor, M. E., Wofsy, S., and Tans, P.: Vertical profiles of CO₂ above eastern Amazonia suggest a net carbon flux to the atmosphere and balanced biosphere between 2000 and 2009, *Tellus Ser. B-Chem. Phys. Meteorol.*, 62, 581-594, 10.1111/j.1600-0889.2010.00484.x, 2010.
- Gatti, L. V., Gloor, M., Miller, J. B., Doughty, C. E., Malhi, Y., Domingues, L. G., Basso, L. S., Martinewski, A., Correia, C. S. C., Borges, V. F., Freitas, S., Braz, R., Anderson, L. O., Rocha, H., Grace, J., Phillips, O. L., and Lloyd, J.: Drought
650 sensitivity of Amazonian carbon balance revealed by atmospheric measurements, *Nature*, 506, 76-80, 10.1038/nature12957, 2014.
- Gelaro, R., McCarty, W., Suárez, M. J., Todling, R., Molod, A., Takacs, L., Randles, C. A., Darmenov, A., Bosilovich, M. G., Reichle, R., Wargan, K., Coy, L., Cullather, R., Draper, C., Akella, S., Buchard, V., Conaty, A., da Silva, A. M., Gu, W., Kim, G.-K., Koster, R., Lucchesi, R., Merkova, D., Nielsen, J. E., Partyka, G., Pawson, S., Putman, W., Rienecker, M., Schubert, S.
655 D., Sienkiewicz, M., and Zhao, B.: The Modern-Era Retrospective Analysis for Research and Applications, Version 2 (MERRA-2), *J. Clim.*, 30, 5419-5454, 10.1175/JCLI-D-16-0758.1, 2017.
- Gockede, M., Markkanen, T., Mauder, M., Arnold, K., Leps, J. P., and Foken, T.: Validation of footprint models using natural tracer measurements from a field experiment, *Agric. For. Meteorol.*, 135, 314-325, 10.1016/j.agrformet.2005.12.008, 2005.
- Gourdji, S. M., Mueller, K. L., Yadav, V., Huntzinger, D. N., Andrews, A. E., Trudeau, M., Petron, G., Nehrkorn, T.,
660 Eluszkiewicz, J., Henderson, J., Wen, D., Lin, J., Fischer, M., Sweeney, C., and Michalak, A. M.: North American CO₂ exchange: inter-comparison of modeled estimates with results from a fine-scale atmospheric inversion, *Biogeosciences*, 9, 457-475, 10.5194/bg-9-457-2012, 2012.
- Gurney, K. R., Law, R. M., Denning, A. S., Rayner, P. J., Baker, D., Bousquet, P., Bruhwiler, L., Chen, Y. H., Ciais, P., Fan, S., Fung, I. Y., Gloor, M., Heimann, M., Higuchi, K., John, J., Maki, T., Maksyutov, S., Masarie, K., Peylin, P., Prather, M.,
665 Pak, B. C., Randerson, J., Sarmiento, J., Taguchi, S., Takahashi, T., and Yuen, C. W.: Towards robust regional estimates of CO₂ sources and sinks using atmospheric transport models, *Nature*, 415, 626-630, 2002.
- Gurney, K. R., Law, R. M., Denning, A. S., Rayner, P. J., Pak, B. C., Baker, D., Bousquet, P., Bruhwiler, L., Chen, Y. H., Ciais, P., Fung, I. Y., Heimann, M., John, J., Maki, T., Maksyutov, S., Peylin, P., Prather, M., and Taguchi, S.: Transcom 3
670 inversion intercomparison: Model mean results for the estimation of seasonal carbon sources and sinks, *Global Biogeochemical Cycles*, 18, 10.1029/2003GB002111, 2004.
- Hartery, S., Commane, R., Lindaas, J., Sweeney, C., Henderson, J., Mountain, M., Steiner, N., McDonald, K., Dinardo, S. J., Miller, C. E., Wofsy, S. C., and Chang, R. Y. W.: Estimating regional-scale methane flux and budgets using CARVE aircraft
675 measurements over Alaska, *Atmos. Chem. Phys.*, 18, 185-202, 10.5194/acp-18-185-2018, 2018.
- Hu, L., Andrews, A. E., Thoning, K. W., Sweeney, C., Miller, J. B., Michalak, A. M., Dlugokencky, E., Tans, P. P., Shiga, Y. P., and Mountain, M.: Enhanced North American carbon uptake associated with El Niño, *Science advances*, 5, eaaw0076,
2019.
- Hugelius, G., Strauss, J., Zubrzycki, S., Harden, J. W., Schuur, E. A. G., Ping, C. L., Schirmer, L., Grosse, G., Michaelson, G. J., Koven, C. D., O'Donnell, J. A., Elberling, B., Mishra, U., Camill, P., Yu, Z., Palmtag, J., and Kuhry, P.: Estimated stocks
680 of circumpolar permafrost carbon with quantified uncertainty ranges and identified data gaps, *Biogeosciences*, 11, 6573-6593, 10.5194/bg-11-6573-2014, 2014.
- Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Muntean, M., and Schaaf, E.: Emissions Database for Global Atmospheric Research, version v4.3.2 part I Greenhouse gases (time-series), in: Joint Research Centre (JRC), v4.3.2 ed., European
Commission, 2017.
- Karion, A., Sweeney, C., Wolter, S., Newberger, T., Chen, H., Andrews, A., Kofler, J., Neff, D., and Tans, P.: Long-term
685 greenhouse gas measurements from aircraft, *Atmos. Meas. Tech.*, 6, 511-526, 10.5194/amt-6-511-2013, 2013.
- Karion, A., Sweeney, C., Kort, E. A., Shepson, P. B., Brewer, A., Cambaliza, M., Conley, S. A., Davis, K., Deng, A., Hardesty, M., Herndon, S. C., Lauvaux, T., Lavoie, T., Lyon, D., Newberger, T., Petron, G., Rella, C., Smith, M., Wolter, S., Yacovitch, T. I., and Tans, P.: Aircraft-Based Estimate of Total Methane Emissions from the Barnett Shale Region, *Environ. Sci. Technol.*,
49, 8124-8131, 10.1021/acs.est.5b00217, 2015.
- 690 Koven, C. D., Ringeval, B., Friedlingstein, P., Ciais, P., Cadule, P., Khvorostyanov, D., Krinner, G., and Tarnocai, C.: Permafrost carbon-climate feedbacks accelerate global warming, *Proc. Natl. Acad. Sci. U. S. A.*, 108, 14769-14774, 10.1073/pnas.1103910108, 2011.

- 695 Lan, X., Tans, P., Sweeney, C., Andrews, A., Dlugokencky, E., Schwietzke, S., Kofler, J., McKain, K., Thoning, K., and Crotwell, M.: Long-Term Measurements Show Little Evidence for Large Increases in Total US Methane Emissions Over the Past Decade, *Geophysical Research Letters*, 46, 4991-4999, 2019.
- Lauvaux, T., Schuh, A. E., Uliasz, M., Richardson, S., Miles, N., Andrews, A. E., Sweeney, C., Diaz, L. I., Martins, D., Shepson, P. B., and Davis, K. J.: Constraining the CO₂ budget of the corn belt: exploring uncertainties from the assumptions in a mesoscale inverse system, *Atmospheric Chemistry and Physics*, 12, 337-354, 10.5194/acp-12-337-2012, 2012.
- 700 Lawrence, D. M., Koven, C. D., Swenson, S. C., Riley, W. J., and Slater, A. G.: Permafrost thaw and resulting soil moisture changes regulate projected high-latitude CO₂ and CH₄ emissions, *Environmental Research Letters*, 10, 10.1088/1748-9326/10/9/094011, 2015.
- McGuire, A. D., Koven, C., Lawrence, D. M., Clein, J. S., Xia, J., Beer, C., Burke, E., Chen, G., Chen, X., Delire, C., Jafarov, E., MacDougall, A. H., Marchenko, S., Nicolsky, D., Peng, S., Rinke, A., Saito, K., Zhang, W., Alkama, R., Bohn, T. J., Ciais, P., Decharme, B., Ekici, A., Gouttevin, I., Hajima, T., Hayes, D. J., Ji, D., Krinner, G., Lettenmaier, D. P., Luo, Y., Miller, P. A., Moore, J. C., Romanovsky, V., Schaedel, C., Schaefer, K., Schuur, E. A. G., Smith, B., Sueyoshi, T., and Zhuang, Q.: Variability in the sensitivity among model simulations of permafrost and carbon dynamics in the permafrost region between 1960 and 2009, *Global Biogeochemical Cycles*, 30, 1015-1037, 10.1002/2016gb005405, 2016.
- 705 Mekkonen, Z. A., Grant, R. F., and Schwalm, C.: Sensitivity of modeled NEP to climate forcing and soil at site and regional scales: Implications for upscaling ecosystem models, *Ecological Modelling*, 320, 241-257, <https://doi.org/10.1016/j.ecolmodel.2015.10.004>, 2016.
- 710 Miller, C. E., Dinardo, S. J., Team, C. S., and Ieee: CARVE: The Carbon in Arctic Reservoirs Vulnerability Experiment, in: 2012 Ieee Aerospace Conference, IEEE Aerospace Conference Proceedings, Ieee, New York, 2012.
- Miller, C. E., Griffith, P. C., Goetz, S. J., Hoy, E. E., Pinto, N., McCubbin, I. B., Thorpe, A. K., Hofton, M., Hodkinson, D., Hansen, C., Woods, J., Larson, E., Kasischke, E. S., and Margolis, H. A.: An overview of ABoVE airborne campaign data 715 acquisitions and science opportunities, *Environmental Research Letters*, 14, 10.1088/1748-9326/ab0d44, 2019.
- Miller, J. B., Gatti, L. V., d'Amelio, M. T. S., Crotwell, A. M., Dlugokencky, E. J., Bakwin, P., Artaxo, P., and Tans, P. P.: Airborne measurements indicate large methane emissions from the eastern Amazon basin, *Geophys. Res. Lett.*, 34, 2007.
- Miller, S. M., Commane, R., Melton, J. R., Andrews, A. E., Benmergui, J., Dlugokencky, E. J., Janssens-Maenhout, G., Michalak, A. M., Sweeney, C., and Worthy, D. E. J.: Evaluation of wetland methane emissions across North America using 720 atmospheric data and inverse modeling, *Biogeosciences*, 13, 1329-1339, 10.5194/bg-13-1329-2016, 2016.
- Molod, A., Takacs, L., Suarez, M., and Bacmeister, J.: Development of the GEOS-5 atmospheric general circulation model: evolution from MERRA to MERRA2, *Geoscientific Model Development*, 8, 1339-1356, 10.5194/gmd-8-1339-2015, 2015.
- Mueller, K., Yadav, V., Lopez-Coto, I., Karion, A., Gourджи, S., Martin, C., and Whetstone, J.: Siting Background Towers to Characterize Incoming Air for Urban Greenhouse Gas Estimation: A Case Study in the Washington, DC/Baltimore Area, 725 *Journal of Geophysical Research-Atmospheres*, 123, 2910-2926, 10.1002/2017jd027364, 2018.
- Oda, T., and Maksyutov, S.: A very high-resolution (1 km x 1 km) global fossil fuel CO₂ emission inventory derived using a point source database and satellite observations of nighttime lights, *Atmospheric Chemistry and Physics*, 11, 543-556, 10.5194/acp-11-543-2011, 2011.
- Ott, L., Duncan, B., Pawson, S., Colarco, P., Chin, M., Randles, C., Diehl, T., and Nielsen, E.: Influence of the 2006 Indonesian biomass burning aerosols on tropical dynamics studied with the GEOS-5 AGCM, *Journal of Geophysical Research: Atmospheres*, 115, 10.1029/2009JD013181, 2010.
- 730 Ott, L. E., Pawson, S., Collatz, G. J., Gregg, W. W., Menemenlis, D., Brix, H., Rousseaux, C. S., Bowman, K. W., Liu, J., Eldering, A., Gunson, M. R., and Kawa, S. R.: Assessing the magnitude of CO₂ flux uncertainty in atmospheric CO₂ records using products from NASA's Carbon Monitoring Flux Pilot Project, *Journal of Geophysical Research-Atmospheres*, 120, 734-765, 10.1002/2014jd022411, 2015.
- 735 Parazoo, N. C., Commane, R., Wofsy, S. C., Koven, C. D., Sweeney, C., Lawrence, D. M., Lindaas, J., Chang, R. Y. W., and Miller, C. E.: Detecting regional patterns of changing CO₂ flux in Alaska, *Proc. Natl. Acad. Sci. U. S. A.*, 113, 7733-7738, 10.1073/pnas.1601085113, 2016.
- Peters, W., Jacobson, A., Sweeney, C., Andrews, A., Conway, T., Masarie, K., Miller, J. B., Bruhwiler, L., Petron, G., Hirsch, A., Worthy, D., Werf, G. v. d., Randerson, J. T., Wennberg, P., Krol, M., and Tan, P.: The atmospheric perspective of carbon-dioxide exchange across North America: CarbonTracker, *Proceedings of the National Academy of Sciences of the United States of America (PNAS)*, 2007.

- 745 Pickett-Heaps, C. A., Jacob, D. J., Wecht, K. J., Kort, E. A., Wofsy, S. C., Diskin, G. S., Worthy, D. E. J., Kaplan, J. O., Bey, I., and Drevet, J.: Magnitude and seasonality of wetland methane emissions from the Hudson Bay Lowlands (Canada), *Atmos. Chem. Phys.*, 11, 3773-3779, 10.5194/acp-11-3773-2011, 2011.
- Poulter, B., Ciais, P., Hodson, E., Lischke, H., Maignan, F., Plummer, S., and Zimmermann, N. E.: Plant functional type mapping for earth system models, *Geoscientific Model Development*, 4, 993-1010, 10.5194/gmd-4-993-2011, 2011.
- 750 Rienecker, M. M., Suarez, M. J., Gelaro, R., Todling, R., Bacmeister, J., Liu, E., Bosilovich, M. G., Schubert, S. D., Takacs, L., Kim, G.-K., Bloom, S., Chen, J., Collins, D., Conaty, A., Da Silva, A., Gu, W., Joiner, J., Koster, R. D., Lucchesi, R., Molod, A., Owens, T., Pawson, S., Pegion, P., Redder, C. R., Reichle, R., Robertson, F. R., Ruddick, A. G., Sienkiewicz, M., and Woollen, J.: MERRA: NASA's Modern-Era Retrospective Analysis for Research and Applications, *J. Clim.*, 24, 3624-3648, 10.1175/jcli-d-11-00015.1, 2011.
- Sasai, T., Okamoto, K., Hiyama, T., and Yamaguchi, Y.: Comparing terrestrial carbon fluxes from the scale of a flux tower to the global scale, *Ecological Modelling*, 208, 135-144, <https://doi.org/10.1016/j.ecolmodel.2007.05.014>, 2007.
- 755 Sasakawa, M., Machida, T., Tsuda, N., Arshinov, M., Davydov, D., Fofonov, A., and Krasnov, O.: Aircraft and tower measurements of CO₂ concentration in the planetary boundary layer and the lower free troposphere over southern taiga in West Siberia: Long-term records from 2002 to 2011, *Journal of Geophysical Research: Atmospheres*, 118, 9489-9498, 10.1002/jgrd.50755, 2013.
- 760 Saunio, M., Stavert, A. R., Poulter, B., Bousquet, P., Canadell, J. G., Jackson, R. B., Raymond, P. A., Dlugokencky, E. J., Houweling, S., Patra, P. K., Ciais, P., Arora, V. K., Bastviken, D., Bergamaschi, P., Blake, D. R., Brailsford, G., Bruhwiler, L., Carlson, K. M., Carrol, M., Castaldi, S., Chandra, N., Crevoisier, C., Crill, P. M., Covey, K., Curry, C. L., Etiope, G., Frankenberg, C., Gedney, N., Hegglin, M. I., Höglund-Isaksson, L., Hugelius, G., Ishizawa, M., Ito, A., Janssens-Maenhout, G., Jensen, K. M., Joos, F., Kleinen, T., Krummel, P. B., Langenfelds, R. L., Laruelle, G. G., Liu, L., Machida, T., Maksyutov, S., McDonald, K. C., McNorton, J., Miller, P. A., Melton, J. R., Morino, I., Müller, J., Murgia-Flores, F., Naik, V., Niwa, Y., Noce, S., O'Doherty, S., Parker, R. J., Peng, C., Peng, S., Peters, G. P., Prigent, C., Prinn, R., Ramonet, M., Regnier, P., Riley, W. J., Rosentreter, J. A., Segers, A., Simpson, I. J., Shi, H., Smith, S. J., Steele, L. P., Thornton, B. F., Tian, H., Tohjima, Y., Tubiello, F. N., Tsuruta, A., Viovy, N., Voulgarakis, A., Weber, T. S., van Weele, M., van der Werf, G. R., Weiss, R. F., Worthy, D., Wunch, D., Yin, Y., Yoshida, Y., Zhang, W., Zhang, Z., Zhao, Y., Zheng, B., Zhu, Q., Zhu, Q., and Zhuang, Q.: The Global Methane Budget 2000-2017, *Earth Syst. Sci. Data Discuss.*, 2019, 1-136, 10.5194/essd-2019-128, 2019.
- 770 Schaefer, K., Lantuit, H., Romanovsky, V. E., Schuur, E. A. G., and Witt, R.: The impact of the permafrost carbon feedback on global climate, *Environmental Research Letters*, 9, 10.1088/1748-9326/9/8/085003, 2014.
- Schmid, H. P.: Footprint modeling for vegetation atmosphere exchange studies: a review and perspective, *Agric. For. Meteorol.*, 113, 159-183, 10.1016/s0168-1923(02)00107-7, 2002.
- 775 Schneider von Deimling, T., Meinshausen, M., Levermann, A., Huber, V., Frieler, K., Lawrence, D. M., and Brovkin, V.: Estimating the near-surface permafrost-carbon feedback on global warming, *Biogeosciences*, 9, 649-665, 10.5194/bg-9-649-2012, 2012.
- Schuur, E. A. G., McGuire, A. D., Schaedel, C., Grosse, G., Harden, J. W., Hayes, D. J., Hugelius, G., Koven, C. D., Kuhry, P., Lawrence, D. M., Natali, S. M., Olefeldt, D., Romanovsky, V. E., Schaefer, K., Turetsky, M. R., Treat, C. C., and Vonk, J. E.: Climate change and the permafrost carbon feedback, *Nature*, 520, 171-179, 10.1038/nature14338, 2015.
- 780 Stephens, B. B., Bakwin, P. S., Tans, P. P., Teclaw, R. M., and Baumann, D. D.: Application of a differential fuel-cell analyzer for measuring atmospheric oxygen variations, *J. Atmos. Ocean. Technol.*, 24, 82-94, 10.1175/jtech1959.1, 2007.
- Strode, S. A., Liu, J., Lait, L., Commane, R., Daube, B., Wofsy, S., Conaty, A., Newman, P., and Prather, M.: Forecasting carbon monoxide on a global scale for the ATom-1 aircraft mission: insights from airborne and satellite observations and modeling, *Atmos. Chem. Phys.*, 18, 10955-10971, 10.5194/acp-18-10955-2018, 2018.
- 785 Sweeney, C., Karion, A., Wolter, S., Newberger, T., Guenther, D., Higgs, J. A., Andrews, A. E., Lang, P. M., Neff, D., Dlugokencky, E., Miller, J. B., Montzka, S. A., Miller, B. R., Masarie, K. A., Biraud, S. C., Novelli, P. C., Crotwell, M., Crotwell, A. M., Thoning, K., and Tans, P. P.: Seasonal climatology of CO₂ across North America from aircraft measurements in the NOAA/ESRL Global Greenhouse Gas Reference Network, *Journal of Geophysical Research-Atmospheres*, 120, 5155-5190, 10.1002/2014jd022591, 2015.
- 790 Sweeney, C., Dlugokencky, E., Miller, C. E., Wofsy, S., Karion, A., Dinardo, S., Chang, R. Y. W., Miller, J. B., Bruhwiler, L., Crotwell, A. M., Newberger, T., McKain, K., Stone, R. S., Wolter, S. E., Lang, P. E., and Tans, P.: No significant increase

- in long-term CH₄ emissions on North Slope of Alaska despite significant increase in air temperature, *Geophysical Research Letters*, 43, 6604-6611, 10.1002/2016gl069292, 2016.
- 795 Sweeney, C., and McKain, K.: ABoVE: Atmospheric Profiles of CO, CO₂ and CH₄ Concentrations from Arctic-CAP, 2017, in, ORNL Distributed Active Archive Center, 2019.
- Sweeney, C., McKain, K., Miller, B. R., and Michel, S. E.: ABoVE: Atmospheric Gas Concentrations from Airborne Flasks, Arctic-CAP, 2017, in, ORNL Distributed Active Archive Center, 2020.
- 800 Takahashi, T., Sutherland, S. C., Wanninkhof, R., Sweeney, C., Feely, R. A., Chipman, D. W., Hales, B., Friederich, G., Chavez, F., Sabine, C., Watson, A., Bakker, D. C. E., Schuster, U., Metzl, N., Yoshikawa-Inoue, H., Ishii, M., Midorikawa, T., Nojiri, Y., Kortzinger, A., Steinhoff, T., Hoppema, M., Olafsson, J., Arnarson, T. S., Tilbrook, B., Johannessen, T., Olsen, A., Bellerby, R., Wong, C. S., Delille, B., Bates, N. R., and de Baar, H. J. W.: Climatological mean and decadal change in surface ocean pCO₂, and net sea-air CO₂ flux over the global oceans, *Deep-Sea Res. Part II-Top. Stud. Oceanogr.*, 56, 554-577, 10.1016/j.dsr2.2008.12.009, 2009.
- 805 Thompson, R. L., Sasakawa, M., Machida, T., Aalto, T., Worthy, D., Lavric, J. V., Myhre, C. L., and Stohl, A.: Methane fluxes in the high northern latitudes for 2005-2013 estimated using a Bayesian atmospheric inversion, *Atmospheric Chemistry and Physics*, 17, 3553-3572, 10.5194/acp-17-3553-2017, 2017.
- USGS: 1 meter Digital Elevation Models (DEMs) - USGS National Map 3DEP Downloadable Data Collection, in: ScienceBase-Catalogue, edited by: Survey, U. S. G., 2017.
- 810 Van Der Werf, G. R., Randerson, J. T., Collatz, G. J., and Giglio, L.: Carbon emissions from fires in tropical and subtropical ecosystems, *Glob. Change Biol.*, 9, 547-562, 10.1046/j.1365-2486.2003.00604.x, 2003.
- Wania, R., Ross, I., and Prentice, I. C.: Integrating peatlands and permafrost into a dynamic global vegetation model: 1. Evaluation and sensitivity of physical land surface processes, *Global Biogeochemical Cycles*, 23, 10.1029/2008gb003412, 2009.
- 815 Weir, B., Ott, L. E., Collatz, G. J., Kawa, S. R., Poulter, B., Chatterjee, A., Oda, T., and Pawson, S.: Bias-correcting carbon fluxes derived from land-surface satellite data for retrospective and near-real-time assimilation systems, *Atmos. Chem. Phys.*, 21, 9609-9628, 10.5194/acp-21-9609-2021, 2021.
- Welp, L. R., Patra, P. K., Rödenbeck, C., Nemani, R., Bi, J., Piper, S. C., and Keeling, R. F.: Increasing summer net CO₂ uptake in high northern ecosystems inferred from atmospheric inversions and comparisons to remote-sensing NDVI, *Atmos. Chem. Phys.*, 16, 9047-9066, 10.5194/acp-16-9047-2016, 2016.
- 820 Wofsy, S. C., Afshar, S., Allen, H. M., Apel, E., Asher, E. C., Barletta, B., Bent, J., Bian, H., Biggs, B. C., Blake, D. R., Blake, N., Bourgeois, I., Brock, C. A., Brune, W. H., Budney, J. W., Bui, T. P., Butler, A., Campuzano-Jost, P., Chang, C. S., Chin, M., Commane, R., Correa, G., Crounse, J. D., Cullis, P. D., Daube, B. C., Day, D. A., Dean-Day, J. M., Dibb, J. E., Digangi, J. P., Diskin, G. S., Dollner, M., Elkins, J. W., Erdesz, F., Fiore, A. M., Flynn, C. M., Froyd, K., Gesler, D. W., Hall, S. R., Hanisco, T. F., Hannun, R. A., Hills, A. J., Hints, E. J., Hoffman, A., Hornbrook, R. S., Huey, L. G., Hughes, S., Jimenez, J.
- 825 L., Johnson, B. J., Katich, J. M., Keeling, R., Kim, M. J., Kupc, A., Lait, L. R., Lamarque, J. F., Liu, J., McKain, K., McLaughlin, R. J., Meinardi, S., Miller, D. O., Montzka, S. A., Moore, F. L., Morgan, E. J., Murphy, D. M., Murray, L. T., Nault, B. A., Neuman, J. A., Newman, P. A., Nicely, J. M., Pan, X., Paplawsky, W., Peischl, J., Prather, M. J., Price, D. J., Ray, E., Reeves, J. M., Richardson, M., Rollins, A. W., Rosenlof, K. H., Ryerson, T. B., Scheuer, E., Schill, G. P., Schroder, J. C., Schwarz, J. P., St.Clair, J. M., Steenrod, S. D., Stephens, B. B., Strode, S. A., Sweeney, C., Tanner, D., Teng, A. P.,
- 830 Thames, A. B., Thompson, C. R., Ullmann, K., Veres, P. R., Vizenor, N., Wagner, N. L., Watt, A., Weber, R., Weinzierl, B., Wennberg, P., Williamson, C. J., Wilson, J. C., Wolfe, G. M., Woods, C. T., and Zeng, L. H.: ATom: Merged Atmospheric Chemistry, Trace Gases, and Aerosols, in, ORNL Distributed Active Archive Center, 2018.
- Wunch, D., Wennberg, P. O., Messerschmidt, J., Parazoo, N. C., Toon, G. C., Deutscher, N. M., Keppel-Aleks, G., Roehl, C. M., Randerson, J. T., Warneke, T., and Notholt, J.: The covariation of Northern Hemisphere summertime CO₂ with surface temperature in boreal regions, *Atmos. Chem. Phys.*, 13, 9447-9459, 10.5194/acp-13-9447-2013, 2013.
- 835 Zhang, Z., Zimmermann, N. E., Kaplan, J. O., and Poulter, B.: Modeling spatiotemporal dynamics of global wetlands: comprehensive evaluation of a new sub-grid TOPMODEL parameterization and uncertainties, *Biogeosciences*, 13, 1387-1408, 10.5194/bg-13-1387-2016, 2016.

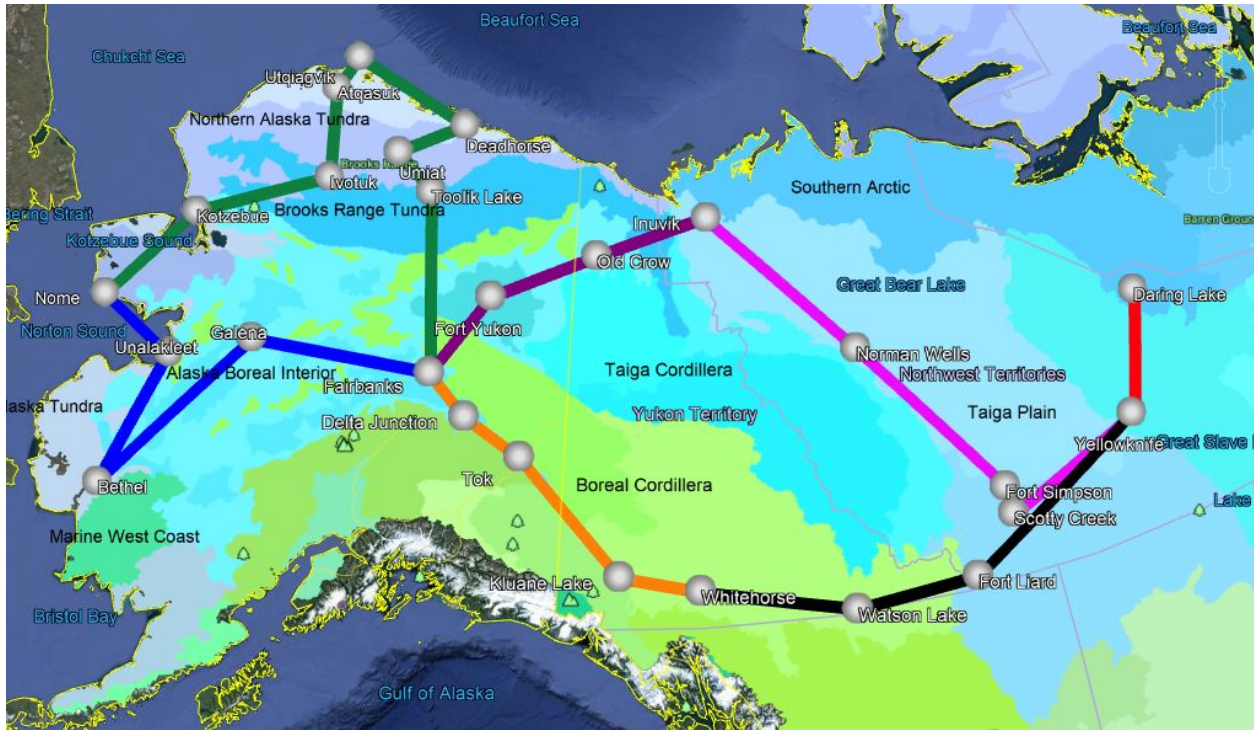
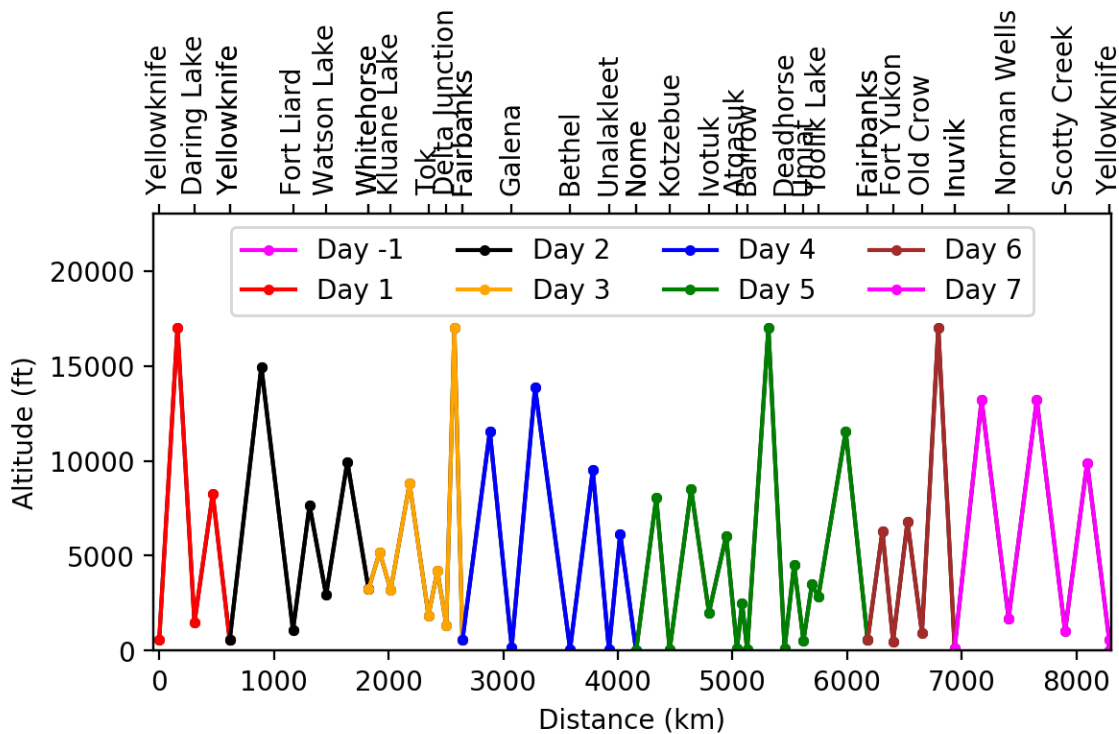
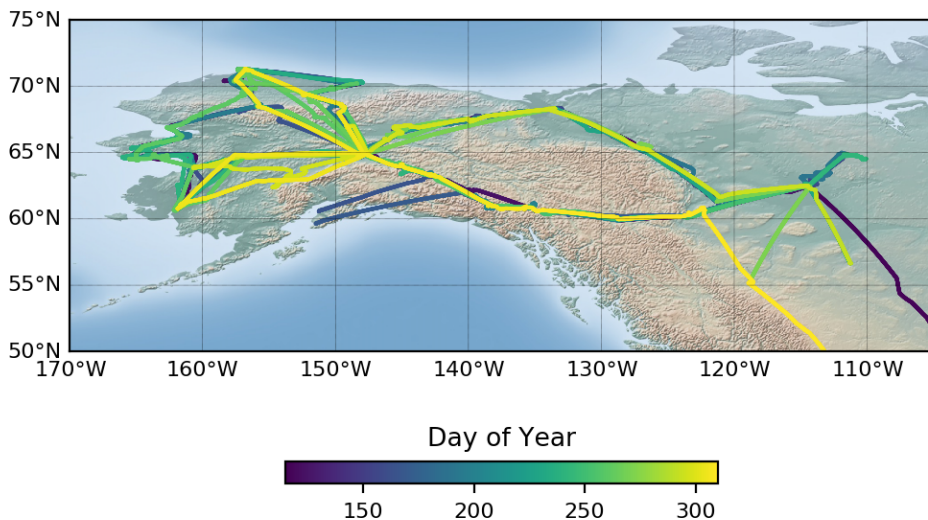


Figure. 1. The Arctic-CAP surveys were designed to sample the Arctic boreal ecosystems of the ABoVE domain. Black text labels represent the six ecoregions covered by this study and white text denote cities and states / provinces. Gray dots depict the locations on which the Arctic-CAP vertical profiles were centered (© Google Earth). Flight track colors represent extent of each (of 7) daily flights (see Figure 2).



850 **Figure 2. Locations and maximum altitudes of the 25 vertical profiles that were acquired during each Arctic-CAP campaign. The colors match the flight lines illustrated in Fig. 1.**



855 **Figure 3 Arctic-CAP flight paths colored by day of year (DOY). Later paths are plotted on top, masking flights from earlier in the year along the same routes. Profile locations span 50-75 °N and 105-165 °W and sampled environmental conditions from the spring thaw (~DOY 125) through the early cold season (> DOY 300) (© Google Maps).**

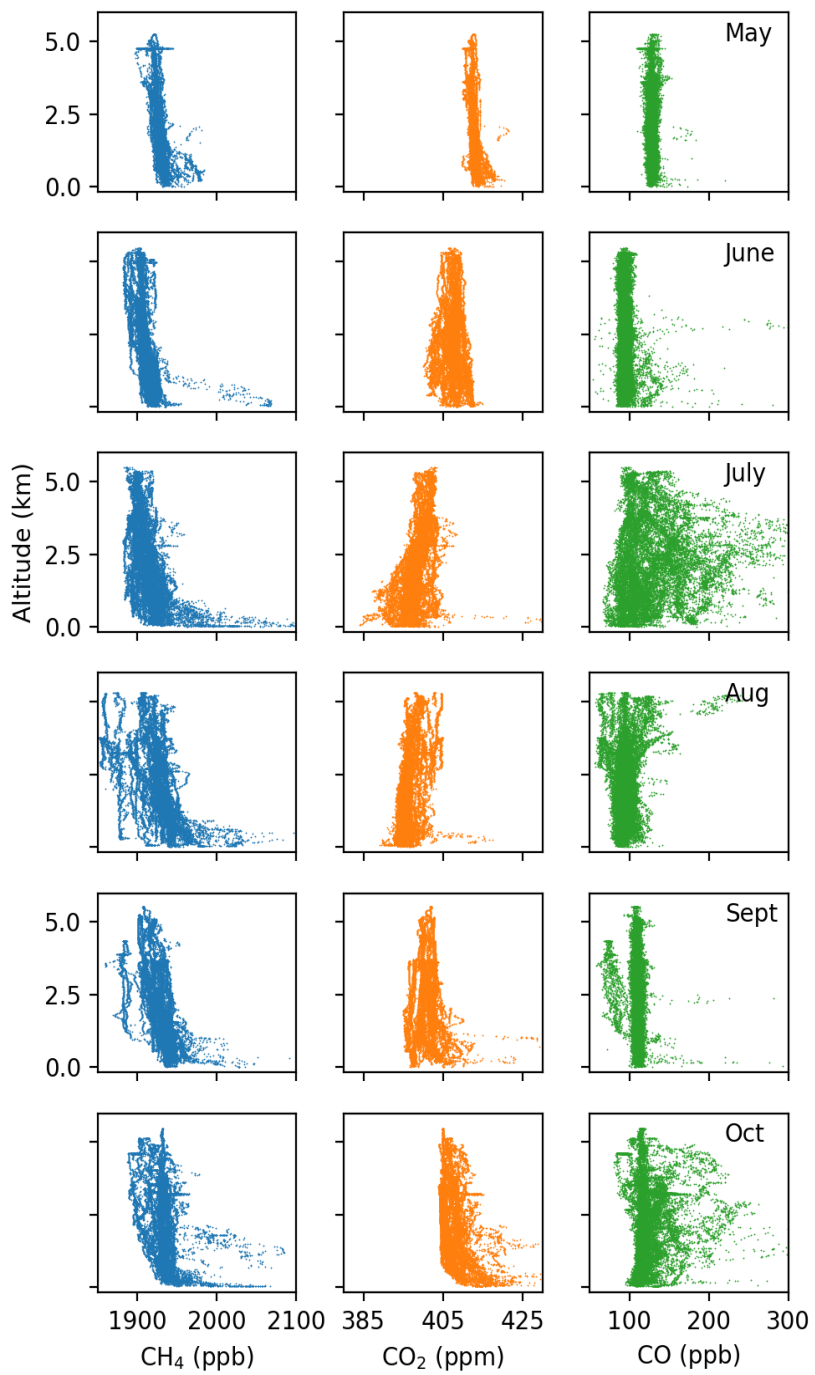


Figure 4. Composite plots of the CH₄ (left column), CO₂ (center column) and CO (right column) measurements acquired during the Arctic-CAP airborne campaign in 2017. Broad seasonal cycle and near surface enhancement (depletions) can be seen as well as the impact of fires to the free tropospheric CO.

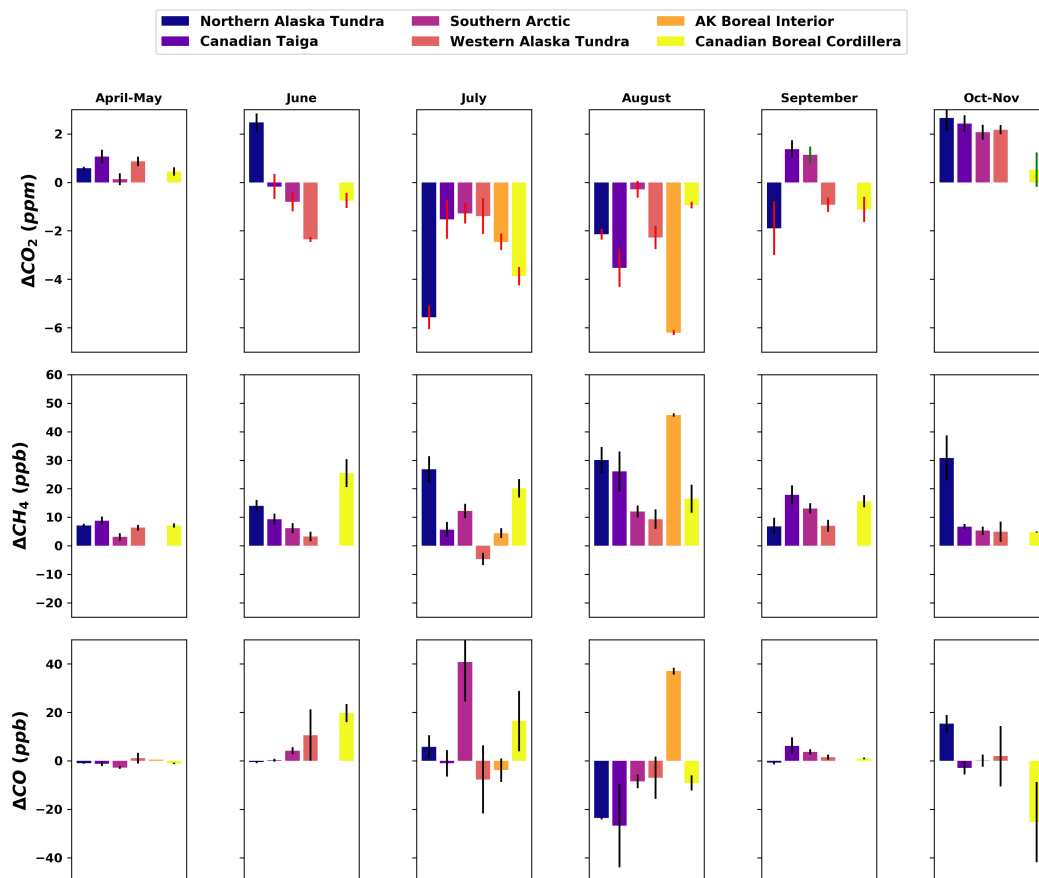


Figure 5. Average gradient between the mean free daily troposphere (> 3000 masl for CO₂ and CH₄ and 4000 masl for CO) and measurements made below 3000 masl during each campaign. Colors refer to the six ecoregions identified in Fig. 1.

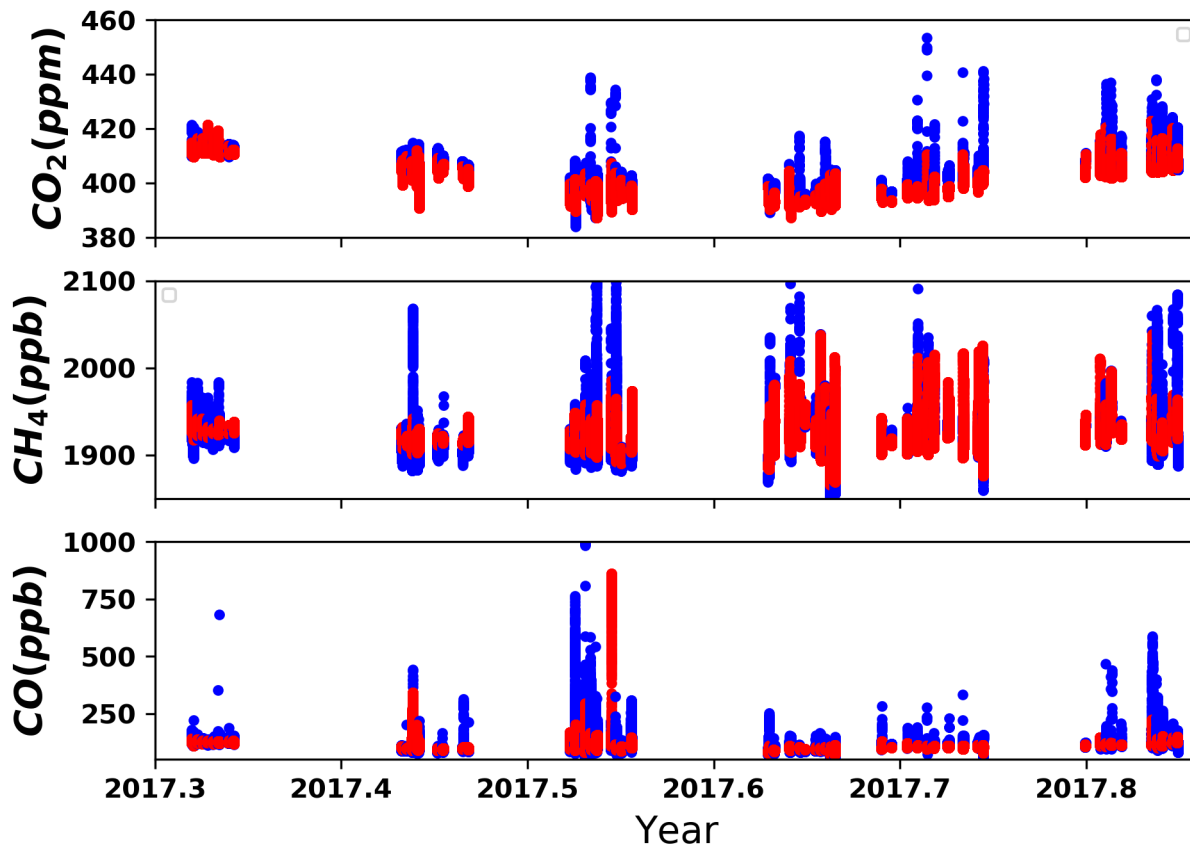
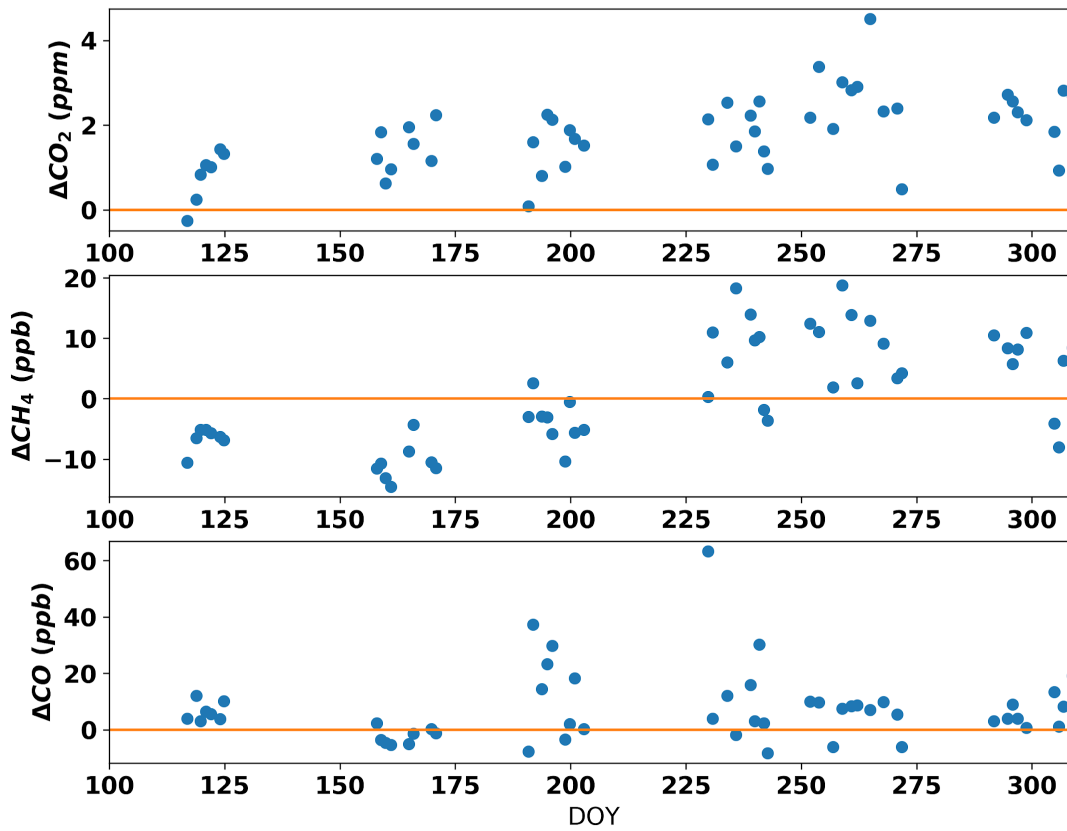


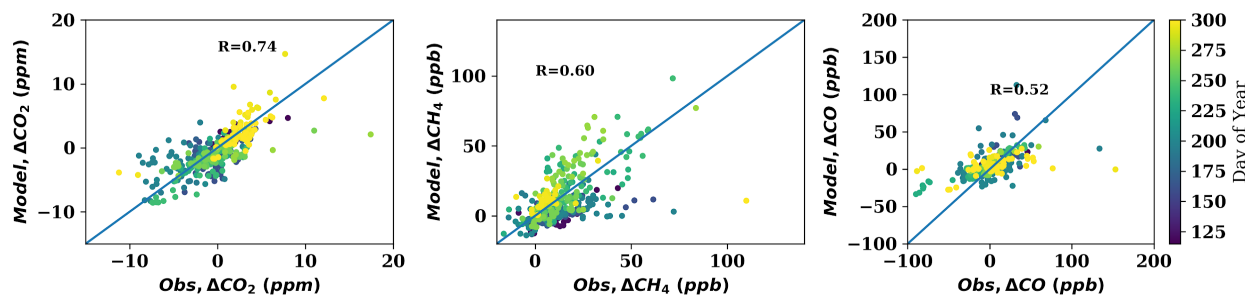
Figure 6. Comparisons of GEOS simulated atmospheric CO₂, CH₄ and CO (red points) versus observed CO₂, CH₄ and CO (blue points) during the Arctic-CAP 2017 campaign show good agreement across campaigns, although the observed data exhibit larger extremes.

870

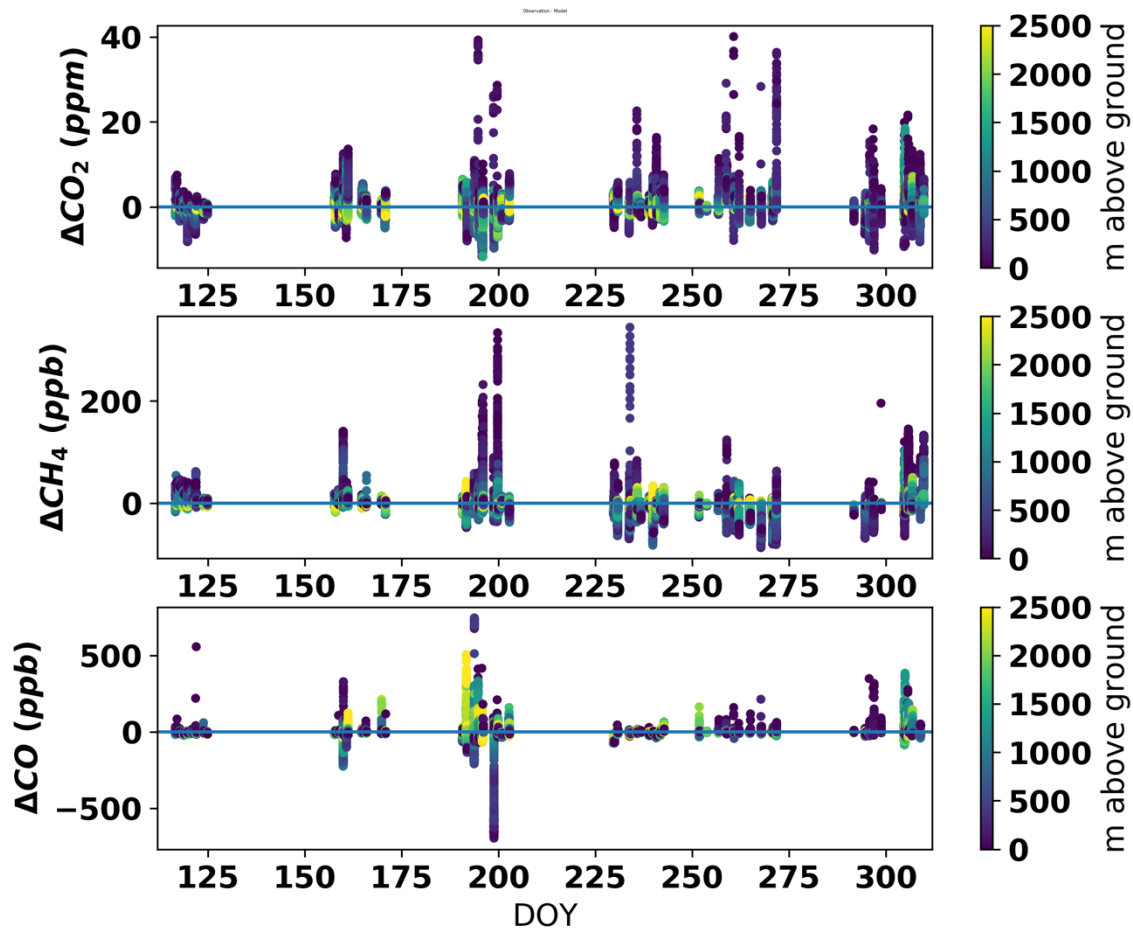


875

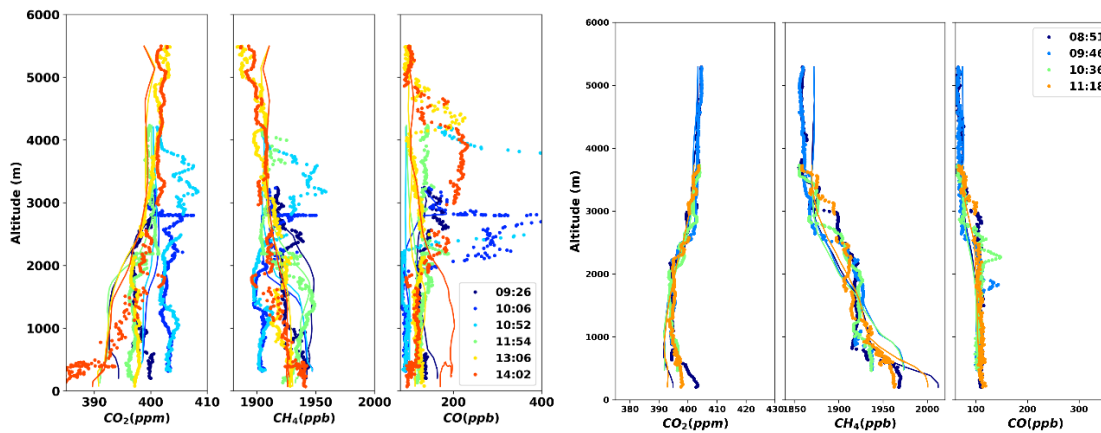
Figure 7. Difference (observations-model) between mean daily free troposphere (3000-5000 masl for CO_2 and CH_4 and 4000-5000 masl for CO) for GEOS simulated and Arctic-CAP observed mole fractions. The GEOS simulations systematically underestimate the mean CO_2 in all months, while the model overestimates CH_4 before DOY 200 and underestimates CH_4 after DOY 200. Simulated CO observations generally agree with the atmospheric observations, although there are sporadic underestimates likely associated with incorrectly modeled fire plumes.



880 **Figure 8.** Modelled versus observed average boundary layer enhancements or depletions in CO_2 , CH_4 and CO for individual profiles from 3000 masl down to the surface level.



885 Figure 9. Observation-model differences in mole fractions below 3000 masl. Corrections have been made for observation-model offsets above 3000 masl (Fig. 7). Colors show the altitude of each deviation. Dark blue indicates differences near the surface while yellow indicates differences near 3000 masl.



890 **Figure 10.** Observation (dotted lines) and model estimates (thin lines) of profiles on July 10, 2017 (left) and August 30, 2017 (right) from a transect up the Mackenzie River in the Northwest Territory of Canada. Dotted lines show observations and thin lines show model estimates corresponding to specific times during the transect.

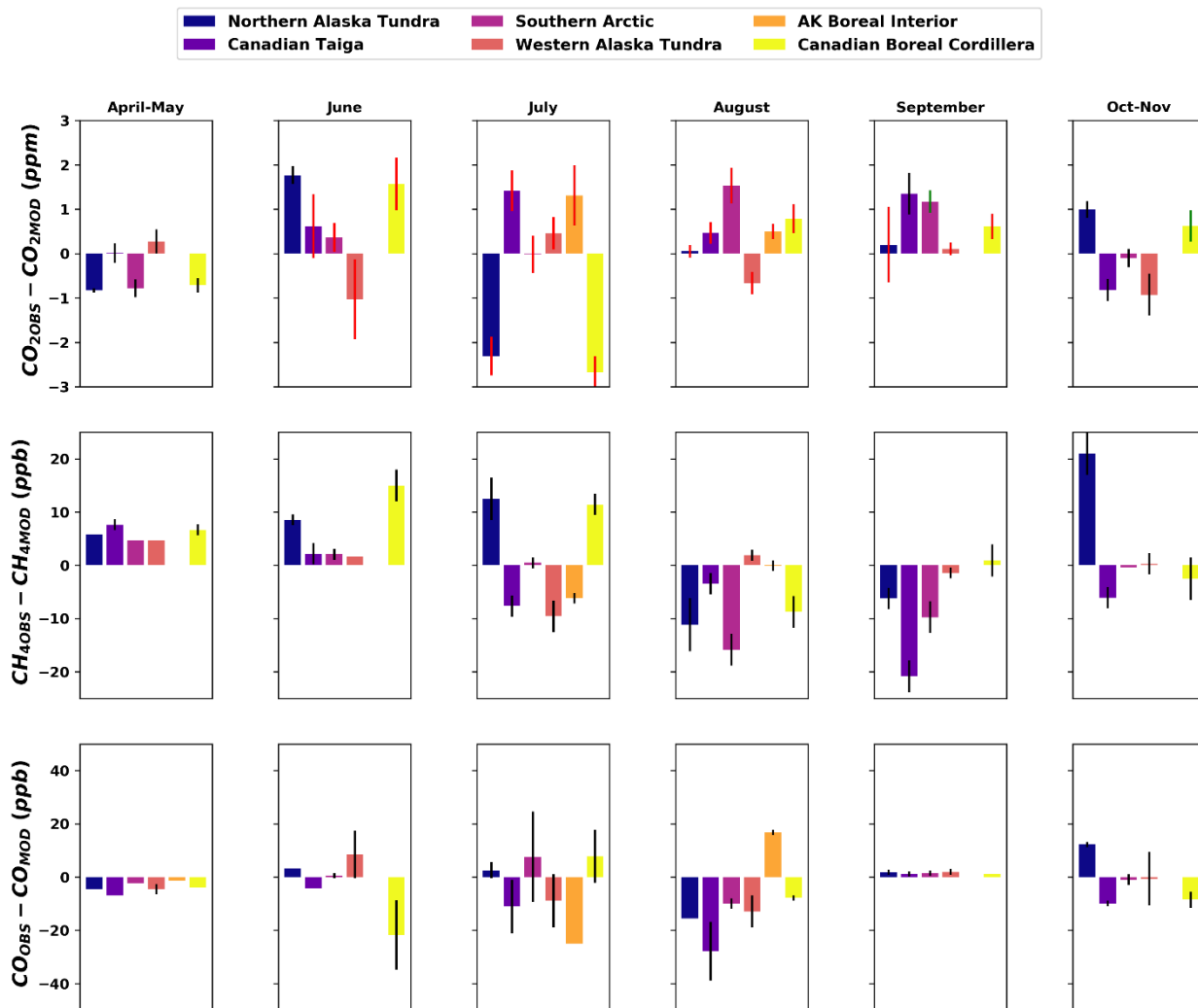


Figure 11. Average observation–model integrated enhancement differences by ecoregion. Standard deviation of differences for each region are shown with black and red bars. Red (black) bars signify a negative (positive) average enhancement below 3000 meters relative to the daily mean tropospheric value above 3000 masl for CO₂ and CH₄ and above 4000 masl for CO.

895

900