

Enhanced CO₂ uptake at a shallow Arctic Ocean seep field overwhelms the positive warming potential of emitted methane

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Continued warming of the Arctic Ocean in coming decades is projected to trigger the release of teragrams (1 Tg = 10⁶ tons) of methane from thawing subsea permafrost on shallow continental shelves and dissociation of methane hydrate on upper continental slopes. On the shallow shelves (<100 m water depth), methane released from the seafloor may reach the atmosphere and potentially amplify global warming. On the other hand, biological uptake of carbon dioxide (CO₂) has the potential to offset the positive warming potential of emitted methane, a process that has not received detailed consideration for these settings. Continuous sea–air gas flux data collected over a shallow ebullitive methane seep field on the Svalbard margin reveal atmospheric CO₂ uptake rates (−33,300 ± 7,900 μmol m^{−2}·d^{−1}) twice that of surrounding waters and ~1,900 times greater than the diffusive sea–air methane efflux (17.3 ± 4.8 μmol m^{−2}·d^{−1}). The negative radiative forcing expected from this CO₂ uptake is up to 231 times greater than the positive radiative forcing from the methane emissions. Surface water characteristics (e.g., high dissolved oxygen, high pH, and enrichment of ¹³C in CO₂) indicate that upwelling of cold, nutrient-rich water from near the seafloor accompanies methane emissions and stimulates CO₂ consumption by photosynthesizing phytoplankton. These findings challenge the widely held perception that areas characterized by shallow-water methane seeps and/or strongly elevated sea–air methane flux always increase the global atmospheric greenhouse gas burden.

methane | carbon dioxide | greenhouse gas emissions | marine biogeochemistry | Arctic Ocean

Contemporary methane emissions from continental shelves to overlying oceans are estimated at 8 Tg to 65 Tg (1 Tg = 10¹² g) annually (1), and will likely increase with future global warming. Inundation of high-latitude continental shelves accompanying sea level rise for the past ~20 ka and superposed, shorter-lived warming events may be the triggers for ongoing methane release from thawing subsea permafrost, dissociating gas hydrate, and/or recent accumulations of microbial methane in newly warmed, organic-rich sediment (2–4). In deeper continental slope waters (200 m to 600 m) at high latitudes and midlatitudes, widespread methane seepage has been observed where warming intermediate ocean waters impinge on the shallowest extent of the gas hydrate stability zone (3, 5–9).

Methane emitted from seafloor seeps may exacerbate global warming if this potent greenhouse gas crosses the sea–air interface. However, most methane released at the seafloor may never reach the uppermost oceanic mixed layer due to a combination of gas exchange during the ascent of bubbles (10, 11), methane dissolution (12), and microbial oxidation of dissolved methane to carbon dioxide (13–15) (CO₂). The exception is methane emitted from the seabed in shallow-water marine settings (<100 m water depth), of which a substantial fraction may enter the atmosphere (2, 16, 17). Studies at these shallow-water locations have taken on urgency given their wide geographic distribution and potential for large

emissions (18). On the other hand, little attention has been given to CO₂ exchange as a component of the net greenhouse gas flux balance at gas seepage sites. If CO₂ efflux is enhanced at seepage sites, it will have a positive effect on radiative forcing. On the other hand, if CO₂ is absorbed, it could mitigate the positive radiative effects from methane efflux.

In this study, we quantify methane and CO₂ sea–air fluxes along the western Svalbard margin (WSM) (Fig. 1A) to determine the net global warming potential for these two important greenhouse gases. The study was conducted in summer when light availability was unlimited, thereby maximizing the potential for CO₂ uptake by primary production (i.e., photosynthesis). The areas surveyed include a stable deep-water gas hydrate system (1,700 m to 2,600 m depth), a gas seepage area (240 m depth) suggested to be geologically linked to deeper climate-sensitive gas hydrate occurrences (5, 7, 19), a shallow-water (80 m to 90 m depth) active gas seep field along the ridge of a glacial moraine (19), and a nearshore (<110 m depth) coastal zone. These sites represent the full depth range of potential methane-emitting sites on high-latitude continental margins (3), including the potential for contributions from thawing subsea permafrost (4, 19).

Materials and Methods

In June 2014, we obtained a 1,600-km-long near-continuous record of methane and CO₂ concentrations and carbon isotopes from surface water of the WSM aboard the R/V *Helmer Hanssen* (Fig. 1A). These data, in addition to methane

Significance

Methane released from the seafloor and transported to the atmosphere has the potential to amplify global warming. At an arctic site characterized by high methane flux from the seafloor, we measured methane and carbon dioxide (CO₂) exchange across the sea–air interface. We found that CO₂ uptake in an area of elevated methane efflux was enhanced relative to surrounding waters, such that the negative radiative forcing effect (cooling) resulting from CO₂ uptake overwhelmed the positive radiative forcing effect (warming) supported by methane output. Our work suggests physical mechanisms (e.g., upwelling) that transport methane to the surface may also transport nutrient-enriched water that supports enhanced primary production and CO₂ drawdown. These areas of methane seepage may be net greenhouse gas sinks.

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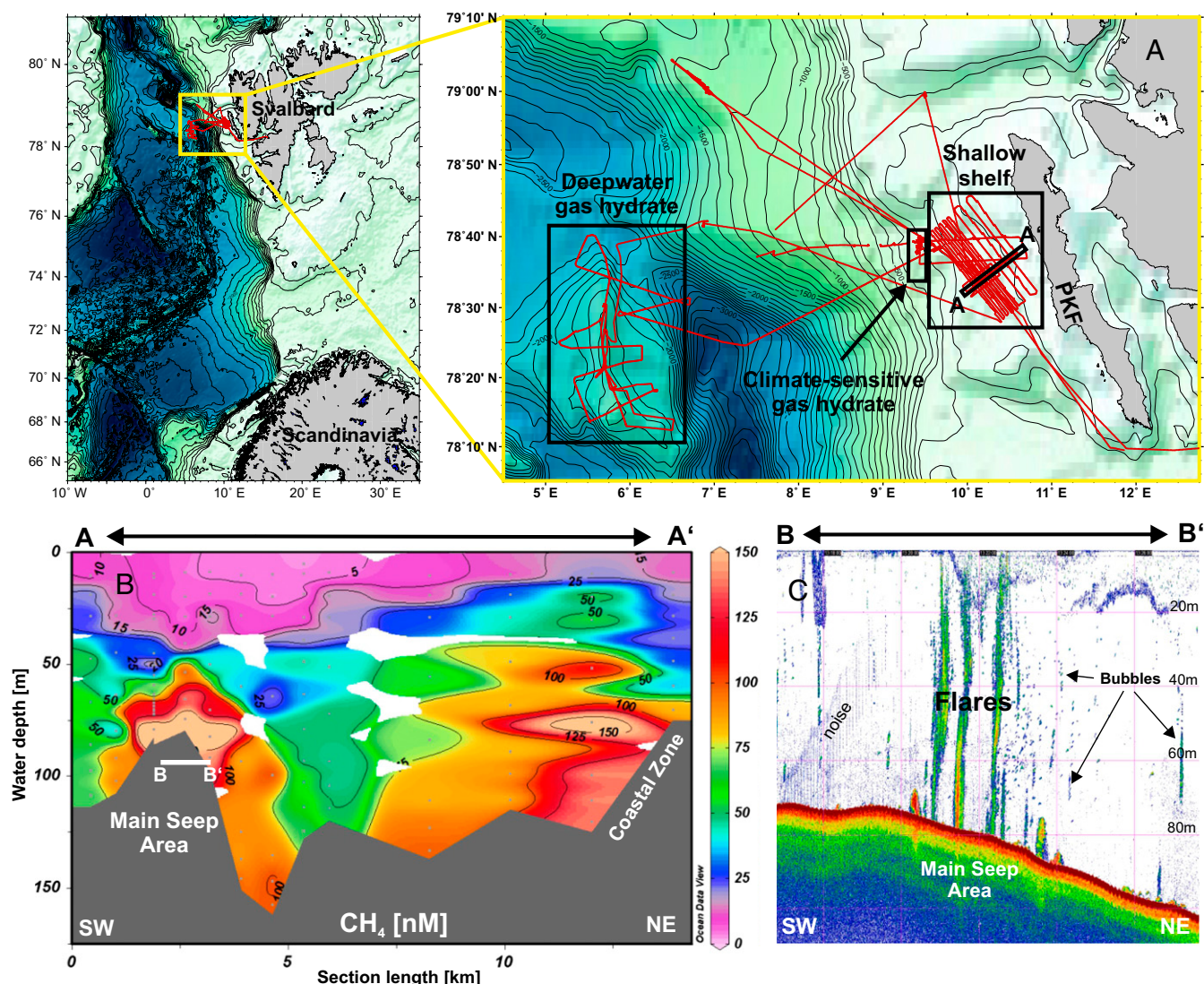


Fig. 1. Site map and water column methane offshore of western Svalbard during the CAGE 14-1 cruise. (A) Gas and gas hydrate sectors surveyed for methane flux, CO_2 flux, and water chemistry. Red lines are the survey tracklines. (B) Distribution of dissolved methane along transect A–A' (see A). (C) Hydroacoustic evidence for gas flares and bubbles overlying the main seep area along transect B–B' (see B). PKF, Prins Karls Forland.

and CO_2 concentrations from the atmospheric marine boundary layer, were acquired with the US Geological Survey-Gas Analysis System (USGS-GAS; *SI Appendix, Fig. S1*). The USGS-GAS is a dual cavity ring-down spectrometer (CRDS) analytical system that constantly circulates gas from the headspace of a Weiss-type equilibrator through a Picarro G-2201i CRDS and sequentially measures air concentrations received from intakes set at three to four different elevations on the ship exterior with a Picarro G-2301f CRDS. Stable carbon isotope values measured with the G-2201i were calibrated against standard gases to obtain accurate $\delta^{13}\text{C}$ values of surface water methane ($\pm 4\%$ at 2 ppm; $\pm 1.5\%$ at 5 ppm) and CO_2 ($\pm 1.5\%$). Gradients in methane and CO_2 concentrations were not detected in air samples collected at different elevations in the atmospheric marine boundary layer, so data from only one elevation (~ 22 m above the sea surface) are reported here. The gas concentration data were combined with meteorological (wind speed, air temperature) and sea surface water environmental parameters (salinity, water temperature; *SI Appendix, Fig. S2*) and averaged at 30-s intervals to determine the flux of methane and CO_2 across the sea–air interface (20) in shallow-water (Fig. 2) and deep-water (*SI Appendix, Fig. S3*) settings (21). To constrain biological activity in surface water, additional environmental parameters [dissolved oxygen (DO), pH, fluorescent dissolved organic matter (fDOM)] were measured in seawater pumped aboard the ship (*SI Appendix, Table S4*).

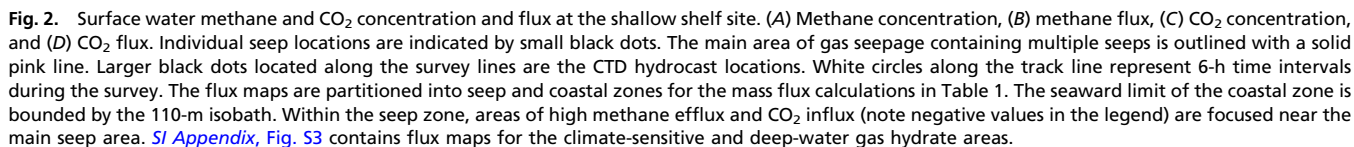
Surface water methane concentrations from 191 discrete water samples analyzed using the traditional gas chromatograph (GC) method and the

USGS-GAS instrumentation were positively correlated ($r^2 = 0.86$, $P < 0.001$) with slope of 0.99 (Fig. 3), which indicates excellent agreement between the analytical methods. The SD of the difference between the methods was 2.1 nM, with a small, but significant, 0.48 nM ($P < 0.001$) bias toward lower values measured by the USGS-GAS system (*SI Appendix, Fig. S7A*). To provide constraints on the subsurface methane distribution and its environmental controls, we also acquired dissolved methane concentration (Fig. 1B) and conductivity-temperature-depth (CTD) vertical profiles (Fig. 4).

Greenhouse Gas Dynamics

Methane Concentration and Fluxes on the WSM. For the deep-water and shelf-edge systems (Fig. 1A and *SI Appendix, Fig. S3*), surface water methane concentrations ranged from 3.2 nM to 4.3 nM, corresponding to saturation anomalies of -1.5 to 36% . Sea–air fluxes ranged from $0.0 \mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ to $2.8 \mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (Table 1 and *SI Appendix, Fig. S3*). The low saturation anomalies and fluxes for the 240-m water depth region are comparable to those in the open ocean (23) and are similar to those previously reported for this site (24), confirming that this setting is not a significant source of methane to the atmosphere.

The highest surface water methane concentrations (Fig. 2A) and dissolved-phase fluxes (Fig. 2B) were detected at the shallow



Gridded and normalized to an area of 100 km², the daily sea-air methane flux from each area ranged from 0.5 kg to 8.8 kg per 100 km², with highest values in the nearshore (*SI Appendix, Table S1*). In the context of a well-constrained global atmospheric methane source (e.g., ruminants), the flux from the shallow-water

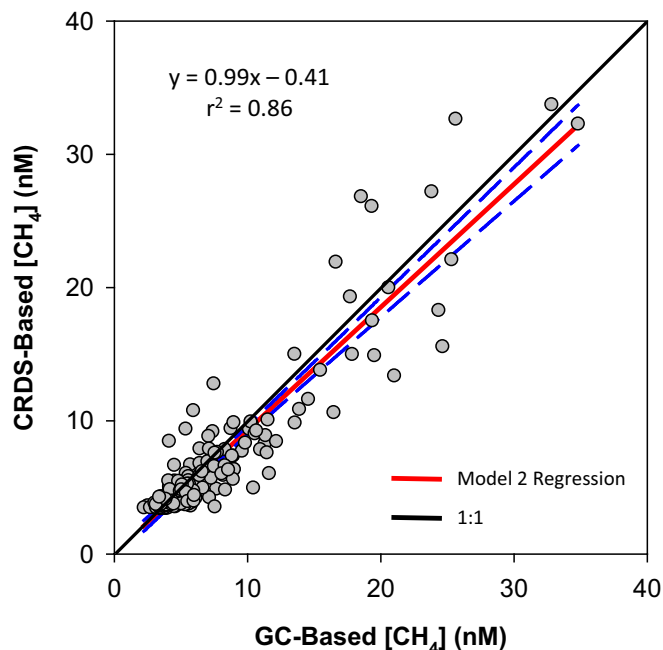


Fig. 3. Regression analysis for GC-based and CRDS-based measurements; 191 methane concentration measurements of discrete samples determined by the GC headspace analysis method compared with values obtained from the continuously measuring USGS-GAS. Blue dashed lines are the 95% confidence interval for the model 2 regression. There was a slight negative bias (-0.5 nM, $P < 0.001$) for the CRDS-based measurement, as determined by the Bland Altman agreement analysis (22) (*SI Appendix, Fig. S7*).

continental shelf seep field [$6.1 \text{ kg CH}_4 (100 \text{ km}^{-2})\cdot\text{d}^{-1}$, *SI Appendix, Table S1*] is equivalent to that from ~ 320 sheep, each emitting $18.9 \text{ g CH}_4 \text{ d}^{-1}$ (26). To match the methane output of the 3×10^7 sheep in New Zealand alone would require more than 90,000 multi-seep clusters of the type investigated here. Although tens of thousands of discrete seeps likely remain undiscovered on global margins (8), there is no evidence that such a large number of

multiseep clusters exists. Even if there were, the annual cumulative atmospheric methane flux would be $\sim 0.15 \text{ Tg CH}_4 \text{ y}^{-1}$, a negligible (0.03%) quantity relative to the 580 Tg of methane emitted to the atmosphere annually (18).

WSM Fluxes Compared with Siberian Shelf Seas. Our data show that shallow arctic methane seeps like those we investigated on the WSM emit negligible methane to the atmosphere. However, in comparison to the seeps we investigated, methane fluxes from the shallow East Siberian Arctic Shelf (ESAS), which may be underlain by thawing subsea permafrost, are ~ 100 times greater. Shakhova et al. (2) report average fluxes of $229 \mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ for “background” areas and $738 \mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ from “hotspots” during the ice-free summertime. These values are comparable to an average ice-free flux for the middle and outer East Siberian Arctic shelf of $238 \mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$, as measured by Thornton et al. (17). Given that (i) gas flares (evidence of seafloor gas ebullition) are prominent features on the WSM (ref. 15 and Fig. 1C) and ESAS (2), (ii) maximum bottom-water methane concentrations at the WSM seeps (Fig. 1B; ~ 300 nM) are comparable to summertime bottom-water concentrations in ESAS hotspots (2), and (iii) WSM gas could also have a component derived from thawing subsea permafrost (4, 19), it is difficult to reconcile why the diffusive fluxes we report from the WSM differ so greatly from those of the ESAS. Methane may be more rapidly oxidized from the WSM water column (13, 24); however, a more likely explanation is that lateral transport of methane from the relatively small and narrow WSM shelf dilutes and disperses methane into the deeper ocean. Similar dispersion and dilution on the shallow, expansive ESAS is not possible, which could permit a greater fraction of methane released from the seafloor to transfer to the atmosphere.

CO₂ Flux and Net Global Warming Potential Flux on the WSM. Within the shallow-water gas seep field, pCO₂ in the surface water was substantially less than in the surrounding area (Figs. 2 C and D and 5.4) and correlates negatively with methane concentration ($r^2 = 0.61$; *SI Appendix, Fig. S4*). These undersaturated pCO₂ values support a CO₂ influx rate of $-33,300 \pm 7,900 \mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ (Table 1), which is about twice that of the surrounding background area ($-16,000 \pm 6,000 \mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$) and more than 1,900 times greater than the efflux of methane ($17.3 \pm 4.8 \mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$). Taking into account the 25 times greater global warming potential

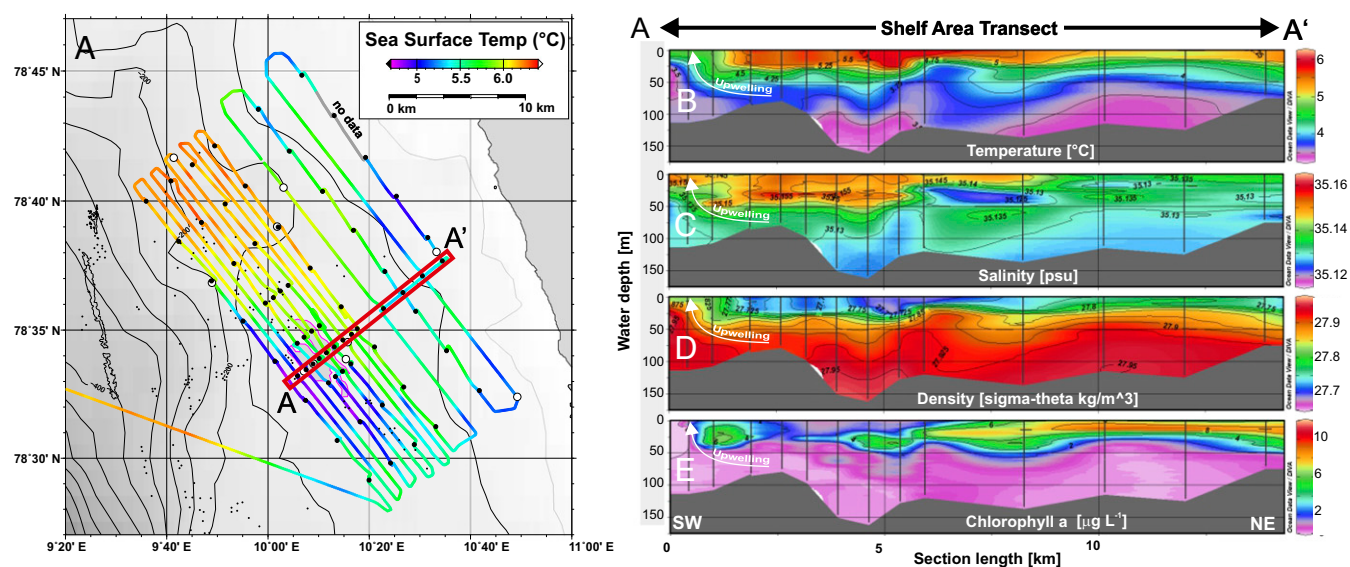


Fig. 4. Physicochemical water properties of the shallow shelf area. (A) Sea surface temperature (SST) during the shallow shelf survey. Colder surface water corresponds with the high-methane, low-CO₂ surface water in Fig. 2. Shore-perpendicular distributions of (B) temperature, (C) salinity, (D) density, and (E) chlorophyll concentration along transect A–A' show that colder bottom water upwells along steeply tilted isopycnals near the termination of a warm surface-water lens extending from the coastline. Elevated chlorophyll fluorescence was observed within the upwelling region.

Table 1. Methane and CO₂ fluxes for areas investigated in this study

Setting	Survey area (km ²)	CH ₄ flux ($\mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)	CO ₂ flux ($\mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$)
Shallow shelf seep field			
All	150.5	3.8 ± 5.5	$-18,037 \pm 8,464$
High flux*	17.6	17.3 ± 4.8	$-33,317 \pm 7,927$
Background	132.9	2.0 ± 1.9	$-16,017 \pm 6,152$
Nearshore coastal zone	38.7	5.5 ± 6.5	$-24,944 \pm 17,818$
Deep shelf seeps	11.5	0.30 ± 0.26	$-2,166 \pm 1,117$
Deep-water gas hydrate	112	1.05 ± 0.61	$-42,001 \pm 24,528$

See [SI Appendix, Table S1](#) for additional site summary details.

*High flux defined as areas with CH₄ flux > 10 μmol·m⁻²·d⁻¹.

of methane relative to CO_2 for a 100-y timescale on a per unit mass basis (18), the strongly negative CO_2 flux at the seep offsets the positive effect of methane expelled by a factor of 231 despite methane's greater global warming potential. Even on a 25-y timescale, for which methane has stronger GWP of 84 (18), the cooling effect of CO_2 uptake is 69 times greater than methane's warming effect. Our comparisons consider only the dissolved phase gas fluxes. However, hydroacoustic imaging (Fig. 1C) and bubble modeling (*SI Appendix, Fig. S8A*) suggest minimal direct bubble transport to the atmosphere. Furthermore, a recent study from the ESAS suggesting that turbulence-driven diffusive methane flux (not ebullition) is the primary transport mechanism for sea-air methane flux (17) supports our assessment that bubble transport of methane to the atmosphere is not important at this setting.

Stimulation of CO₂ Uptake over Shallow-Water Methane Seeps. At least two processes could be responsible for the reduced concentrations of CO₂ observed over the shallow-water methane seeps: (i) Methane bubbles ascending from the seafloor dissolve methane, strip CO₂ from the water column, and transport this CO₂ to the sea–air interface and release it to the atmosphere (12), or (ii) a physical and/or biological mechanism stimulates photosynthesis, and thus CO₂ drawdown, above the seep area. To test the first hypothesis, we applied a numerical bubble-stripping model (12). Reproducing the low CO₂ concentrations requires (i) bubble diameters of 14 mm, which is much larger than the most frequent diameter of ~6 mm (range 2 mm to 16 mm) observed in the area (27), and (ii) a volumetric gas flux of 34 L·m⁻²·min⁻¹ from the seabed at 90 m (~13.6 mol/min, at 4 °C), compared with reported values of 3 mL·min⁻¹ to 41 mL·min⁻¹ per seep at 385 m (5.4 mmol/min to 74.5 mmol/min, at 4 °C) (19). Bubble stripping is therefore not a plausible mechanism for removing CO₂.

The alternate hypothesis for lower surface-water $p\text{CO}_2$ is that upwelling of cold, nutrient-rich water stimulated CO_2 assimilation by phytoplankton, a phenomenon also observed in areas of strong upwelling associated with eastern boundary currents of major ocean basins (28). Surface water within the high-methane, low- CO_2 seep area was 0.65°C colder than the surrounding surface water (Figs. 44 and 5C and *SI Appendix, Table S1*), and the estimated $\delta^{13}\text{C}$ of the seabed-sourced methane measured at the sea surface (-54.6‰ ; *SI Appendix, Methane Isotopic Mass Balance for Determination of Seabed*) was similar to that reported at the seafloor (29) and emanating from seeps downslope (19). We are therefore confident the cold and methane-rich surface water originated from near the seafloor close to the seep area. Furthermore, CO_2 uptake rates we measured ($2,200\text{ }\mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ to $42,000\text{ }\mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$; Table 1) are comparable to primary production rates reported from nearby Kongsfjorden (30) ($600\text{ }\mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$ to $184,000\text{ }\mu\text{mol}\cdot\text{m}^{-2}\cdot\text{d}^{-1}$), confirming the plausibility that phytoplankton-related processes altered the surface water CO_2 budget. A possible subsurface manifestation of high surface productivity is that benthic chlorophyll and phaeopigment

concentrations at this seep were the highest among nine stations investigated in the western Svalbard–Barents Sea region (31).

Upwelling on the WSM shelf is driven by Ekman transport during northerly or onshore wind events that can occur during any season (32). On a smaller scale, the topographically steered Spitsbergen Polar Current encountering the high-relief glacial moraine may upwell locally along steeply tilted isopycnals (Fig. 4D). Bubble-driven buoyancy and entrainment of bottom waters may also transport bottom water to the photic zone from depths as great as 1,000 m (33), a mechanism invoked to explain elevated surface-water chlorophyll above a Gulf of Mexico hydrocarbon seep (34). The relatively low seafloor methane flux at the WSM seep sites between 240 m and 385 m water depth (19) renders it unlikely that bubble-associated buoyancy caused the upwelling, supporting the assumption that physical oceanographic processes alone are responsible for upwelling, independent of the presence of gas seepage.

Regardless of the upwelling mechanism, multiple lines of evidence support the interpretation that primary production and consequent CO₂ drawdown are enhanced where methane-charged bottom water emerges: (i) Chlorophyll-fluorescence, a proxy for photosynthesis, is elevated (Fig. 4E); (ii) DO, a product of photosynthesis, is ~1 mg/L higher in surface waters with high methane and low CO₂ concentrations (Fig. 5D); (iii) pH, which increases when CO₂ is removed from solution by photosynthesis, is elevated by as much as 0.6 units compared with background (Fig. 5D); and (iv) δ¹³C-CO₂, a metric that becomes more

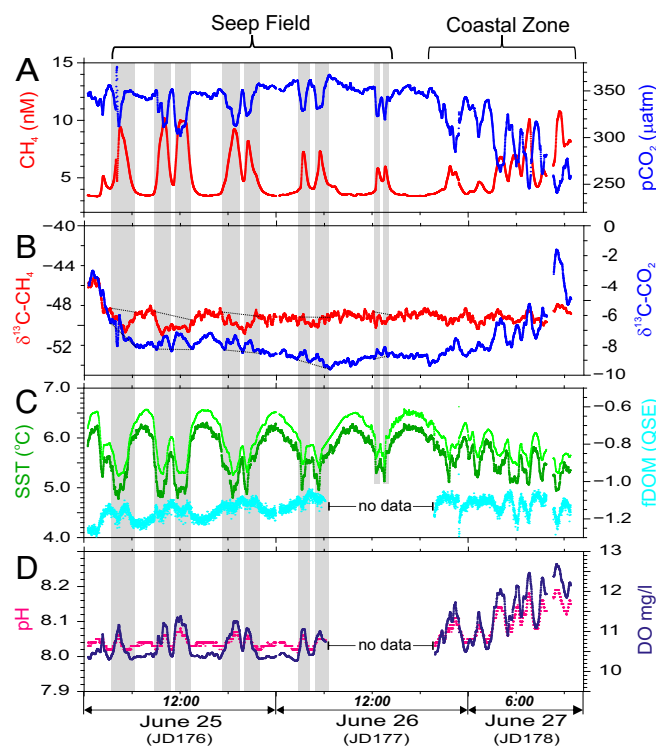


Fig. 5. Surface water time series data for the shallow shelf survey. (A) Dissolved methane concentration and $p\text{CO}_2$; (B) $\delta^{13}\text{C}\text{-CH}_4$ and $\delta^{13}\text{C}\text{-CO}_2$; (C) SST temperature [dark green, hull-mounted sensor; light green, EXO2 sensor (YSI Incorporated)] and fDOM; and (D) pH and DO. Seep crossings (highlighted with gray bars) are characterized by colder water containing elevated concentrations of ^{13}C -depleted methane and lower concentrations of ^{13}C -enriched CO_2 . Isotopic excursions are demarcated by dashed lines that connect $\delta^{13}\text{C}$ values from the margins of the seep crossings. Within the seep crossings, fDOM, pH, and DO are elevated. The combined evidence suggests upwelling of cold, methane-charged (and presumably nutrient-rich) bottom water originating from the seep-stimulated phytoplankton activity in the surface water that enhanced the consumption of CO_2 . Similar trends occur within the nearshore coastal zone.

positive when algae preferentially remove $^{12}\text{CO}_2$ during photosynthesis, is ^{13}C -enriched (more positive) by as much as 2‰ within the upwelling area of methane-charged bottom water (Fig. 5B).

Similar, yet more pronounced, patterns of high methane, low CO_2 , and changes in water chemistry indicative of upwelling-induced photosynthesis were observed in the nearshore coastal zone (Fig. 5 and *SI Appendix*, Fig. S4). However, the coastal zone lacks pervasive discrete bubble-releasing methane seeps (Fig. 2A). Most methane in that region (up to 150 nM in bottom waters; Fig. 1B) likely originates from in situ production in organic-rich, anoxic sediment. Elevated methane in marine surface waters can also be a product of dimethylsulfoniopropionate demethylation (35), but the high bottom-water methane content and $\delta^{13}\text{C}$ signature of the nearshore methane are most consistent with a sediment source.

Despite the spatiotemporal coincidence between high concentrations of methane and enhanced CO_2 uptake at seeps on the WSM continental shelf (Fig. 5A) and in some other settings such as the Santa Barbara Basin seep field (36), we suggest that high methane concentrations are an indicator of, but not a necessary condition for, enhanced CO_2 drawdown. Instead, the surface-water methane observed on the WSM is a chemical tracer for cold, nutrient-rich upwelled water that supports enhanced photosynthesis within the euphotic zone. A relationship of higher methane efflux and CO_2 influx that correlated with colder surface waters was also observed near the >2,000-m deep-water gas hydrate site (*SI Appendix*, Fig. S5). This observation suggests enhanced CO_2 drawdown is likely to occur whenever deep nutrient-rich (and perhaps methane-charged) waters are upwelled to the surface, and conditions for photosynthesis are suitable.

Methane seepage from high-latitude shallow continental margins is an atmospheric methane source (2) that could become more substantial as the climate continues to warm. Evidence that the cooling potential from CO_2 influx at this shallow-water arctic methane seep overwhelms the greenhouse warming potential from the emitted methane suggests that methane seeps can nevertheless be net sinks for climate-forcing gases. If the sedimentary efflux of nutrients that support photosynthesis is related to methane discharge intensity from the seafloor, a positive feedback between accelerated methane release from the seafloor and amplified atmospheric warming may be offset by atmospheric CO_2 drawdown. Further investigation of sea–air greenhouse gas fluxes at methane seep sites where upwelling-driven outputs are counteracted by photosynthetic CO_2 drawdown (including light-limited wintertime conditions) would provide data to constrain which processes are responsible for enhanced CO_2 uptake, quantify net greenhouse gas fluxes globally for shallow-water methane seepage areas, and determine if accelerated seafloor methane release will be offset by enhanced CO_2 uptake at the sea–air interface in the future.

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