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Pan-Arctic land–atmospheric fluxes of methane and carbon dioxide in response to climate change over the 21st century

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Abstract

Future changes of pan-Arctic land-atmospheric methane (CH₄) and carbon dioxide (CO₂) depend on how terrestrial ecosystems respond to warming climate. Here, we used a coupled hydrology-biogeochemistry model to make our estimates of these carbon exchanges with two contrasting climate change scenarios (no-policy versus policy) over the 21st century, by considering (1) a detailed water table dynamics and (2) a permafrost-thawing effect. Our simulations indicate that, under present climate conditions, pan-Arctic terrestrial ecosystems act as a net greenhouse gas (GHG) sink of -0.2 Pg CO_2 -eq. yr⁻¹, as a result of a CH₄ source $(53 \text{ Tg CH}_4 \text{ yr}^{-1})$ and a CO₂ sink (-0.4 Pg C yr⁻¹). In response to warming climate, both CH₄ emissions and CO₂ uptakes are projected to increase over the century, but the increasing rates largely depend on the climate change scenario. Under the non-policy scenario, the CH₄ source and CO_2 sink are projected to increase by 60% and 75% by 2100, respectively, while the GHG sink does not show a significant trend. Thawing permafrost has a small effect on GHG sink under the policy scenario; however, under the no-policy scenario, about two thirds of the accumulated GHG sink over the 21st century has been offset by the carbon losses as CH₄ and CO₂ from thawing permafrost. Over the century, nearly all CO₂-induced GHG sink through photosynthesis has been undone by CH₄-induced GHG source. This study indicates that increasing active layer depth significantly affects soil carbon decomposition in response to future climate change. The methane emissions considering more detailed water table dynamics continuously play an important role in affecting regional radiative forcing in the pan-Arctic.

Keywords: methane, carbon dioxide, greenhouse gases, climate change, Arctic, terrestrial ecosystem model, variable infiltration capacity

S Online supplementary data available from stacks.iop.org/ERL/8/045003/mmedia



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1. Introduction

Future changes in carbon stocks of pan-Arctic terrestrial ecosystems in response to climate change are mainly determined by the increases in net primary productivity (NPP), during warmer and longer growing seasons (Euskirchen et al 2006), and in heterotrophic respiration (R_h) , in warmer and deeper soil active layers (Hollesen et al 2011). The soil organic carbon (SOC) of terrestrial ecosystems in pan-Arctic permafrost region ($\sim 16\%$ of the global soil area) is estimated to be about 50% of the global SOC (Tarnocai et al 2009). The disproportionally large SOC has been accumulated historically due to slow decomposition rate with low temperature, but warmer and wetter future climate may accelerate soil biogeochemical cycling, leading to loss of carbon (Webster et al 2003, ACIA 2004, Davidson and Janssens 2006). Warming and thawing permafrost results in thicker soil active layers, which could lead to both enhanced decomposition and plant growth (Mack et al 2004), and previously frozen soil carbon is being converted and released as CH₄ and CO₂ (Harden et al 2012), which, as two major greenhouse gases (GHG), may significantly feedback to climate given the large size of SOC and warming climate in the pan-Arctic (Schuur et al 2008).

The uncertainty in boreal carbon budgets and their responses to climate change over the 21st century are among the largest remaining gaps in assessing regional carbon dynamics in pan-Arctic terrestrial ecosystems (e.g., McGuire et al 2009, Koven et al 2011). To date, the amounts and rates of land-atmospheric CH₄ and CO₂ fluxes are poorly constrained, as is the ratio of CH₄ to CO₂ (e.g., Whiting and Chanton 2001). The ratio, as an index of land-atmospheric GHG carbon exchange (Gorham 1991), is important in terms of carbon-climate feedbacks given higher radiative efficiency of CH₄. The relative amount of CH₄ and CO₂ released from soil carbon mineralization depends on the amount of available oxygen or dry/wet conditions in the soil (Segers 1998). More future precipitation and thermokarst may result in wetter soil conditions (Walter et al 2006), which favor CH4 production, while water redistribution towards thermokarst depressions may lead to drier soils in adjacent higher areas (Osterkamp *et al* 2009), which favor CO_2 production. Thus, how the quantity and quality (i.e., the ratio of CH_4 to CO_2) of regional carbon budgets will change over the 21st century depends on how pan-Arctic terrestrial ecosystems respond to potentially dramatic environmental changes, including rising temperature, change of precipitation regimes, and resultant changes in soil thermal and hydrological conditions (Romanovsky et al 2007, Schulze et al 2001, Zhuang et al 2006, Solomon et al 2007).

To evaluate regional carbon budgets of CH₄ and CO₂ and their respective impacts on climate radiative forcing, the net methane exchanges (NME) and net ecosystem exchange (NEE) need to be quantified. To date, most regional carbon dynamics studies focus on either NME (e.g., Walter *et al* 2001, Melton *et al* 2013) or NEE (e.g., Clein *et al* 2002, Carrasco *et al* 2006). Only a few process-based models were developed to simulate both NME and NEE at region scales, but they assumed uniform water table depth (WTD) over large grid cells (e.g., 0.5°) without considering the sub-grid spatial heterogeneity of water table dynamics (e.g., Zhuang *et al* 2006), which is important in the quantification of regional carbon budgets (Bohn and Lettenmaier 2010). In this study, we used a coupled hydrology–biogeochemistry model framework, incorporating sub-grid spatial variation in the WTD, to estimate land–atmospheric NME and NEE over the pan-Arctic during the 21st century. We have three main objectives: (1) to explore how NME and NEE respond to future climate change, (2) to analyze the relative contribution of CH₄ and CO₂ to regional carbon budgets and radiative forcing, (3) to examine the effects of permafrost thawing on projected regional carbon dynamics.

2. Methods

2.1. Model and data

A macro-scale hydrological model (variable infiltration capacity, VIC Liang et al 1994, Cherkauer et al 2003) and a biogeochemical model (terrestrial ecosystem model, TEM Raich *et al* 1991, Zhuang *et al* 2004) were coupled (figure 1) to make estimates of land-atmospheric NME and NEE over the pan-Arctic, which is defined as the watersheds of major rivers that drain into the Arctic Ocean (Lammers et al 2001). This coupled VIC-TEM model framework was developed and applied to estimate NME (Lu and Zhuang 2012). Here, we extended the model framework to estimate both NME and NEE. The VIC model simulates soil thermal and hydrological dynamics using physically based formulations (Liang et al 1994), and has been applied in cold region hydrology (Su et al 2005, 2006) by the incorporation of a frozen soil/permafrost algorithm (Cherkauer et al 2003). The TEM model explicitly simulates carbon and nitrogen dynamics of vegetation and soils, and has been used to examine terrestrial CH₄ and CO₂ dynamics in high latitudes (Zhuang et al 2004, 2006, McGuire et al 2010). TEM has its hydrological module (Zhuang et al 2002) to simulate water dynamics of terrestrial ecosystems. However, like many existing biogeochemistry models, the hydrological module is formulated as a simple 'single-bucket'. To improve the estimates of soil moisture profile, a key controlling factor for biogeochemical processes of CH₄ and CO₂, more sophisticated hydrological models, like VIC, are needed (e.g., Bohn and Lettenmaier 2010, Lu and Zhuang 2012). In TEM, the NME is calculated as the difference between CH₄ production (methanogenesis) and CH₄ oxidation (methanotrophy). The NEE is calculated as the difference between CO₂ uptakes, through plant photosynthesis, and CO₂ emissions, from $R_{\rm h}$ and accompanied releases of CO₂ associated with methanogenesis and methanotrophy (Conrad et al 1989, Tang et al 2010). The size of SOC pool in TEM is determined by the balance between litter production (proportional to vegetation carbon pool) and $R_{\rm h}$. To consider the effect of thawing permafrost on carbon dynamics, a permafrost-switch was set in this study to control the newly available SOC due to permafrost thawing. When the permafrost-switch was on, the newly available SOC was simply added into SOC pool of the TEM once in a year (at the end of each year). When the switch was off, this effect was deactivated. The magnitude of newly available SOC was calculated based on the annual change of simulated active layer depth (ALD) weighted by spatially



Figure 1. Conceptual framework of coupled models to estimate net fluxes of methane and carbon dioxide. Shown are external spatial inputs (blue) for driving or calibrating models, internal information exchange (yellow) between models, and final model outputs (green). Arrows indicate the direction of information exchange among all components. See text and supplementary materials for details (available at stacks.jop.org/ERL/8/045003/mmedia).

explicit SOC profiles derived from Harden *et al* (2012) (see the supplementary materials for details, available at stacks. iop.org/ERL/8/045003/mmedia).

This study used two contrast climate change scenarios simulated by the MIT's integrated global systems model (IGSM) (Reilly et al 1999, Prinn et al 1999, Webster et al 2003, Sokolov et al 2009) with a median transient climate response (Webster et al 2012): a no-policy scenario (a reference case assuming no explicit climate policy) and a policy scenario (a stabilization case assuming a specified climate policy). The projected atmospheric CO₂ mole fraction (parts per million, ppm) in 2100 is about 900 ppm and 480 ppm for no-policy and policy scenarios, respectively. Air temperature over the region is projected to increase by 8 °C and 2°C, and annual precipitation is projected to increase by 200 mm and 70 mm, respectively. Further details on climate change scenarios and all other spatially explicit inputs (figure 1) for the VIC-TEM model framework are provided in the supplementary materials.

2.2. Simulation protocol

In the VIC–TEM model framework, the VIC model was first run to solve soil moisture and energy balance and provide soil freeze/thaw fronts, soil temperature, ice and moisture profiles. All these VIC outputs were fed directly into TEM model except soil moisture profile, which was used to estimate WTD using the soil moisture deficit method (Bohn *et al* 2007) and then fed into the TEM model after redistributing WTD based on a TOPMODEL parameterization scheme (see the supplementary materials). To examine how NME and NEE respond to future climate change, we conducted two model CO₂ concentrations and climate forcing from each of the two climate change scenarios: no-policy and policy scenarios. The permafrost-thawing effect was considered in the simulations by turning on the permafrost-switch. The spatio-temporal dynamics of NME and NEE over the pan-Arctic and their relative impacts on climate system were analyzed based on these two model simulations: no-policy and policy simulation. The warming/cooling impacts of NME and NEE on climate system were compared using the global warming potentials (GWP) (denoted as CO₂-eq.), assuming one gram of CH₄ is equivalent to 25 g of CO₂ over a 100-year time horizon (Solomon et al 2007). In addition, in order to separate the effects of permafrost thawing from others, we conducted another two model simulations using the same two scenarios but with the permafrost-switch turned off. We calculated the effect of permafrost thawing as the difference between the two sets of simulations with and without considering the permafrost-thawing effect.

simulations over the 21st century using future atmospheric

3. Results and discussion

3.1. Spatial patterns of CH_4 and CO_2 fluxes

Both NME and NEE estimates showed substantial spatial variations over the region, with consistent spatial patterns between no-policy (figures 2(a)-(d)) and policy (figures S1(a)-(d), available at stacks.iop.org/ERL/8/045003/mmedia) simulations. As indicated by the mean annual estimates of the 2000s and 2090s, the spatial patterns of both NME and NEE seemed not to change over time for each simulation. The highest positive NME, or CH₄ source, occurred in the



Figure 2. Mean annual net methane exchange (NME) (top row), net ecosystem exchange (NEE) (middle row), and net global warming potential (GWP) (bottom row) over the pan-Arctic during the 2000s (left column) and the 2090s (right column), under the no-policy scenario.

West Siberian Lowlands and the Hudson Bay Lowlands, with CH₄ sources up to 8 g CH₄ m⁻² yr⁻¹ during the 2000s. The negative NME, or CH₄ sink, existed over the southern parts of the study region, with CH₄ sinks up to -0.2 g CH₄ m⁻² yr⁻¹ during the 2000s.

The magnitude and spatial variations of NME were generally consistent with previous regional CH₄ studies focusing on northern high latitudes (e.g., Zhuang *et al* 2004, Petrescu *et al* 2010, Zhu *et al* 2011). In terms of the similarity in spatial patterns, the CH₄ sinks seemed to



Figure 3. Comparison of the observed and simulated active layer depths (b), CH_4 fluxes (c), and CO_2 fluxes (d) at field sites over the pan-Arctic (a). The observed active layer depths are multi-year mean values of annual end-of-season thaw depth from 155 circumpolar active layer monitoring (CALM) sites (Brown *et al* 2000). The observed net fluxes of CH_4 and CO_2 are mean daily fluxes during the growing season (May–Sep.), and the information of 22 CH_4 flux sites and 14 CO_2 flux sites can be found in tables S1 and S2 (available at stacks.jop.org/ERL/8/045003/mmedia), respectively. Solid and dashed lines represent fitted and 1:1 lines, respectively.

be dominated by WTD (figure S2, available at stacks.iop. org/ERL/8/045003/mmedia), while the CH₄ sources seemed to be dominated by soil temperature (figure S3, available at stacks.iop.org/ERL/8/045003/mmedia). For NEE, most of the study region had a negative NEE, or CO2 sink, with a sink of atmospheric CO₂ up to -80 g C m⁻² yr⁻¹ during the 2000s. The West Siberia acted as a strong CO₂ sink, while other regions were weak sinks or small sources. The magnitude of NEE was comparable to other model simulations of northern terrestrial ecosystems (e.g., Wania et al 2009, Koven et al 2011). The spatial pattern of NEE was partly consistent with WTD (figure S2), since we only considered the heterotrophic respiration in unsaturated soils above WTD given that decomposition rates were very low under anaerobic conditions in saturated soils (Freeman et al 2001).

We evaluated our simulations with observations at field sites over the pan-Arctic (figure 3(a)). The comparison between the simulated and observed ALD at 155 circumpolar active layer monitoring (CALM) sites showed that our simulations underestimated ALD, especially for those soils with thick active layers (figure 3(b)). And the comparison between the simulated and observed mean daily CH₄ fluxes over the growing season (May-September) at 22 CH₄ flux sites (table S1, available at stacks.iop.org/ERL/8/045003/ mmedia) indicated that our simulations were able to well simulate the fluxes of CH_4 (figure 3(c)). However, our simulations overestimated the net fluxes of CO_2 (figure 3(d)), based on the comparison of the simulated and observed mean daily CO₂ fluxes (negative values indicate carbon sinks) over the growing season at 14 eddy covariance flux sites (table S2, available at stacks.iop.org/ERL/8/045003/mmedia).



Figure 4. Annual net methane exchange (NME) (a), net ecosystem exchange (NEE) (b), global warming potential (GWP) (c), and the ratio of CH_4 - and CO_2 -induced GWP (d) over the pan-Arctic for the 21st century, under the no-policy (dashed lines) and policy (solid lines) scenarios. Gray lines indicate the simulations without considering the permafrost-thawing effect. Absolute values of GWP are used for the calculation of the ratios. A 5-year running mean is applied to all the curves.

For WTD, we compared our estimates (figure S2) with the simulations of global equilibrium WTD (Fan *et al* 2013) and found that the spatial pattern and magnitude of our estimates of WTD over the pan-Arctic was comparable with their results. The regions with shallow WTD well corresponded to major wetland regions over the pan-Arctic, including West Siberian Lowlands and Hudson Bay Lowlands.

3.2. Inter-annual variations of regional CH₄ and CO₂

The annual regional total NME and NEE over the pan-Arctic exhibited a significant inter-annual variability over this century, and the difference in the estimates between no-policy and policy simulations increased over time, especially from the middle of this century (figures 4(a) and (b)). During the 2000s, the regional CH₄ source and CO₂ sink were estimated to be 53 Tg CH₄ yr⁻¹ and -0.4 Pg C yr⁻¹, respectively. For the no-policy simulation, the magnitudes of CH₄ source and CO₂ sink were projected to be 85 Tg CH₄ yr⁻¹ and -0.7 Pg C yr⁻¹ during the 2090s, with annual changing rates of 0.38 Tg CH₄ yr⁻² and -2.8 Tg C yr⁻², respectively (table 1). However, for the policy simulation, the annual

changing rates of regional CH_4 sources and CO_2 sinks were much smaller, about a quarter of the changing rates of the no-policy simulation (table 1). The big differences in the estimated NME and NEE resulted from the differential response of terrestrial ecosystems to various climate forcing and atmospheric CO_2 concentrations.

Our estimates of regional CO2 sink under present climate conditions $(-0.4 \text{ Pg C yr}^{-1})$ was comparable to net carbon flux of this region based on previous model studies (-0.8-0 Pg C yr⁻¹; McGuire *et al* 2009, Koven *et al* 2011, Schaphoff et al 2013), and contemporary regional CH₄ source (53 Tg CH₄ yr⁻¹) was within the range of previous estimates (20–157 Tg CH₄ yr⁻¹; Christensen *et al* 1996, Zhuang et al 2004, Petrescu et al 2010). For the no-policy simulation, the 60% increase in the projected CH₄ source by 2100 was comparable to the projected increase of future CH₄ emissions (51%) in northern Eurasia using similar climate change scenarios (Zhu et al 2011); and the projected regional CO_2 sink by the 2090s (-0.7 Pg C yr⁻¹, a 75% increase over the 1990s level) was at the high end of the multi-model estimates of CO₂ sink (-0.3 ± 0.3 Pg C yr⁻¹; Qian *et al* 2010). Our simulations showed a consistent increase in regional

Table 1. Annual changing rate of key variables over the 21st century, under the no-policy and policy scenarios. The annual changing rates are determined as the slopes from a least square linear regression over the century, over the pan-Arctic. All estimates except those for GWP are statistically significant at P < 0.01.

	No-policy			Policy		
	Estimate	95% CI	R^2	Estimate	95% CI	R^2
Soil temperature ^a (°C yr ⁻¹)	0.042	(0.040, 0.043)	0.96	0.011	(0.010, 0.012)	0.91
Active layer depth (cm yr^{-1})	0.538	(0.512, 0.564)	0.95	0.136	(0.118, 0.154)	0.70
Water table depth ^b (cm yr ^{-1})	-0.107	(-0.112, -0.101)	0.94	-0.036	(-0.041, -0.031)	0.69
NME (Tg CH ₄ yr ^{-2})	0.375	(0.361, 0.404)	0.91	0.113	(0.107, 0.119)	0.62
NPP (Tg C yr^{-2})	-10.208	(-10.723, -9.754)	0.91	-2.542	(-2.654, -2.423)	0.70
$R_{\rm h}$ (Tg C yr ⁻²)	8.154	(7.747, 8.558)	0.89	2.029	(1.928, 2.130)	0.76
NEE (Tg C yr ^{-2})	-2.771	(-2.911, -2.632)	0.90	-0.607	(-0.637, -0.576)	0.25
GWP (Tg CO_2 -eq. yr ⁻²)	-0.779	(-0.827, -0.741)	0.05	0.543	(0.516, 0.571)	0.03

^a Top-layer (0–5 cm) soil temperature.

^b Mean water table depth during the growing season (May–Sep.).

net CO_2 sink, suggesting that NPP will increase faster than soil decomposition in response to future climate warming over the century. This is not consistent with previous model studies (e.g., Koven *et al* 2011, Schaphoff *et al* 2013), which projected that, during the second half of this century, regional net CO_2 sink will weaken and gradually turn into a source. The differential changing patterns of regional net CO_2 sink in response to future climate change could be caused by different model assumptions and climate change scenarios. For example, the TEM used in our projections explicitly took into account the nitrogen limitation on plant. The inclusion of carbon–nitrogen interaction in the model projections might have greatly influenced how vegetation and soil carbon respond to future climate in the pan-Arctic.

3.3. Regional global warming potentials of CH₄ and CO₂

The spatial pattern of net GWP was a combination of the spatial variations of NME and NEE for both no-policy and policy simulations (figure 2 and figure S1 available at stacks. iop.org/ERL/8/045003/mmedia). Those regions with high CH₄ emissions tended to be a net GHG source (up to 100 g CO_2 -eq. m⁻² yr⁻¹), while other regions were generally a net GHG sink (up to -150 g CO₂-eq. m⁻² yr⁻¹) (figures 2(e) and (f)). The regional net GHG sink, the result of positive CH₄-GWP (GHG source) and negative CO₂-GWP (GHG sink), was estimated to be -0.2 Pg CO_2 -eq. yr⁻¹ during the 2000s, and the annual GHG did not have a significant trend during the 21st century although the magnitude fluctuated over the century (figure 4(c)). Our estimate of the ratio of CH₄ emissions to CO₂ uptakes during the 2000s, $\sim 10\%$ on a molar basis, was within the range of field observations (3-20% Whiting and Chanton 1993, 2001). Although CH₄ source was relatively small in comparison with the CO₂ sink, the CH₄-GWP offset \sim 90% of CO₂-GWP (figure 4(d)), which highlighted the importance of CH₄ in regional total radiative forcing. Over the 21st century, there was no significant trend for the ratio of CH₄-GWP to CO₂-GWP for both no-policy and policy simulations.

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3.4. Effects of permafrost thawing on regional carbon budgets

Under future warming climate conditions, the thawing permafrost could lead to both enhanced decomposition and plant growth. The sign and magnitude of net carbon balance depends on the relative intensity of stimulation on soil decomposition and plant growth. In this study, two sets of model simulations, with the permafrost-switch on/off, were conducted, and the effect of permafrost thawing on regional carbon budgets was calculated as the difference between these two sets of simulations. Our results indicated that, by excluding the permafrost-thawing effect, our no-policy simulations showed a 8% decrease and a 14% increase in the absolute magnitude of regional CH_4 source (figure 4(a)) and CO_2 sink (figure 4(b)) over the 21st century, respectively, which implied that soil decomposition will be more intensified due to thawing permafrost, resulting in a net decrease in ecosystem carbon storage. The net losses of CH₄ and CO₂ associated with permafrost thawing were estimated to about 0.6 Pg CH₄ and 7.5 Pg C over the century. Taken together, the net accumulated GHG sink were reduced by 41.5 Pg CO₂-eq. over the century, which was almost twice as the accumulated GHG sink $(-24.1 \text{ Pg CO}_2\text{-eq.})$ simulated with a permafrost-thawing effect (figure 4(c)). Thus, the comparison between the no-policy simulations with and without the permafrost-thawing effect suggested that the thawing permafrost resulted in a 63% decrease in accumulated GHG sink over the 21st century. Compared with the no-policy simulation, there were small differences in the estimated carbon and GHG budgets between policy simulations with and without a permafrost-thawing effect.

Over the century, the simulated mean ALD, over the regions where permafrost still exists by 2100 (within the top 3 m), will change from ~1 m during the 2000s to ~1.5 m during the 2090s, and the annual changing rate of ALD (~0.5 cm yr⁻¹) was comparable with the observed changes during recent two decades at CALM sites (Brown *et al* 2000). However, the underestimation of the simulated ALD (figure 3(b)) suggested that actual net carbon loss

due to permafrost thawing might be larger than what we have estimated (8.0 Pg C over the century). Our estimation of net carbon loss associated with permafrost thawing was lower than our previous estimates (Zhuang *et al* 2006), but our estimates did not consider other disturbances in this region (e.g., fires), which could also lead to higher carbon losses.

In addition, our calculations of net carbon loss due to permafrost thawing could be greatly influenced by the specification of the vertical distribution of SOC. To assess the influence of the choice of SOC profile data, we conducted one additional no-policy simulation using a biome-dependent SOC profile dataset (Jobbágy and Jackson 2000), which includes a global summary of vertical SOC distribution of each biome (SOC profiles for two major biomes in the pan-Arctic, boreal forest and tundra, were shown in figure S5, available at stacks.iop.org/ERL/8/045003/mmedia). Compared to the previous no-policy simulations with SOC profile data derived from Harden et al (2012), this additional no-policy simulation, during the 2000s, showed a 12% decrease in CH₄ source (from 53 to 47 Tg CH₄ yr⁻¹) and a 20% increase in CO₂ sink (from -0.4 to -0.5 Pg C yr⁻¹), which together tripled the GHG sink (from -0.2 to -0.6 Pg CO_2 -eq. yr⁻¹). However, in spite of the difference in the magnitude of carbon/GHG budgets, the changing patterns of NME, NEE and GWP from this additional no-policy simulation did not change in comparison with the previous simulations.

4. Conclusions

Using a coupled hydrology-biogeochemistry model framework, the net carbon dynamics (CH₄ and CO₂) and their combined GHG over the pan-Arctic were analyzed over the 21st century under two contrast climate change scenarios. Under present climate conditions, the region acted as a CH₄ source (53 Tg CH₄ yr⁻¹) and a CO₂ sink (-0.4 Pg C yr⁻¹), resulting in a net GHG sink of -0.2 Pg CO₂-eq. yr⁻¹. In response to future warming climate, both the CH₄ source and the CO₂ sink strengthened, but the net GHG sink did not show a significant trend. The increasing rates mainly depended on the climate change scenarios. Under the no-policy climate change scenario, the CH₄ source and the CO₂ sink were projected to increase 60% and 75% by 2100, respectively. The consistent increase in the CO₂ sink in our simulations suggested that NPP will increase faster than R_h in response to future climate warming over the 21st century. Although the amount of CH₄ emissions was small ($\sim 10\%$ of the amount of carbon uptakes on a molar basis), they played an important role in affecting regional radiative forcing since CH₄-GWP had offset almost all CO₂-GWP over the century. Our simulations suggested that the thawing permafrost will enhance both CH₄ and CO₂ emissions, and the magnitude of future GHG sink will highly depend on whether a permafrost-thawing effect is included or not. The inclusion of a permafrost-thawing effect led to a 63% loss of GHG sink over the century.

The coupled hydrology-biogeochemistry model framework was operated at a fine spatial resolution, representing the critical effects of hydrological dynamics on GHG cycling of terrestrial ecosystems. The consideration of soil carbon distribution and thawing permafrost in quantification was another improvement to our previous quantification (e.g., Zhuang et al 2006). However uncertainties in the projection of GHG source/sink remained large due to two main sources. One the one hand, uncertainties came from the projection of future climate change, which itself was greatly affected by the climate policy implemented in the model and climate model parameterizations (e.g., climate sensitivity). In this study, we only considered the difference in climate policymaking (a reference case and a stabilization case), and a same climate model parameterization (median transient climate response) was applied in both cases (Webster et al 2012). One the other hand, large uncertainties could result from incomplete understanding of biogeochemical and physical processes in terrestrial ecosystems and the capacity of process-based models to represent underlying mechanisms at different temporal and spatial scales. Also, the choice of SOC profile data did influence our estimates especially for GWP, although it did not affect the changing patterns of carbon/GHG budgets over the century.

Although the CO₂-fertilization, carbon-nitrogen interaction, and thawing permafrost were considered in the model to examine how CH₄ and CO₂ respond to climate change over the century, several other mechanisms, including fire disturbances (Carrasco et al 2006), changes in biome composition (Wilmking et al 2004), and cryoturbation in permafrost-affected soils (Bockheim 2007), can affect the quantification of pan-Arctic carbon balance and need to be included in future work. In particular, the CO₂ emissions from forest fires (~ 0.2 Pg C yr⁻¹; Zhuang *et al* 2006) might have offset a half of net CO2 sinks under present climate conditions. In addition, vegetation shifts (e.g., expansion of boreal forest Euskirchen et al 2009) and small-scale permafrost-hydrology interactions (e.g., thermokarst erosion van Huissteden et al 2011) are also needed to be considered in future assessment of carbon dynamics in the pan-Arctic.

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