

Strong atmospheric chemistry feedback to climate warming from Arctic methane emissions

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[1] The magnitude and feedbacks of future methane release from the Arctic region are unknown. Despite limited documentation of potential future releases associated with thawing permafrost and degassing methane hydrates, the large potential for future methane releases calls for improved understanding of the interaction of a changing climate with processes in the Arctic and chemical feedbacks in the atmosphere. Here we apply a “state of the art” atmospheric chemistry transport model to show that large emissions of CH₄ would likely have an unexpectedly large impact on the chemical composition of the atmosphere and on radiative forcing (RF). The indirect contribution to RF of additional methane emission is particularly important. It is shown that if global methane emissions were to increase by factors of 2.5 and 5.2 above current emissions, the indirect contributions to RF would be about 250% and 400%, respectively, of the RF that can be attributed to directly emitted methane alone. Assuming several hypothetical scenarios of CH₄ release associated with permafrost thaw, shallow marine hydrate degassing, and submarine landslides, we find a strong positive feedback on RF through atmospheric chemistry. In particular, the impact of CH₄ is enhanced through increase of its lifetime, and of atmospheric abundances of ozone, stratospheric water vapor, and CO₂ as a result of atmospheric chemical processes. Despite uncertainties in emission scenarios, our results provide a better understanding of the feedbacks in the atmospheric chemistry that would amplify climate warming.

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1. Introduction

[2] Methane (CH₄) is an important greenhouse gas with a radiative forcing (RF) of 0.48 Wm⁻², due to anthropogenic activity since preindustrial time [Forster *et al.*, 2007], being second only to CO₂ among the anthropogenic greenhouse gases. Its distribution and growth are well documented [Forster *et al.*, 2007; Ramaswamy *et al.*, 2001] showing a significant increase in atmospheric concentrations since preindustrial times. Analyses of ice core data for the last 650,000 years show that atmospheric CH₄ concentrations varied from approximately 400 ppb during glacial periods to approximately 700 ppb during interglacial periods. The tro-

pospheric average concentration is currently about 1,800 ppb, representing an approximate 2.5 increase since preindustrial time. The atmospheric concentrations in 2005 correspond to an atmospheric burden of 4,900 Tg CH₄ (1 Tg = 10¹² g). Observations since 1984, for which there are continuous measurements, show an increase in atmospheric abundances of CH₄ by about 10%. Growth rates have decreased significantly since the early 1990s, but with pronounced interannual variations [Rigby *et al.*, 2008].

[3] CH₄ is a chemically active greenhouse gas, and its long-term distribution is affected by changes in both emissions and the atmospheric oxidation rate. Estimates of current global CH₄ emissions are in the range 500 to 600 Tg CH₄ yr⁻¹ [Denman *et al.*, 2007].

[4] Atmospheric CH₄ is removed through oxidation by the hydroxyl radical (OH), mainly in the troposphere:



[5] A small fraction is also removed by surface deposition. In the stratosphere, where water vapor is in the range of only a few ppm, CH₄ oxidation contributes to water vapor buildup. Since reaction (R1) also represents a significant

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loss path for OH, additional CH₄ emission will suppress OH and thereby increase the CH₄ lifetime, implying further increases in atmospheric CH₄ concentrations [Isaksen and Hov, 1987; Prather et al., 2001]. This represents a positive chemical feedback, with a feedback factor estimated to be about 1.4 (uncertainty range 1.3 to 1.7) for current atmospheric conditions [Prather et al., 2001]. The nonlinearity in the chemical system could result in a significantly enhanced feedback factor for large CH₄ emissions causing large perturbations [Isaksen, 1988].

[6] The growth in atmospheric CH₄ concentrations, a result of an imbalance between sources and sinks, observed over several decades, is reduced markedly during recent years [Dlugokencky et al., 2003]. For instance, estimates reported in IPCC AR4 give close to a balance (about 1 Tg CH₄ yr⁻¹ difference between emissions and loss) for the time period 2000 to 2005 [Denman et al., 2007], compared to 33 Tg CH₄ yr⁻¹ during the 1980s. Although there are no clear indications whether the changes in trends are due to stabilization in emissions or enhanced atmospheric loss through increased OH levels [Prather et al., 2001], Dlugokencky et al. [2003] point to a possible stabilization of emissions in recent decades. This could be a result of changes in emissions from anthropogenic sources and from wetlands [Bousquet et al., 2006]. However, analyses of atmospheric CH₄ changes in 2007 indicate a renewed growth of CH₄ [Rigby et al., 2008].

[7] Increased CH₄ emissions affect climate in several ways: Directly through increased CH₄ concentrations and indirectly through the chemical feedback on CH₄ levels and through production of O₃ and stratospheric H₂O. Furthermore, CO₂ will increase since it is the end product of atmospheric CH₄ oxidation. In the current atmosphere the indirect RF is approximately the same as the direct methane RF, taking into account the effect on its own lifetime, on ozone, and on stratospheric water vapor [Forster et al., 2007].

[8] In this study we use a global Chemical Transport Model (CTM), the Oslo CTM2 [Isaksen et al., 2005; Søvde et al., 2008], to estimate the impact of additional CH₄ emissions on the atmospheric concentrations of the climate gases CH₄, O₃, stratospheric H₂O, and CO₂, and on RF from these forcing agents. The study covers a wide range of hypothetical methane emission scenarios, up to about 5 times the current emission rate. Although there is no evidence supporting the higher emission in this range, we include them in order to demonstrate the particularly strong positive feedback in the chemical system from large methane releases and the general impact on atmospheric composition and on climate forcing.

[9] We further consider two potentially important natural sources of atmospheric CH₄ in relation to the adopted scenarios: (1) Conversion of organic carbon to CH₄, and release when permafrost thaws; and (2) release of CH₄ hydrates in marine sediments. Earlier studies have demonstrated that large releases of CH₄ from natural sources during warming events can have significant impacts on atmospheric CH₄ levels and may have potential synergistic effects leading to increased and/or sustained global warming. Observed surface temperatures in recent years show significant warming, indicating Arctic warming of more than a factor 2 greater than the global mean value [Hansen et al., 2007]. Permafrost thawing could be more extensive than previously

predicted [Camill, 2005; Osterkamp, 2005], with large potential for methane emission.

[10] Atmospheric CH₄ has a global average atmospheric lifetime of approximately 8 to 10 years [Denman et al., 2007]. Thus, atmospheric composition changes initiated by methane emissions from Arctic warming depend critically on the time horizon of the CH₄ releases from thawing permafrost and degassing from marine hydrates (centennial to millennium timescales).

[11] Our calculations are based on a range of hypothetical future emission scenarios for CH₄. There are currently no studies that quantify large CH₄ emission from permafrost thawing and destabilization of marine gas hydrates in the Arctic in a future warmer climate.

2. CH₄ Emissions From the Arctic Region

[12] We consider two major sources of CH₄ emissions from the warming Arctic: (1) Methane produced from microbial degradation of labile organic carbon that becomes bioavailable as permafrost thaws; and (2) methane released from gas hydrate deposits as they dissociate in response to climate warming. Thawing permafrost may also promote emissions from other methane sources in the Arctic, but the amount of methane that could potentially be produced by microbial processes in thawed soils or release of methane from gas hydrates far exceeds that associated with other Arctic sources. There is evidence that continuous permafrost is actively thawing in many circum-Arctic regions, both onshore and in the shallow offshore continental shelves [Rachold et al., 2007].

2.1. Methane and Thawing Permafrost

[13] Thawing of permafrost at a rate of 0.04–0.10 m yr⁻¹ has been observed in some terrestrial upland regions [Osterkamp, 2005], and it is shown that temperatures have increased at depths as great as 25 to 30 m below the surface at some locations in the Arctic during the last two decades [Isaksen et al., 2007; Osterkamp and Jorgenson, 2006]. Camill [2005] finds that if the current rate of temperature increase (which is at least twice that of the global mean temperature increase) continues at his sites in boreal peatlands in Arctic Canada, the permafrost will have disappeared by the end of this century. The observations at this measurement site are consistent with a warming of at least 7–8°C in a high-CO₂-emission scenario.

[14] Northern soils sequester an estimated 1,672 Pg (1 Pg = 1e+15 g) of organic C, 88% of which is stored in perennially frozen ground [Tarnocai et al., 2009]. The yedoma ice complex within the continuous permafrost in Northern Siberia is of particular interest for understanding the interplay among thawing permafrost, organic carbon, and methane emissions. The yedoma complex consists of ice-rich frozen loess and loess-related sediments that cover approximately 1 million km². These sediments have organic carbon content of approximately 27 kg C/m³ [Walter et al., 2006] and represent a large reservoir (~450 Pg C) of labile organic carbon [Zimov et al., 1997]. An additional 240 Pg C are sequestered in continuous permafrost in thick fluvial deltaic deposits. Thawing of this permafrost and particularly of the yedoma ice complex could produce large CH₄ emissions via anaerobic degradation of the labile carbon, as

demonstrated in laboratory incubations by *Zimov et al.* [1997]. In order to have sustained anaerobic conditions in thermokarsting soils, meltwater needs to be retained in the yedoma complex.

[15] Thermokarst lakes (lakes that have an underlying thaw bulb and that are widespread in some parts of the Arctic) are among the best places to study the potential release of CH₄ during permafrost degradation. *Plug and West* [2009] consider thermokarst expansion rates of up to 8 m in a single year. A strong amplification of CH₄ emissions from Arctic lakes due to thawing of permafrost has already been observed in response to ongoing climate warming. For instance, *Walter et al.* [2006] described significant increases (~60%) in CH₄ emissions from North Siberian thermokarst lakes between 1974 and 2000, associated with a more moderate (15%) increase in lake area during the same period. Calculations by *Walter et al.* [2007] suggest that ~50 Pg CH₄ will be released from Siberian thermokarst (thaw) lakes as yedoma permafrost thaws over a timescale of centuries to millennia. CH₄ emission from lakes in other nonyedoma permafrost-dominated regions in the Arctic is also expected as thermokarst lakes form and expand in conjunction with warming and thawing of permafrost.

[16] The remainder of nonyedoma permafrost in the Arctic contains ~1,000 Pg C [*Tarnocai et al.*, 2009]. Assuming that 10% of this permafrost will thaw beneath lakes and 17% of the C in the thawed permafrost will be converted to CH₄, similar to the efficiency of CH₄ production in yedoma, we estimate that 23 Pg CH₄ can be released from nonyedoma thermokarst lakes. Together, thawing of yedoma and nonyedoma permafrost could thus release up to 73 Pg CH₄ to the atmosphere.

[17] Several studies indicate that high-latitude wetlands have CH₄ emissions that are sensitive to increased temperatures. For example, northern peatlands that experience permafrost thaw [*Christensen et al.*, 2004; *Wickland et al.*, 2006] have enhanced CH₄ emissions. Due to uncertainties associated with future surface moisture and redox regimes, enhanced wetland emissions are not explicitly included as a methane source here.

[18] The CH₄ emission rate and the magnitude of its accumulation in the atmosphere strongly depend on the rate of permafrost thaw. Under current assumptions of warming in the Arctic, permafrost thaw and CH₄ release are likely to occur over timescales of centuries to millennia. In scenarios of more enhanced warming of the Arctic than considered in current climate models [*IPCC*, 2007] the timescale for CH₄ release due to permafrost thawing could be shorter. Although the current global contribution from North Siberian thermokarst lake emissions of CH₄ are small, *Walter et al.* [2006] indicate that these emissions have increased significantly (~60%) between 1974 and 2000.

2.2. Methane Hydrates

[19] Gas hydrates (clathrates) are a solid, ice like form of mostly methane, which occur beneath and possibly within [*Dallimore and Collett*, 1995] onshore permafrost and also in subsea permafrost that persists in some high-latitude regions to water depths as great as ~90 m. Both permafrost-associated gas hydrates and the shallowest part of the deepwater marine gas hydrate system are susceptible to

dissociation (breakdown to methane and water) under conditions of a warming Arctic climate.

[20] The most recent review of the numerous published estimates of the amount of methane sequestered in global gas hydrate deposits converges on a range of 3 to 40 × 10¹⁵ m³ of methane [*Boswell and Collett*, 2011], which converts to a range of ~1,600 to 21,000 Pg C. This consensus range brackets some older estimates (3000 Pg C in the work of *Buffett and Archer* [2004]) and a recent estimate of 1,000 to 10,000 Pg C by *Krey et al.* [2009]. Based on the estimates by *Soloviev et al.* [1987], *Shakhova et al.* [2010a] conclude that one quarter of the Arctic ocean shelf contains 540 Pg CH₄ in gas hydrates. This yields an estimated ~1,600 Pg C within gas hydrates associated with subsea permafrost on the Arctic Ocean continental shelves. It is important to note that the formerly terrestrial sediments on these very shallow shelves contain significant additional carbon in nonhydrate form. Like the carbon trapped in terrestrial permafrost, this additional carbon is subject to microbial degradation and CO₂ and CH₄ production as the subsea permafrost thaws.

[21] In the deep geologic past, CH₄ releases from gas hydrates may have been triggered by, but also possibly exacerbated, the extreme warming event at ~55 Ma before present [*Dickens et al.*, 1995, 1997; *Lamarque et al.*, 2006; *Renssen et al.*, 2004; *Schmidt and Shindell*, 2003]. In this study we estimate a range of potential future methane emissions from the various Arctic gas hydrate populations:

[22] 1. Subsea Permafrost: There is substantial evidence that subsea permafrost is undergoing rapid degradation at high northern latitudes [e.g., *Rachold et al.*, 2007; *Shakhova et al.*, 2005]. The current rate of subsea permafrost degradation is unknown, and acceleration in this degradation with recent changes in sea ice cover and thus ocean temperatures is expected, but not yet fully documented. Still, dissociation of methane hydrate that is currently capped by or contained within subsea permafrost is very likely occurring now [e.g., *Shakhova et al.*, 2010b] and should increase as warming affects the ocean-atmosphere system. Methane released from these hydrates would be emitted into shallow seas where relatively little is likely to be oxidized before reaching the atmosphere. *Shakhova et al.* [2008] speculate that 50 Pg CH₄ could be released abruptly at any time from gas hydrates associated with subsea permafrost. Although there is no basis for estimating the rate of such a release, this value is used as a worst case scenario for the numerical model studies.

[23] 2. Methane hydrates in terrestrial permafrost near the top of the gas hydrate stability zone are unlikely to contribute significantly to the net methane flux from these deposits. A critical exception may be beneath deep thermokarst thaw bulbs, where the superposition of climate-induced surface warming and possible thaw bulb growth may so perturb temperatures near the top of the gas hydrate stability zone that methane hydrate at depths of only a few hundred meters might be susceptible to dissociation.

[24] 3. Deepwater marine hydrates: Most deepwater gas hydrates are unlikely to be a major source of atmospheric methane in climate scenarios that consider less than several millennia. Dissolution and oxidation are likely to strongly reduce the amount of methane that reaches the atmosphere once it is emitted at the seafloor. One exception is gas hydrate in continental margin sediments at pressure-temperature conditions close to the gas hydrate phase boundary. For

example, contemporary climate warming may be triggering gas hydrate dissociation and methane emissions along the West Spitsbergen margin [Westbrook *et al.*, 2009]. In that case, much of the methane is emitted rapidly and in bubble streams, meaning that significant amounts could reach the atmosphere. In other cases, oxidation and dissolution might be expected to prevent much methane from crossing the ocean-atmosphere interface. The numerical model studies presented here do not include a potential contribution from this population of potentially degrading gas hydrates.

[25] Submarine landslides associated with gas hydrates and/or underlying free gas are possible for CH₄ release from marine sediments (e.g., discussion and references in the work of Archer [2007]). It is assumed that single landslides can release up to about 5 Pg C as CH₄, based on estimates of past major landslides. This can be regarded as an upper estimate from single landslides, since a recent estimate indicates lower release [Paull *et al.*, 2007]. Our models do not include this as a potential carbon source.

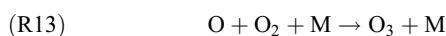
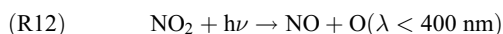
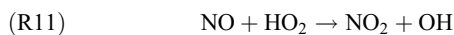
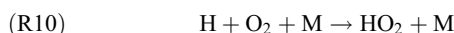
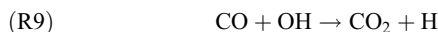
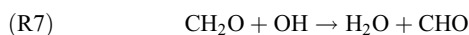
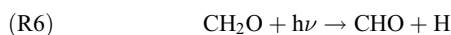
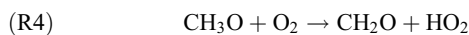
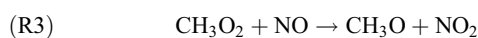
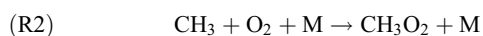
2.3. Model Inputs

[26] Clearly, large uncertainties exist in the amount of carbon stored in Arctic permafrost as methane hydrates, the fraction emitted as CH₄ from thawing permafrost and the timing and magnitude of potential emissions from thawing permafrost and gas hydrates. Combining these with our limited knowledge of the rate and degree of long-term global warming it is clear that our estimates of the impact of future emissions of CH₄ on atmospheric composition and on climate are also associated with large uncertainties. In order to demonstrate the potential contribution of high-latitude release of CH₄ to global warming, several scenarios are considered here. The scenarios discussed in section 5 are based on the potential future emissions of CH₄ estimated for the Arctic permafrost region and for CH₄ stored in hydrates. The estimates include (1) sustained emissions over a specified time horizon from permafrost thawing necessary to reach a defined level of atmospheric CH₄ enhancement and (2) a hypothetical release of 50 Pg of CH₄ from the hydrate pool as a pulse over a limited period of time (1 year) or as sustained emissions over several decades. Although the high-emission scenarios are unlikely to occur, they are compatible with the current knowledge of the cumulative magnitude of CH₄ that might be emitted from permafrost thawing and from CH₄ hydrate destabilization.

3. Atmospheric CH₄ Oxidation

[27] This section provides a brief description of the atmospheric chemistry leading to the formation of greenhouse gases from CH₄ emissions.

[28] CH₄ oxidation leads to enhanced formation of ozone in the troposphere and lower stratosphere through a sequence of reactions involving NO_x compounds. The CH₃ resulting from reaction (R1) is oxidized and the reaction products are photolyzed in the presence of sunlight:



[29] M is an air molecule (usually N₂), participating in a three-body reaction, and “hν” represents the solar photon flux. Through this cycle ozone is efficiently formed in the presence of NO_x, CO and CH₄. The end product of the OH and HO₂ formation from CH₄ is water vapor. As two H₂O molecules are formed from each CH₄ molecule, water vapor enhancements due to additional CH₄ releases can be important in relative terms in the dry stratosphere. Reaction (R9) yields CO₂, constituting another important product from CH₄ oxidation. The result of the CH₄ oxidation chain is thus the formation of the three greenhouse gases O₃, H₂O, and CO₂, which comes in addition to the enhancement of CH₄ concentrations due to direct emissions.

4. Experimental Setup

4.1. Model Description

[30] We apply the global-scale Oslo CTM2 model (Oslo Chemical Transport Model). The model runs with meteorology from the European Centre for Medium Range Weather Forecasts (ECMWF) and uses the Second-Order Moments Scheme [Prather, 1986] for advective transport. Surface emissions are based on the EDGARv3.2 database [Olivier and Berdowski, 2001] for anthropogenic emissions, and on the work of Müller [1992] for natural emissions. CH₄ levels are prescribed in the troposphere with latitudinal distributions based on observations from the ESRL network [Dlugokencky *et al.*, 2010; Masarie *et al.*, 1991]. The model has a resolution, which is 2.8 × 2.8 degrees (T42) horizontally and 60 vertical layers in the troposphere and stratosphere, and has extensive tropospheric and stratospheric chemistry which has been validated in model-model and model-measurement inter comparisons of relevance to CH₄ perturbations [Gauss *et al.*, 2006; Isaksen *et al.*, 2005;

Table 1. Changes in CH₄ Burden Chosen for the 2.5 × CH₄, 4 × CH₄, 7 × CH₄, and 13 × CH₄ Cases Addressed by Oslo CTM2 Model Runs, Along With the Lifetimes τ_2 as Modeled by Oslo CTM2 and the Increase With Respect to the Base Case Lifetime τ_1 ^a

Name of Case	$\Delta M/M_1$	Lifetime of CH ₄ (years), τ_2	τ_2/τ_1 ($=\beta$)	Δe in Pg CH ₄ yr ⁻¹	$\Delta e/e_1$	η
2.5 × CH ₄	1.5	12.4	1.36	0.45	0.8	1.8
4 × CH ₄	3	14.7	1.61	0.80	1.5	2.0
7 × CH ₄	6	18.0	1.98	1.37	2.6	2.4
13 × CH ₄	12	22.6	2.50	2.28	4.2	2.9
100 Tg	0.27	9.7	1.07	0.1	0.2	1.5
200 Tg	0.58	10.4	1.15	0.2	0.4	1.6
Hydrate	10.78 ^b	21.2 ^b	2.34 ^b	50 (1 year only)	93	2.7 ^b

^aAlso shown are the emission enhancements required to bring about the chosen enhancements in atmospheric CH₄. $M_1 = 4.9$ Pg CH₄, $e_1 = 540$ Tg CH₄ yr⁻¹, and $\tau_1 = 9.1$ years refer to CH₄ burden, emission and lifetime in the base case. ΔM and Δe denote the absolute increases from the base to the perturbation cases. The enhancement factor η (see text) is a measure of the positive feedback in the chemistry of CH₄ and is calculated from the changes in CH₄ lifetime and burden at the new equilibrium. The three last rows show additional cases, which have not been simulated by the Oslo CTM2 model, but are based on emission estimates assuming thawing of permafrost, and a sudden release of CH₄ from marine sediments.

^bNumbers apply to peak concentrations reached after 1 year.

Shindell et al., 2006]. The model is further documented by *Berglen et al.* [2004] and *Søvde et al.* [2008].

4.2. Simulations and Definitions

[31] In the Oslo CTM2 model simulations atmospheric CH₄ levels are fixed. The estimates of corresponding emissions are made with a forward explicit box model and described in more detail in section 5. Five simulations are made with 2003 meteorology. A base case simulation with present-day CH₄ levels, and 4 perturbation cases with tropospheric CH₄ levels multiplied by 2.5 (“2.5 × CH₄”), four (“4 × CH₄”), seven (“7 × CH₄”), and thirteen (“13 × CH₄”). For chlorine and bromine levels we use values suggested by *WMO* [2003] for 2100, as we are considering long-term future changes. In each of the simulations the global lifetime of CH₄ is calculated from a mass-weighted average of loss through reaction with OH. The base case simulation was spun up for several model years until a stable chemical composition was reached in both the troposphere and the stratosphere. Starting from the resulting chemical fields, both the base case and the perturbation cases were run for an additional 4 years to reach a new equilibrium corresponding to the respective CH₄ levels.

[32] In the calculations we have used current atmospheric water vapor content. Since water vapor is expected to increase in a future warmer climate the calculations were repeated for a 40% increase in tropospheric water vapor (but no other changes). Such perturbations in tropospheric water vapor are expected to be representative for strongly perturbed future climate conditions and key compounds in the interactive climate-chemistry system like OH and O₃ will be affected. We found that the calculated tracer and lifetime perturbations were only slightly affected by this increase (less than 10% impact).

[33] We define an enhancement factor η to quantify the effect of CH₄ emissions on its own concentration and lifetime. η is given as the ratio of relative change in CH₄ burden over relative change in CH₄ emissions, i.e., $\eta = (M_2 - M_1)/M_1/(e_2 - e_1)/e_1$, where M and e represent the global CH₄ burden and global CH₄ emission at steady state, respectively. Subscript 1 is for the base case, and subscript 2 is for the perturbation cases. The feedback in atmospheric concentration from emissions is positive if $\eta > 1$. We define

further the ratio of burden $\alpha = M_2/M_1$ and the ratio of CH₄ lifetimes as $\beta = \tau_2/\tau_1$.

5. Results

5.1. Atmospheric Enhancement of CH₄

[34] Estimates of current atmospheric CH₄ lifetime are in the range 8 to 10 years [*Shindell et al.*, 2006]. In the Oslo CTM2 model the lifetime of CH₄ amounts to 9.1 years in the base case. Table 1 summarizes the main results for the four perturbation cases. For example, in the 4 × CH₄ case we choose a fourfold increase in atmospheric CH₄, i.e., $\alpha = 4$ (or $\Delta M/M_1 = 3$) in the Oslo CTM2 model calculation and obtain a new CH₄ lifetime of $\tau_2 = 14.7$ years, giving a relative increase in lifetime of $\beta = 1.61$. The 7 × CH₄ case with $\alpha = 7$ almost doubles the CH₄ lifetime, $\beta = 1.98$.

[35] Figure 1 shows β as a function of CH₄ increase. The circles show the increases of CH₄ lifetime as represented by $\beta = \tau_2/\tau_1$ for the four different cases of enhancement modeled by Oslo CTM2. To obtain intermediate values of β we apply cubic interpolation, which is assumed to approximate the relation between β and α ($= M_2/M_1$) well within the considered range.

5.2. Calculated Atmospheric Changes in H₂O, OH, and Ozone

[36] Figure 2 shows model calculated changes in zonal-mean atmospheric water vapor, OH, and ozone for the 4 × CH₄, 7 × CH₄, and 13 × CH₄ cases. Related to the negative height dependence of CH₄ mixing ratio, stratospheric water vapor increases strongly with height. More CH₄ leads to more stratospheric water vapor, and this increase is also significant below 30 km where stratospheric water vapor is radiatively efficient. OH decreases strongly in most of the troposphere due to increased CH₄ oxidation. The strongest decrease in OH is found in the lower troposphere with reductions by more than 50% in the three cases. In the stratosphere, OH increases as a result of enhanced production from water vapor. The ozone increase is particularly large in the upper tropical troposphere where the ozone radiative forcing is strong [*Ramaswamy et al.*, 2001], while in the lower stratosphere the ozone increase is modest, with regions where ozone even decreases slightly when CH₄ is perturbed.

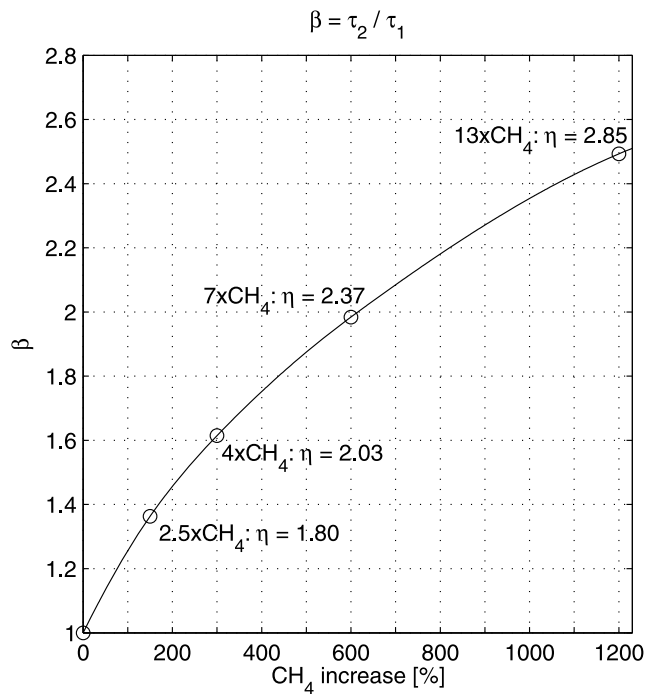


Figure 1. The increase in CH₄ lifetime as a function of the increase in CH₄ burden.

In the upper stratosphere ozone is reduced due to the more efficient removal by HO_x (odd hydrogen) reactions.

5.3. Relating the Chosen CH₄ Enhancements to Emissions and Available Storage

[37] A forward explicit box model is applied to calculate the evolution of CH₄ as function of time assuming different (sustained) CH₄ emission rates. The calculation starts at present CH₄ levels, and at each time step (0.01 years) it uses the CH₄ lifetime that corresponds to the CH₄ concentration calculated by the box model for the current time step. The relation between CH₄ lifetime and concentration calculated in the Oslo CTM2 simulations (Figure 1) is taken as input to the box model. The resulting evolution of CH₄ is shown in Figure 3. 96%, 92% and 87% of the chosen increases are reached after 50 years in the 2.5 × CH₄, 4 × CH₄ and 7 × CH₄ cases, respectively, reflecting the increase in lifetime with increasing emission.

[38] Table 1 also lists the additional CH₄ emissions (Δe) needed to reach the adopted equilibrium enhancement values in the different perturbation experiments. These additional emissions are switched on in the forward explicit box model from the beginning. The base case level is consistent with an emission of 540 Tg CH₄ yr⁻¹. Total equilibrium emissions can thus be obtained by adding 540 Tg CH₄ yr⁻¹ to the values given in Table 1. However, an important question in relation to the equilibrium value is the timescale for the additional emission.

[39] Our highest estimate for thermokarst lakes is a 100 to 200 Tg CH₄ yr⁻¹ emission, which can be sustained for a

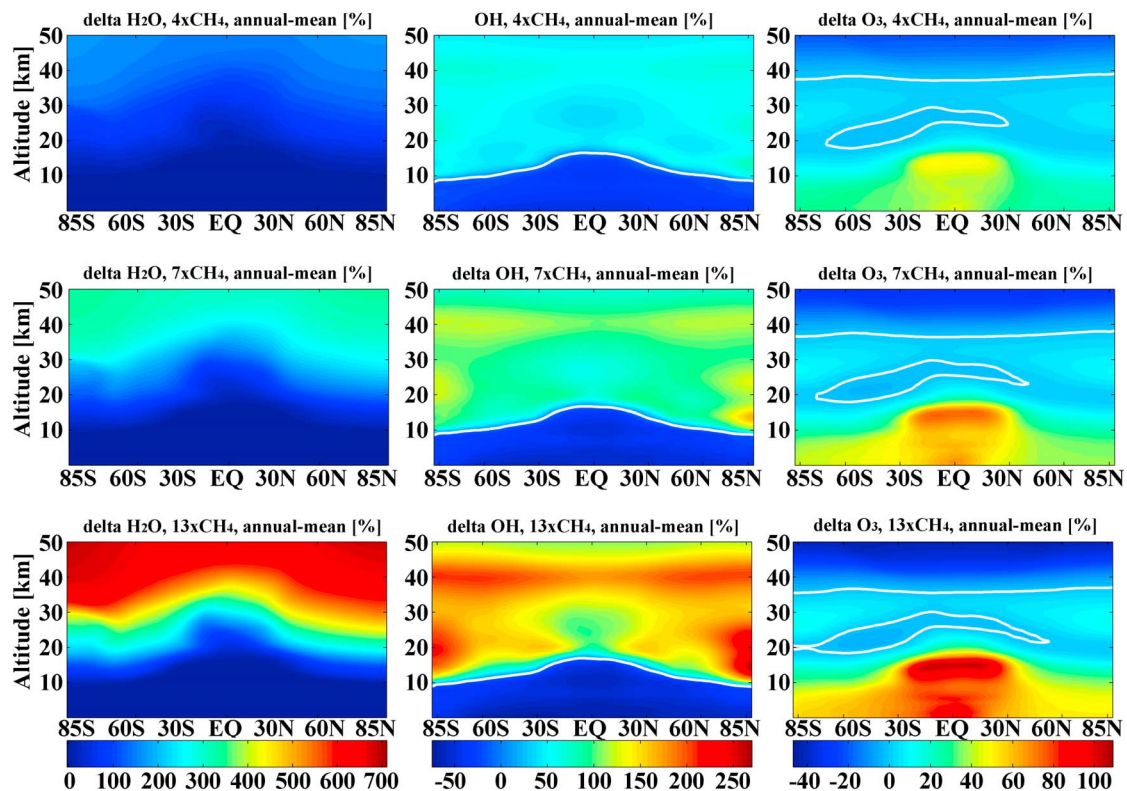


Figure 2. Annual and zonal mean relative change in stratospheric water vapor, the hydroxyl radical, and ozone for 4, 7, and 13 times current CH₄ levels. White contour lines indicate zero change.

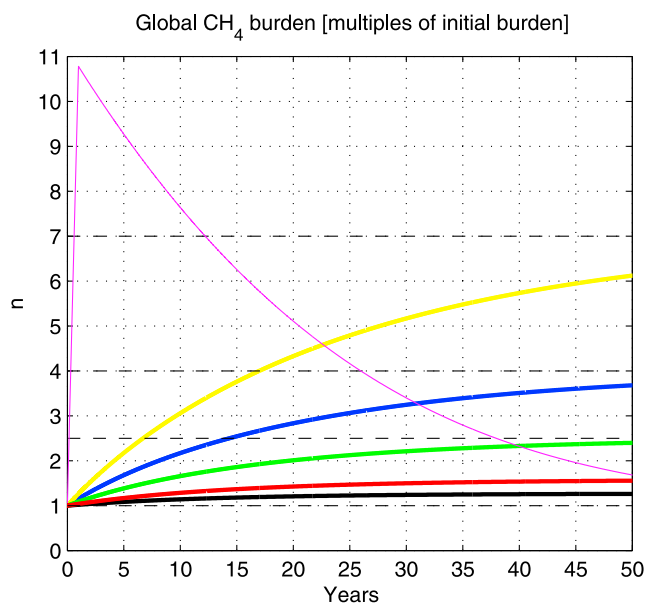


Figure 3. Global CH₄ burden as function of time for scenarios $2.5 \times \text{CH}_4$ (green), $4 \times \text{CH}_4$ (blue), and $7 \times \text{CH}_4$ (yellow). Additional scenarios are indicated, for which Oslo CTM2 simulations were not performed, but the lifetime dependence of CH₄ on its own concentration is taken into account: $100 \text{ Tg}(\text{CH}_4) \text{ yr}^{-1}$ sustained (black), $200 \text{ Tg}(\text{CH}_4) \text{ yr}^{-1}$ sustained (red), and a 1 year $50 \text{ Pg}(\text{CH}_4) \text{ yr}^{-1}$ emission scenario (“hydrate” scenario, purple). Horizontal gray lines denote current global burden (1) and the three enhanced levels 2.5, 4, and 7.

time period of up to 500 years. Such enhanced emissions would increase the CH₄ lifetime by about 7 to 15%, and the CH₄ burden by factors of 1.3 to 1.6 (Table 1).

[40] To illustrate the atmospheric impact on CH₄ of large pulse type releases, we assume a hypothetical case of 50 Pg CH₄ emission, corresponding to CH₄ hydrate releases from the Siberian Arctic shelf (as defined in section 2). The atmospheric impact for a pulse emission (over 1 year) and for sustained emissions over several decades, are considered. Our calculations (Figure 3) show that a 1 year pulse emission of 50 Pg CH₄ would lead to abundances peaking at about 11 times its current level. Table 1 gives the yearly emissions needed to give equilibrium abundances in the atmosphere 2.5 times, 4 times and 7 times current abundances. A total of 50 Pg CH₄ can sustain the additional emissions in the three cases for 111, 62.5, and 36.5 years, respectively, giving enhancements that are approaching the equilibrium values.

[41] For the sustained 100 Tg CH₄ yr⁻¹, 200 Tg CH₄ yr⁻¹ and the 1 year 50 Pg CH₄ emission cases, the CH₄ lifetime dependence derived from the four explicit Oslo CTM2 simulations (shown in Figure 1) is taken into account in the calculation of CH₄ evolution.

5.4. Atmospheric Enhancement of CO₂

[42] CH₄ is oxidized to CO₂ in the atmosphere, and CO₂ decays according to different loss processes. Our calculation of the evolution of CO₂ (shown in Figure 4) from oxidation of the CH₄ released from thawing permafrost or hydrate

storage is based on the formula of IPCC-AR4 [Forster et al., 2007], according to which the decay of a pulse of CO₂ with time t is given by

$$a_0 + \sum_{i=1}^3 a_i \cdot \exp(-t/\tau_i)$$

Where $a_0 = 0.217$, $a_1 = 0.259$, $a_2 = 0.338$, $a_3 = 0.186$, $\tau_1 = 172.9$ years, $\tau_2 = 18.51$ years, and $\tau_3 = 1.186$ years. The pulses at each time step are set equal to the amount that is oxidized from the additional atmospheric CH₄ from permafrost emissions. After 50 years, the CO₂ values are 4.4, 7.3, and 11.3 ppm (ppm) for the $2.5 \times \text{CH}_4$, $4 \times \text{CH}_4$, and $7 \times \text{CH}_4$ cases, respectively. For comparison with Figure 3, Figure 4 also includes the CH₄ hydrate case for which the CO₂ enhancement peaks at around 10 ppm after 40 to 50 years. It has to be noted that the CO₂ shown in Figure 4 does not include directly emitted CO₂ from permafrost thaw. The inclusion of CO₂ in these calculations is consistent with the inclusion of CH₄ oxidation to CO₂ in the GWP for CH₄ [Boucher et al., 2009].

5.5. Radiative Forcing

[43] Detailed off-line atmospheric radiative transfer schemes are used in the calculations of radiative forcing (RF), from changes in ozone and stratospheric water vapor (sH₂O) [Myhre et al., 2000; Myhre et al., 2007]. For CO₂ and CH₄, expressions given by Ramaswamy et al. [2001] are used. The radiative forcing is not related to a particular year but to the time when the CH₄ increase is reached. The $13 \times$

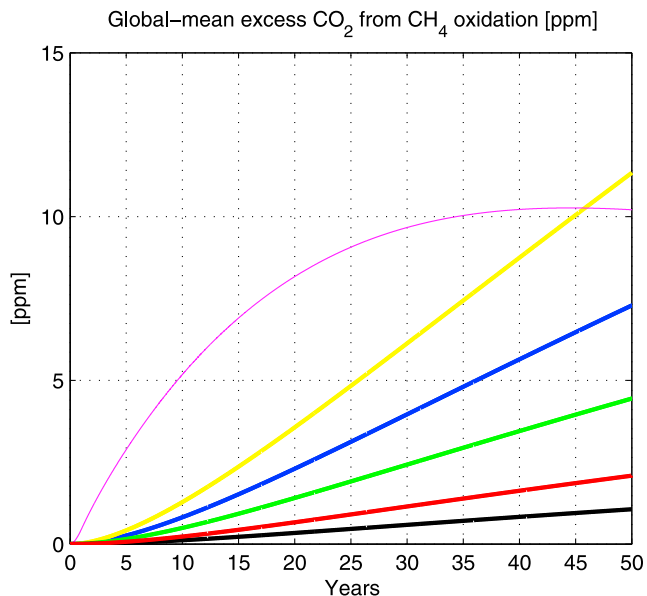


Figure 4. Global-mean mixing ratio of excess CO₂ from additional CH₄ oxidation for scenarios $2.5 \times \text{CH}_4$ (green), $4 \times \text{CH}_4$ (blue), and $7 \times \text{CH}_4$ (yellow). Additional scenarios are indicated, for which Oslo CTM2 simulations were not performed, but the lifetime dependence of CH₄ on its own concentration is taken into account: 100 Tg CH₄ yr⁻¹ sustained (black), 200 Tg CH₄ yr⁻¹ sustained (red), and 50 Pg CH₄ yr⁻¹ switched off after 1 year (“hydrate” scenario, purple).

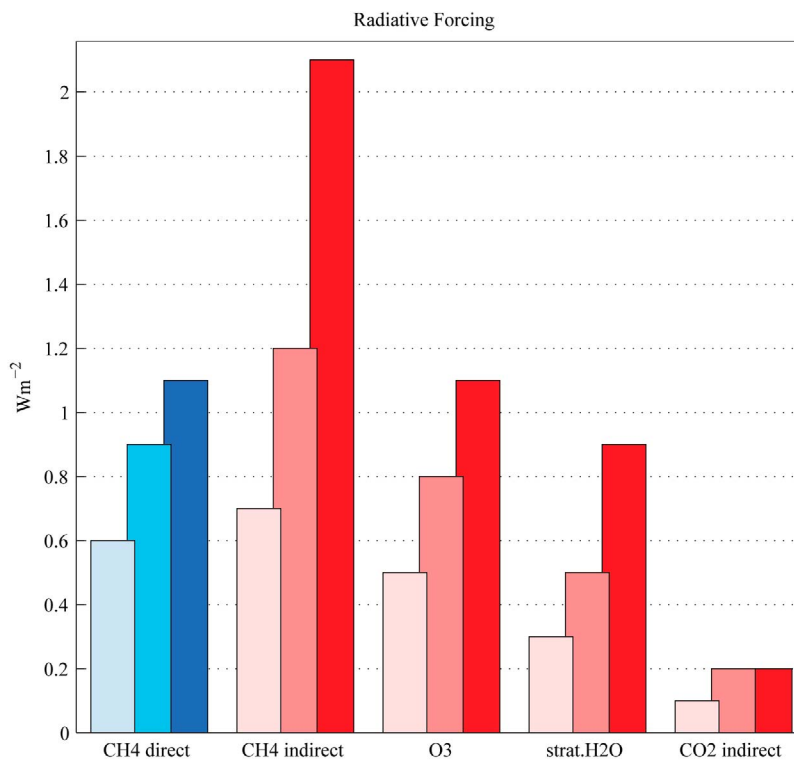


Figure 5. Radiative forcing resulting from CH₄ increases. Blue bars denote contributions from direct emissions, and red bars are contributions from CH₄ initiated changes in atmospheric composition. “CH₄ indirect” represents the CH₄ enhancement that is due to the increase in its lifetime. “CO₂ indirect” relates to the CO₂ enhancement due to oxidation of the additional CH₄. The lightest colors refer to the 4 × CH₄, medium light colors to the 7 × CH₄ and dark colors to the 13 × CH₄ case. The radiative forcing is not related to a particular year but to the time when the CH₄ increase is reached. The 13 × CH₄ case refers to a shorter time horizon (30 years) while the other cases are based on a 50 year time horizon.

CH₄ case refers to a shorter time horizon (30 years) while the other cases are based on a 50 year time horizon.

[44] Results are shown in Figure 5 for the 4 × CH₄, 7 × CH₄ and 13 × CH₄ cases. Radiative forcings due to CH₄ are listed in Table 2. In the 4 × CH₄ case the RF caused by direct emissions of CH₄ and the RF due to the lifetime increase are comparable and add up to 1.3 Wm⁻². The RF of 0.5 Wm⁻² from ozone, calculated for the same case, is probably stronger than the forcing since preindustrial time [Gauss *et al.*, 2006]. For sH₂O we calculate nearly quadrupling (0.3 Wm⁻²) of the current sH₂O forcing (0.08 Wm⁻²) [Myhre *et al.*, 2007]. In the 7 × CH₄ and 13 × CH₄ cases the indirect CH₄ RF clearly dominates the direct one, due to more pronounced OH changes. The total RF is 2.2, 3.6, and 5.4 Wm⁻² for the 4 × CH₄, 7 × CH₄ and 13 × CH₄ cases, respectively.

[45] The importance of CH₄ emissions through atmospheric chemistry perturbation is illustrated by a 3.5- to fivefold increase in the total RF compared to the contribution from direct emissions, increasing in importance with the increase in CH₄ emission. The impact of chemical perturbations by CH₄ emission increases is substantially higher than what is included in the GWP value for CH₄ used in IPCC [Forster *et al.*, 2007], where a twofold increase in RF was adopted to account for indirect effects at current conditions. The CH₄ lifetime feedback represents the most important contribution among the additional radiative for-

ings due to changes in atmospheric composition, followed by ozone (in particular tropospheric ozone) and sH₂O.

[46] According to our simulations, sustained CH₄ emissions of 100 Tg yr⁻¹ to 200 Tg yr⁻¹ due to permafrost thawing will correspond to a RF of 0.3 to 0.6 Wm⁻² in a new equilibrium (reached after a few decades). Although the amount of CH₄ released to the atmosphere is large, the short lifetime (8 to 10 years) compared to the time horizon for

Table 2. Radiative Forcing Due to Directly Emitted CH₄ (CH₄ Direct Forcing), the Lifetime Change of CH₄ (CH₄ Indirect Forcing), and the Chemical Enhancements Due to the Feedbacks in the 4 × CH₄, 7 × CH₄, and 13 × CH₄ Cases^a

Name of Case	CH ₄ Direct Forcing	CH ₄ Indirect Forcing	Indirect Forcing From O ₃ , strat. H ₂ O and CO ₂
4 × CH ₄	0.6 Wm ⁻²	0.7 Wm ⁻²	0.9 Wm ⁻²
7 × CH ₄	0.9 Wm ⁻²	1.2 Wm ⁻²	1.5 Wm ⁻²
13 × CH ₄	1.1 Wm ⁻²	2.1 Wm ⁻²	2.2 Wm ⁻²
<i>100 Tg</i>	<i>0.1 Wm⁻²</i>	<i>0.1 Wm⁻²</i>	<i>0.1 Wm⁻²</i>
<i>200 Tg</i>	<i>0.2 Wm⁻²</i>	<i>0.2 Wm⁻²</i>	<i>0.2 Wm⁻²</i>

^aFor the “100 Tg” and “200 Tg” cases (in the table listed in italics) no Oslo CTM2 calculations have been made. Thus the split into direct and indirect components of radiative forcing is based on extrapolated values for β. Contributions to indirect radiative forcing from perturbations of O₃, stratospheric H₂O and CO₂ are not included in the table.

emission (up to 500 years) means that equilibrium in atmospheric concentration is reached at an early stage in the time period. As an illustration of the potential of permafrost CH₄ releases, the increase of the CH₄ concentration by a factor 2.5 compared to current CH₄ concentrations would correspond to an extra emission of 450 Tg CH₄ yr⁻¹ from permafrost thawing and give a radiative forcing of about 1.5 Wm⁻². However, the emission would be over a shorter time horizon than in the 100 Tg CH₄ yr⁻¹ to 200 Tg CH₄ yr⁻¹ emission cases.

[47] A release of 50 Pg (CH₄) from gas hydrates over a 30 year time period could give a RF in excess of 4 Wm⁻². The larger calculated RF, results from the fact that this case represents a more rapid emission than our 7 × CH₄ case.

[48] The release of up to 5 Pg C estimated for single submarine landslides (see discussion in section 2) can enhance atmospheric CH₄ by nearly 2.5 ppm, and give a RF slightly larger than 1 Wm⁻². This is substantially higher than the 0.2 Wm⁻² (only the direct RF was taken into account) from a similar CH₄ release discussed by Archer [2007]. It has to be noted, however, that more recent estimates of the emission from the Storegga Landslide are lower than 5 Pg C [e.g., Paull et al., 2007], and that there is no evidence that the CH₄ released escaped to the atmosphere after the event.

6. Discussion and Conclusion

[49] Relating the magnitude and timing of CH₄ release in a warming Arctic to the adopted scenarios applied in the model studies is challenging due to uncertainties associated with the rate of permafrost warming and thawing, the size and distribution of the large organic C pool [Tarnocai et al., 2009] and its fate in microbial decomposition under aerobic versus anaerobic conditions [Schuur et al., 2008; Walter et al., 2007], and the size and stability of the hydrate reservoir. However, given the magnitude and vulnerability of the permafrost organic carbon pool and the large permafrost-associated CH₄ hydrate reservoir, quantification of the feedbacks associated with the release of CH₄ from these systems is important for climate modeling.

[50] There is a potential for the release of large amounts of carbon stored in the Arctic to the atmosphere as CH₄ following the thawing of permafrost and warming of oceans [Buffett and Archer, 2004; Schuur et al., 2008; Walter et al., 2007, 2006]. Considering the positive feedback (increase in lifetime) and the non linearity in atmospheric chemistry through amplification of atmospheric CH₄ (represented by the enhancement factor η), we have applied a wide range of CH₄ emission scenarios to explore the impact of such interactions. The impact on methane lifetime does not only apply to the additionally emitted CH₄ but also to background levels of CH₄.

[51] Although the high-CH₄ scenarios applied in this study are unlikely, they demonstrate the strong CH₄ feedbacks in the climate system, with large amplification of atmospheric composition changes and RF compared to the direct RF of CH₄ emissions.

[52] The Oslo CTM2 has been thoroughly tested with respect to OH and CH₄ lifetime through model comparisons and through perturbation studies [Hoor et al., 2009; Shindell et al., 2006]. These studies show that there are noticeable differences in model estimates. Another model uncertainty

in the Oslo CTM2 calculations may arise from the use of today's meteorology, uncertainties in emissions of other gases than CH₄ (e.g., emissions of NO_x and CO), and the formulation of dry deposition (O₃) and wet deposition (NO_x) [Isaksen et al., 2005; Solberg et al., 2008]. Yet, the uncertainties in the atmospheric chemistry calculations are likely to be small compared to the uncertainties in the amount and time horizon for CH₄ emission from thawing permafrost or degassing hydrates.

[53] Since current atmospheric CH₄ lifetime ranges between 8 and 10 years, the RF of CH₄ emissions from permafrost thawing and CH₄ hydrate degassing depends critically on the rate of emissions. Additional studies linking CH₄ emissions to the possibilities for large future warming in the Arctic are needed.

[54] Although CH₄ emissions from permafrost thawing and hydrate degassing are likely to have a very different spatiotemporal distribution than anthropogenic emissions, the impact on climate will be global in the same way as the impact from CH₄ emitted in other regions, when we take into account the multiyear lifetime of CH₄.

[55] Fossil fuel CO₂ emissions have increased substantially over the last decade and is now 40% higher than in 1990 [Le Quéré et al., 2009; Myhre et al., 2009]. The continued increase in greenhouse gas emissions toward the end of this century has the potential to produce significant warming at high northern latitudes well beyond what has been observed during the last decades [Hansen et al., 2007; IPCC, 2007]. There is a possibility that the Arctic temperature increases could be followed by extensive permafrost thawing, with enhanced CH₄ emission from thermokarst lakes [Walter et al., 2006], with later release of CH₄ from gas hydrates that would eventually be affected by warming temperatures. Considering the large, nonlinear atmospheric chemistry feedbacks discussed here, future CH₄ emissions from permafrost deposits could be a larger concern for climate warming than previously thought.

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