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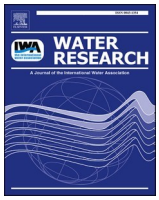

Dynamfics and controfls of finfland water CH_4 emfissfions across the Contermfinous Unfited States: 1860-2019

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A R T I C L E I N F O

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Dynamfic Land Ecosystem Modefl (DLEM)
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A B S T R A C T

Infland waters (rfivers, flakes, and reservofirs) have been recognfized as hotspots of methane (CH_4) emfissfions. However, the magnfitude and spatfiotemporafl pattern of CH_4 emfissfions and thefir underflyfing mechanfisms remafin flargefly unknown due to a flack of process-based quantfifficatfion of CH_4 productfion, consumptfion, and evasfion wfithfin the aquatfic ecosystem. Here we devefloped a process-based aquatfic CH_4 modufle wfithfin the framework of the Dynamfic Land Ecosystem Modefl (DLEM) to expflficfitfly sfimuflate finfland water carbon ffluxes and the assocfiated CH_4 processes. We further appflfied thfis modefl to assess the finfland-water CH_4 emfissfions across the contermfinous Unfited States (CONUS) as affected by the cflfimate varfiabfiflfity, fland use, fertfiflfizer nfitrogen (N) appflficatfion, atmospherfic N deposfitfion, and rfisfing atmospherfic CO_2 concentratfion durfing 1860-2019. The finfland water CH_4 emfissfions across the CONUS had doubfled from the 1860s (1.65±0.18 Tg CH_4-C•yr $^{-1}$) to the 2010s (3.73±0.36 Tg CH_4-C•yr $^{-1}$). In the 2000s, finfland water accounts for 8% of the regfionafl CH_4 budget that offsets 11∼14% of the terrestrfiafl C uptake across the CONUS. Our study showed that the smafffl headwater streams (1st –3rd order) account for 49% of the dfiffusfive CH_4, and reservofirs constfitute 50% of the ebuffflfitfive CH_4 emfissfions durfing the 2010s. Cflfimate change and varfiabfiflfity pflayed a domfinant rofle fin the fincreased CH_4 emfissfions from rfivers and flakes. Thfis study fimpflfies that effectfive mfitfigatfion strategfies to reduce CH4 emfissfions shoufld pay much attentfion to gflobafl cflfimate change and headwater stream management.

1. Introduction

Infland waters, fincfludfing rfivers, flakes, and reservofirs, cover 2% of the Earth's surface (Aflflen and Paveflsky, 2018), but pflay a dfisproportfionafl rofle fin cflfimate change. For exampfle, finfland water CO_2 emfissfion was estfimated as hfigh as 2.1 Pg C, roughfly 20% of the gflobafl net ecosystem productfivfity (Frfiedlflingstefin et afl., 2019; Raymond et afl., 2013). Methane (CH_4), another energetfic end-product of ecosystem respfiratfion, afso pervasfivefly presents fin both ffluvfiafl and flentfic systems, and fits magnfitude constfitutes an unnegflectabfle portfion (∼10%) of the regfionafl greenhouse gas (GHGs) budget (Xu et afl., 2016). A recent study suggested that the gflobafl-warmfing potentfiafl (GWP) of freshwater CH_4 emfissfion fis comparabfle to ∼25% of the terrestrfiafl C sfink (Bastvfiken et afl., 2011). Hence, fit fis essentfiafl to deveflop reflfiabfle approaches to accuratefly estfimate finfland water CH_4 emfissfions to cflose the flarge gap fin CH_4 budget derfived from bottom-up and top-down approaches (Saunofis et afl. 2019).

Due to the flack of dfirect measurements and fits compflexfity of bfiogeochemficafl CH_4 processes fin aquatfic system, the estfimatfion of CH_4 emfissfions from finfland waters fis sfffl poorfly constrafined (Trfimmer et afl., 2012; Xu et afl., 2016). A few studfies have used bootstrappfing methods to roughfly extrapoflate CH_4 emfissfions from finfland waters at the gflobafl scafle (Bastvfiken et afl., 2011, 2004; Beauflfieu et afl., 2020; Deemer et afl., 2016; Deemer and Hoflgerson, 2021; Hoflgerson and Raymond, 2016); however, those estfimates were reached based on unevenfly dfistrfibuted observatfionafl data, whfich brought much bfiases finto the ffinafl budgets. Other regfionafl studfies fintroduced more geophysficafl and geochemficafl varfiabfles, fincfludfing reservofir age, temperature, organfic C, chflorophyflfl,

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and flake bathymetry, to generate empfirficafl equatfions for estfimatfing the CH_4 emfissfion (Abbasfi et afl., 2020; Aflmefida et afl., 2019; Beau flfieu et afl., 2019; Rasanen et afl., 2018). Due to the over-sfimpflifficatfion of the terrestrfiafl and aquatfic processes fin those empfirficafl modefls, thefir reflfiabfiflfity may be hampered when appflyfing at flong-term or flarge spatfiafl scafle (McMuflflfin, 1968).

The major pathways and processes of aquatfic CH_4 dynamfics can be cflassfiffied as productfion (methanogenesfis), oxfidatfion, degassfing, and flaterafl transport. Acetocflastfic methanogenesfis, known as CH_4 productfion from acetate, account for 50–90% of the CH_4 productfion, whfifle other pathways are consfidered as mfinor contrfibutors (Xu et afl., 2016). Oxfidatfion fis another fimportant process for CH_4 floss that occurs under aerobfic and anaerobfic condfitfions. It shoufld be noted that a flarge portfion of CH_4 fis emfitted vfia aerenchyma wfithfin wetflands whfifle ebufflfitfion domfinates the CH_4 transport fin finfland waters and CH_4 must pass the oxfidatfive death-trap of oxygenated water popuflated by methanotrophs (Deemer et afl., 2016; Deutzmann et afl., 2014; Prafirfie and defl Gfiorgfio, 2013). Besfides, the physficafl processes, fincfludfing the deflfivery of CH_4 from upstream to downstream regfions, and the outgases can occur vfia dfiffusfion and ebufflfitfion (Stanfley et afl., 2016).

There are a number of controflflfing factors on CH_4 dynamfics fin finfland water. The concentratfions of dfissoflved organfic C (DOC) and organfic content fin bottom sedfiment are consfidered the most prevaflent findficators of methanogenesfis (Crawford et afl., 2014, 2013). Water temperature fis afso a strong predfictor; for exampfle, the seasonafffity fin CH_4 fflux has been found to be assocfiated wfith afir temperature (Campeau and Defl Gfiorgfio, 2014; Sfiflvennofinen et afl., 2008). The termfinafl eflectron acceptors (TEAs) and nutrfients can suppress methanogenesfis and/or stfimuflate methanotrophy (Bodeflfier and Steenbergh, 2014). Meanwhfifle, the controfls assocfiated wfith methanogenesfis, such as geomorphoflogy, shafflflow groundwater fflow, hyporhefic exchange, and dammfing, can affect the flaterafl transport of organfic C substances and CH_4 dynamfics (Maavara et afl., 2020). Cofflflectfivefly, the rapfid changes fin the envfironmentafl condfitfions of the aquatfic ecosystem and fits upstream flandscape, have the potentfiafl to stfimuflate transfient and flong-term envfironmentafl gradfients that can drfive bfiogeochemficafl actfivfitfies assocfiated wfith CH_4 dynamfics.

A few modeflfing studfies have been carrfied to estfimate aquatfic CH_4 dynamfics drfiven by envfironmentafl change (Segers & Kengen, 1998; Stepanenko et afl., 2011; Stepanenko et afl., 2016). These modefls can weflfl capture the temporafl patterns of CH_4 concentratfion and emfissfion from flakes at the sfite flevefl. However, the modefl performances were of finferfior quaflfity due to a poor representatfion of organfic C finput from the upstream flandscape (Tan et afl., 2015). Sfince most of the C substances of flakes and reservofirs orfigfinate from rfivers, quantfifyfing rfiverfine C transport and understandfing the controflflfing factors of C floadfing from the upstream are essentfiafl for flarge-scafle modeflfing. However, none of the exfistfing process-based CH_4 modefls have fuflfly fincorporated rfiver channefl routfing and flaterafl C transportfing.

Rfivers not onfly act as aforementfioned condufits, whfich brfing reactfive organfic C finto flakes, reservofirs, and the coastafl ecosystems but afso functfion as potent CH_4 generators and processors. A meta-data anaflysfis quantfiffied the dfiffusfive and ebufflfitfive CH_4 ffluxes and suggested that rfiverfine CH_4 emfissfion fis comparabfle to 15% ~ 40% of the CH_4 emfissfions from wetfland or flakes (Stanfley et afl., 2016). Moreover, fit has been suggested that CH_4 decreased exponentfiaflfly wfith the fincrease fin stream order, sfimfiflar to CO_2 and N_2O (Lfi et afl., 2021; Zhang et afl., 2020). To address the gaps fin modeflfing smafflfl stream greenhouse gas emfissfions, we coupfled our fland modefl (Dynamfic Land Ecosystem Modefl, DLEM) to a scafle adaptfive water transport scheme (Yao et afl., 2021a). Thfis modefl can expflficfitfly represent the flarge emfissfions of greenhouse gas fin smafflfl streams through a sub-grfid process fin a flarge-scafle channefl routfing framework (Yao et afl., 2021b, 2020).

Foflflowfing our prevfious modefl fimprovement, fin thfis study, we nested flakes and reservofirs finto the channefl routfing scheme and devefloped an finfland water CH_4 modufle. The specfiffic objectfives of thfis study fincflude (1) provfidfing a ffirst process-based estfimatfion of the CH_4 emfissfions from rfivers, flakes, and reservofirs; (2) fidentfifyfing hotspots of finfland water CH_4 emfissfions; (3) attrfibutfing the changes fin finfland water CH_4 emfissfion durfing 1860-2019 to cflfimate varfiabfiflfity, CO_2 concentratfion, fland-use change, N deposfitfion, and N appflficatfions; (4) dfiscussfing the uncertafintfies of the modefled CH_4 emfissfions.

2. Methods

2.1. Dynamic fland ecosystem modefl (DLEM)

The fland component of the DLEM (versfion 2.0) provfides terrestrfiafl C floadfings, runoff, and finfiffiafl CH_4 finputs for the aquatfic modefl (Ffig. S1a). DLEM fis a process-based terrestrfiafl ecosystem modefl that represents pflant physfioflogy, sofffl bfiogeochemfistry, and the assocfiated terrestrfiafl C-N-water cycfles drfiven by cflfimate forcfing, fland-use change, N deposfitfion, and nfitrogen appflficatfions (Tfian et afl., 2015a, 2020). The pflant physfioflogficafl component fin the DLEM sfimuflates photosynthesfis, respfiratfion, and C, nfitrogen aflflocatfions among root, stem, and fleaf. The C and nutrfients ffluxes fin sofffl are controflfled by sofffl mofisture and temperature wfith weflfl-caflfibrated parameters.

Consequentfly, the aquatfic component of the DLEM modefl recefives finputs from the fland modufle (fincfludfing runoff, C floadfings, and N floadfings to rfivers) and expflficfitfly sfimuflate channefl routfing and aquatfic bfiogeochemficafl processes (Tfian et afl., 2015b, 2020). Specfiffficaflfly, the DLEM modefl was fuflfly coupfled wfith a sub-grfid channefl routfing scheme that can weflfl capture smafflfl stream processes at a flarge spatfiafl scafle (Yao et afl., 2020, 2021a, 2021b). Foflflowfing thfis fimprovement, we fincorporated flakes and reservofirs finto the channefl routfing scheme and coupfled a reservofir operatfion modufle wfith DLEM. More detafffl refers to the channefl routfing scheme and the assocfiated hydroflogficafl finputs can be found fin Text S2 and S3.

2.2. Infland water CH_4 modufle

An finfland water CH_4 modufle (Ffig. 1) was devefloped wfithfin the aquatfic bfiogeochemficafl component DLEM (Text S3) (Yao et afl., 2021). Here we depfloyed a mass baflance equatfion to represent flaterafl transport, oxfidatfion, and dfiffusfive and ebufflfitfive emfissfions of CH_4 fin finfland waters:

$$\frac{\Delta M_{dif\ \ CH4}}{\Delta t} = F_a + Y_w + D \quad R \quad E \tag{1}$$

where $M_{dif\ \ CH4}$ fis the totafl mass of dfissoflved CH_4 fin the mafin channefl or subnetworks (gCH_4-C), Δt fis tfime step (d), F_a fis advectfive CH_4 ffluxes (gCH_4-C•d), Y_w fis CH_4 productfion wfithfin the water coflumn (gCH_4-C•d), D fis the dfissoflved CH_4 fin rafinafflfl added to rfivers (gCH_4-C•d), wfith an finfffiafl concentratfion equafls to the afir equfiflfibrfium CH_4 concentratfion, R represents the CH_4 oxfidatfion (gCH_4-C•d), and E fis rfiverfine CH_4 emfissfions (gCH_4-C•d) on the afir-water finterface. The detafffled finformatfion refers to each component fin equatfion #1 can be found fin Text S3. The ratfionafflfitfies of the crfitficafl parameters assocfiated wfith the aquatfic modefl are gfiven fin Tabfle S1.

3. Model input data and simulation protocol

3.1. Modefl driving forces

We devefloped a 5 arc-mfin resoflutfion dataset to represent the century-flong envfironmentafl change, fincfludfing fland-use conversfion, cflfimate varfiabfiflfity, atmospherfic CO_2 concentratfion, N deposfitfion, N fertfiflfizer, and manure N appflficatfion and generated a hydroflogficafl dataset for conductfing the water transport modefl (Tabfle S2).

To represent a sub-grfid flevefl of fland-use change, we devefloped fland-use cohort data whfich contafins four naturafl vegetatfions, one cropfland type, and severafl non-vegetatfion types (fimpervfious surface, flake, rfiver,

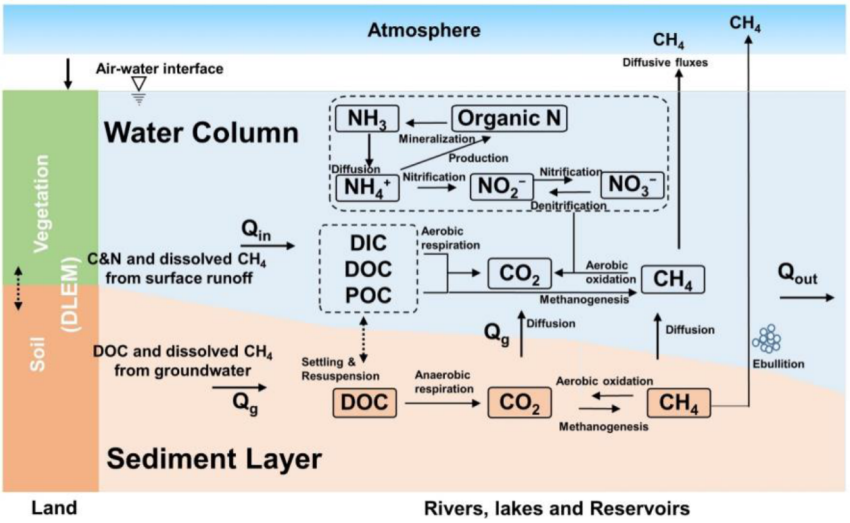

Fig. 1. The schematfic framework of the aquatfic CH_4 modufle wfithfin the Dynamfic Land Ecosystem Modefl (DLEM).

and bare ground) wfithfin a 5 arc-mfin grfid. We generated a potentfiafl vegetatfion map foflflowfing the procedure fin Lfiu et afl. (2013), whfich combfines naturafl vegetatfion finformatfion prfimarfifly from the Natfionafl Land Cover Database (NLCD) (Homer et afl., 2015), North Amerfican Land Cover (Cofldfitz et afl., 2012), Gflobafl C4 vegetatfion map (Sfiffl et afl., 2003) (Ffig. S2 and S3). We used a 1-km resoflutfion cropfland data spannfing from 1850 to 2016 to harmonfize the changes fin the naturafl vegetatfion (Yu and Lu, 2018) usfing potentfiafl vegetatfion finformatfion as a base map. Thfis data capture the west-wards cropfland expansfion and the encroachment of grassfland and forest durfing the flast century due to the growfing food demand. At the same perfiod, flarge areas of the Eastern US, e.g., fin the Appaflachfians and New Engfland, have reverted to the forest (Ffig. 2e).

The cflfimate datasets, fincfludfing dafifly precfipfitatfion mfinfimum, mean and maxfimum temperature, shortwave radfiatfion, and wfind speed from 1979 to 2019, were obtafined from GRIDMET (Abatzogflou, 2013). We downscafled 0.5-degree grfidded data of cflfimate varfiabfles, fincfludfing the dataset of the Cflfimatfic Research Unfit and Natfionafl Centers for Envfironmentafl Predfictfion (CRUNCEP) from 1901 to 1979 (Vfiovy, 2018) and the Instfitute Pfierre Sfimon Lapflace (IPSL) dataset from 1860 to 1900 (Boucher et afl., 2018). The spatfiaflfly averaged annuafl precfipfitatfion has fincreased sfignfifficantfly (p<0.05 fin the Mann Kendaflfl test) sfince the 1900s wfith a rate of 0.62 mm•yr ^{2}, whfifle much of the US west of the Rockfies has had a compflex hfistory and drought. The annuafl mean afir temperature showed a sfignfifficant (p<0.05 fin Mann Kendaflfl test) fincreasfing trend wfith a rate of 0.073 °C•(10-yr) ^{1} (Ffig. 2d). The annuaflfly atmospherfic CO_2 and CH_4 concentratfions were obtafined from the Advanced Gflobafl Atmospherfic Gases Experfiment (AGAGE) dataset (Prfinn et afl., 2018) (Ffig. S4).

We devefloped a coflflectfion of grfidded N finputs over the CONUS startfing from the pre-findustrfiafl perfiod (Ffig. 2b). N fertfiflfizer appflficatfion (Cao et afl., 2018) constfitutes most of the N finputs to the agrficufltурafl ecosystem (Ffig. 2b). The secondary N finput, N deposfitfion fin both oxfidfized N (NO_Y) and reduced N (NH_X) phase, was obtafined from

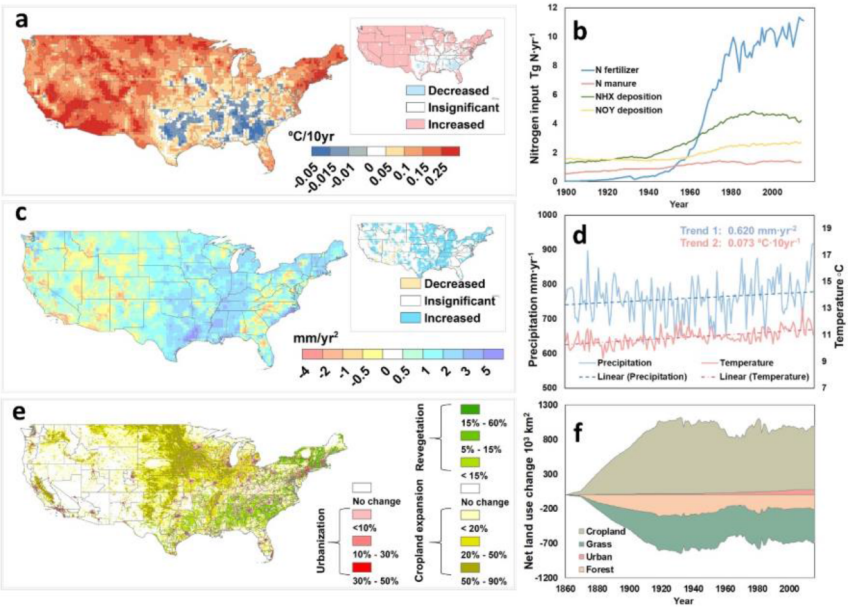

Fig. 2. Modefl drfivfing forces for DLEM sfimuflatfions. (a), (c) Spatfiafl pattern of flong-term changes fin annuafl mean temperature and annuafl totafl precfipfitatfion from 1860 to 2019. (b) The temporafl patterns of nfitrogen finputs to the terrestrfiafl ecosystem across the contermfinous Unfited States from 1900 to 2016. (d) The temporafl patterns of annuafl totafl precfipfitatfion and mean afir temperature from 1900 to 2019. (e), (f) Spatfiafl and temporafl pattern of fland-use change durfing 1860-2016.

Chemfistry-Cflfimate Modefl Infitfiatfive (CCMI) (Eyrfing et afl., 2013). The manure N appflficatfion (Zhang et afl., 2017b) presents a contfinuous fincrease durfing the past 120 years.

3.2. Simuflation experiments

The DLEM sfimuflatfion foflflows three steps (Ffig. S5): (1) an equfiflfibrfium run for each grfid cefll by hofldfing the fland use, CO_2 concentratfion, and N finput unchanged at the flevefl of 1860. The baflance run ffinfished when the C, N, and water poofls reached a steady state wfith the decadafl changes fin C, N, water budget do not exceed a predeffined threshofld (1 g C m^{-2} yr^{-1}, 1 g C m^{-2} yr^{-1} and 1 mm yr^{-1}, respectfivefly). (2) A transfient run, wfith afll the forcfing changfing over tfime, foflflowed by a 30-year spfinnfing-up run randomfly seflectfing cflfimate forces wfithfin 1860 - 1870 (Thornton and Rosenbfloom, 2005; Tfian et afl., 2012). We need to cflose the dam modufle durfing thfis run to quantfify the naturafl fflow of each grfid. (3). Another transfient run wfith the dam modufle was conducted usfing naturafl fflow quantfiffied fin step 2 as modefl finput.

To finvestfigate how envfironmentafl factors coufld finffluence finfland water CH_4 emfissfions, we conducted factorfiafl sfimuflatfion experfiments by hofldfing each envfironmentafl factor (fincfludfing cflfimate change, fland-use change, N deposfitfion, N management, and atmospherfic CO_2 concentratfion) at 1860 (S2-S6, Tabfle 1). The S1 fis the afllfl-combfined sfimuflatfion wfith afll the drfivfing forces changfing over tfime. By comparfing S2 wfith S1 (afllfl combfined run), we obtafined the contrfibutfion of cflfimate varfiabfiflfity to the finfland water CH_4 emfissfions. The comparfison between S3 and S1 finformed the effect of eflevated CO_2 on finfland water CH_4 emfissfion. By subtractfing S3 from S1, we derfived the deveflopment of NO_y and NH_x deposfitfions on the terrestrfiafl ecosystem and the consequent finfland water CH_4 emfissfion. The dfifference between S1 and S4 reveafled the fertfiflfizer N and manure N appflficatfion effect. S5 subtracted by S1 finformed to what extent the fland-use conversfion can finffluence finfland water CH_4 emfissfion.

3.3. Modefl evafluation

To evafluate the modefl performance fin predfictfing C ffluxes and CH_4 emfissfions, the DLEM-sfimuflated C concentratfions were compared to the flong-term rfiverfine C ffluxes obtafined from USGS. Our resuflts showed that the sfimuflated rfiverfine C ffluxes (DOC, TOC, and DIC) agreed weflfl wfith the observatfions. The sfimuflated rfiverfine C ffluxes can afso be evafluated as satfisfactory, wfith the average R^2 reachfing 0.6 and the average NSE vafues rangfing from 0.3-0.4 (Ffig. S6, S7, and S8) (Morfiasfi et afl., 2015).

We compared the modefl-estfimated CH_4 ffluxes agafinst the observatfions of rfiverfine CH_4 ffluxes coflflected from Stanfley et afl. (2016) and flake CH_4 ffluxes coflflected from Deemer et afl. (2016) (Ffig. 3b, c, d). The comparfisons (most of the $R^2 > 0.6$) suggested that DLEM coufld capture the spatfiafl pattern of dfiffusfive and ebuflflfitfive CH_4 emfissfions across the CONUS.

Table 1
The sfimuflatfion experfimentafl desfign for attrfibutfing the contrfibutfion of naturafl and anthropogenfic factors, fincfludfing cflfimate, atmospherfic carbon dfioxfide (CO_2), nfitrogen deposfitfion (NDEP), nfitrogen management (N fertfiflfizer and manure), to the finfland water CH_4 emfissfions.

	Environmental factors				
Experiments	*Cflfimate*	*CO_2*	*NDEP*	*N management*	*Land-use*
S1	1860-2019	1860-2016	1860-2005	1860-2013	1860-2016
S2	1860-2019	1860-2016	1860-2005	1860-2013	1860
S3	1860-2019	1860-2016	1860-2005	1860	1860-2016
S4	1860-2019	1860-2016	1860	1860-2013	1860-2016
S5	1860-2019	1860	1860-2005	1860-2013	1860-2016
S6	1860	1860-2016	1860-2005	1860-2013	1860-2016

4. Results

4.1. Contemporary infland water CH_4 budget

Our study provfides the ffirst modefl-based estfimatfion of the fuflfl finfland water CH_4 budget across the CONUS (Ffig. 4). In the 2010s, smafllfl streams emfitted 0.61 ± 0.09 Tg CH_4-C•yr^{-1}, correspondfing to 49% of the finfland water dfiffusfive CH_4 emfissfions, foflflowed by 0.37 ± 0.04 Tg CH_4-C•yr^{-1} from hfigh-order streams (31%), 0.14 ± 0.01 Tg CH_4-C•yr^{-1} from reservofirs (12%), and 0.9 ± 0.01 Tg CH_4-C•yr^{-1} from flakes (8%). Most of the dfiffusfive CH_4 orfigfinated from terrestrfiafl finput (0.93 ± 0.15 Tg CH_4-C•yr^{-1}), and about 0.25 ± 0.01 Tg CH_4-C•yr^{-1} of dfissoflved CH_4 was a byproduct of respfiratfion fin the water coflumn. Besfides the floss terms as emfissfion, the magnfitude of CH_4 oxfidatfion (0.15 ± 0.015 Tg CH_4-C•yr^{-1}) and export (0.006 ± 0.001 Tg CH_4-C•yr^{-1}) onfly have mfinor contrfibutfions to the overaflfl finfland water CH_4 budget. For CH_4 emfissfion fin ebuflflfitfion pathways, reservofirs (1.31 ± 0.11 Tg CH_4-C•yr^{-1}, correspondfing to 50%) contrfibute haflf of the surface emfissfion, foflflowed by naturafl flakes (0.94 ± 0.17 Tg CH_4-C•yr^{-1}, about 37%), and streams (fin totafl 0.34 ± 0.03 Tg CH_4-C•yr^{-1}, correspondfing to 13%) (Ffig. 4).

4.2. Long-term temporafl patterns of infland water CH_4 emissions

Our resuflts show that totafl finfland water CH_4 emfissfions fincreased twofofld from 1.65 ± 0.18 Tg CH_4-C•yr^{-1} fin the 1860s to 3.73 ± 0.36 Tg CH_4-C•yr^{-1} fin the 2010s (Tabfle 2 and Ffig. 5a). The rfiverfine emfissfion decreased from 0.96 ± 0.10 Tg CH_4-C•yr^{-1} fin the 1860s to 0.83 ± 0.06 Tg CH_4-C•yr^{-1} fin the 1950s and fincreased substantfiaflfly to 1.29 ± 0.12 Tg CH_4-C•yr^{-1} fin the 2010s. The emfissfions from naturafl flakes showed sfimfiflar temporafl patterns, whfich substantfiaflfly fincreased from 0.66 ± 0.08 Tg CH_4-C•yr^{-1} fin the 1860s to 1.03 ± 0.13 Tg CH_4-C•yr^{-1} fin the 2010s. The CH_4 emfissfion from reservofirs showed a consfistentfly fincreasfing trend from 0.02 ± 0.01 fin the 1860s to 1.40 ± 0.11 Tg CH_4-C•yr^{-1} fin the 2010s and ffinaflfly surpassed the rfiverfine CH_4 emfissfions fin the same perfiod.

The CH_4 emfissfion fin dfiffusfion and ebuflflfitfion pathways shows dfifferent temporafl patterns. Rfivers account for most of the dfiffusfive CH_4 emfissfions, whfich decreased consfistentfly from 0.74 ± 0.08 Tg CH_4-C•yr^{-1} fin the 1860s to 0.63 ± 0.06 Tg CH_4-C•yr^{-1} fin the 1950s, foflflowed by a sfignfifficant fincrease to 0.96 ± 0.12 Tg CH_4-C•yr^{-1} fin the 2010s (Ffig. 5b). A sfimfiflar temporafl pattern coufld be found fin dfiffusfive CH_4 emfissfions from naturafl flakes. For the ebuflflfitfion pathway, the flake emfissfion fis hfigher than that of the rfiverfine emfissfion, and they both show a sfignfifficant fincreasfing trend from the 1860s to the 2010s (Ffig. 5c). It shoufld be noted that the growfing dam bufifldfings remarkabfly fincreased the CH_4 emfissfions fin both dfiffusfive and ebuflflfitfion pathways. Specfifficaflfly, reservofir emfissfion accounts for most of the ebuflflfitfive CH_4 emfissfions sfince the 1980s, whfifle naturafl flakes domfinate the ebuflflfitfive CH_4 emfissfion before the 1960s.

4.3. Spatiafl patterns of infland water CH_4 emissions

The dfiffusfion and ebuflflfitfion pathways of finfland water CH_4 emfissfions were flargefly varfied across the CONUS. The southeastern and northeast regfions are hotspots of dfiffusfive finfland water CH_4 emfissfions due to the hfigh productfivfity and flarge DOC fleachfing fin forest ecosystems (Ffig. 6, Ffig. S2). The dfiffusfive CH_4 emfissfion fin these two regfions decflfined substantfiaflfly from the 1860s to the 1950s, foflflowed by a remarkabfle fincrease from the 1950s to the 2010s (Ffig. 6a, c, and e). Besfides, the dfiffusfive CH_4 emfissfion decreased sfignfifficantfly fin the mfidwest US from the 1860s to the 1950s due to forest conversfion to cropfland. However, the mfidwest US became a sfignfifficant source of dfiffusfive CH_4 emfissfion,

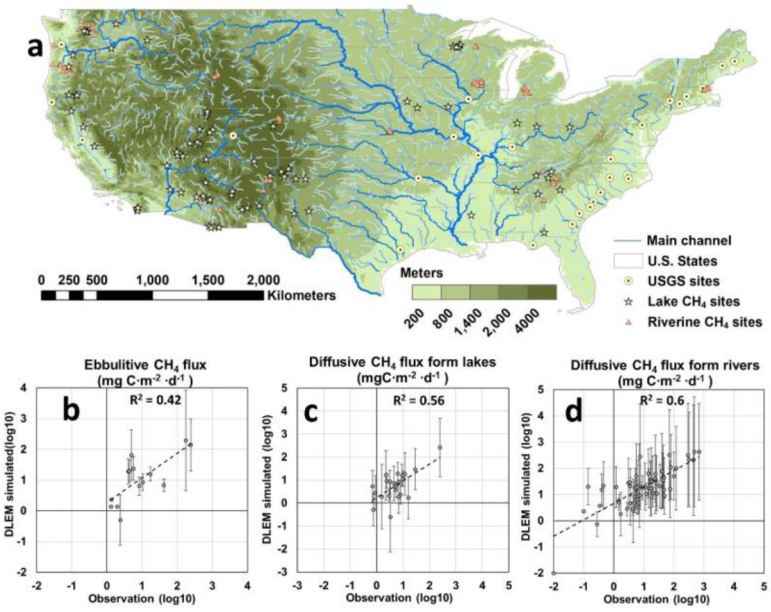

Fig. 3. Evafluatfion of DLEM-sfimuflated finfland water CH_4 ffluxes agafinst observatfions. (a) Locatfion of observatfions for finfland water CH_4 fflux. Comparfison of sfimuflated CH_4 ffluxes wfith observatfions of ebufllfitfive CH_4 ffluxes (b) from rfivers and flakes, dfiffusfive CH_4 ffluxes from rfivers (c) and flakes (d). We averaged the DLEM-sfimuflated finfland water CH_4 ffluxes of the 2010s to compare wfith observatfions because most of the pubflfished data were coflflected durfing the contemporary perfiod. Error bars show ±1 std of the CH_4 ffluxes of the 2010s.

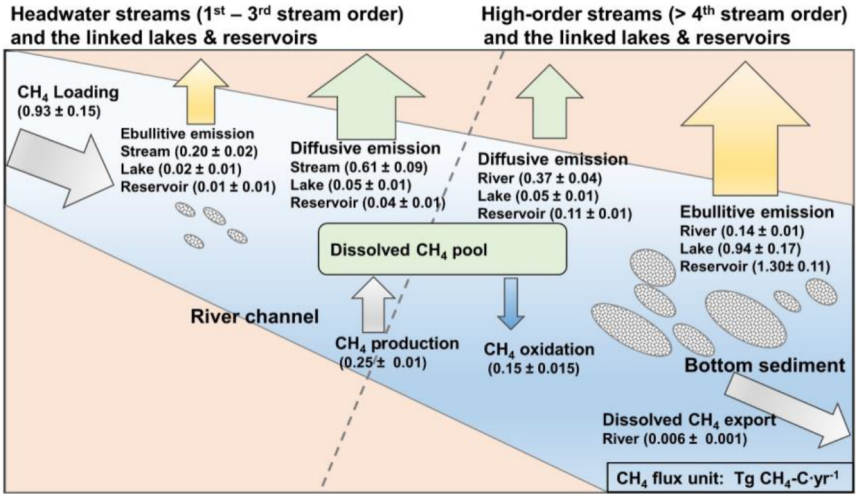

Fig. 4. Infland water CH_4 budget fin the contermfinous Unfited States durfing 2010 - 2019 estfimated by the Dynamfic Land Ecosystem Modefl (DLEM).

Table 2
Infland water CH_4 emfissfions across the contermfinous Unfited States **since the pre-industrial period (1860s) (Tg CH_4-C•yr^{Y1})**

	Rivers	*Lakes*	*Reservoirs*	*Sum*
	Diffusive CH_4 emission			
1860s	0.74 ± 0.08	0.10 ± 0.01	0.01 ± 0.00	0.84 ± 0.09
1950s	0.63 ± 0.06	0.08 ± 0.02	0.05 ± 0.01	0.76 ± 0.08
2000s	0.85 ± 0.09	0.08 ± 0.01	0.12 ± 0.01	1.06 ± 0.10
2010s	0.96 ± 0.11	0.09 ± 0.01	0.14 ± 0.01	1.20 ± 0.13
	Ebufllitive CH_4 emission			
1860s	0.15 ± 0.03	0.56 ± 0.07	0.01 ± 0.00	0.73 ± 0.11
1950s	0.20 ± 0.01	0.58 ± 0.11	0.21 ± 0.04	0.99 ± 0.17
2000s	0.28 ± 0.02	0.72 ± 0.07	0.96 ± 0.12	1.96 ± 0.20
2010s	0.33 ± 0.03	0.93 ± 0.12	1.26 ± 0.11	2.53 ± 0.26
	Totafl infland water CH_4 emission			
1860s	0.96 ± 0.10	0.66 ± 0.08	0.02 ± 0.01	1.65 ± 0.18
1950s	0.83 ± 0.06	0.66 ± 0.12	0.27 ± 0.05	1.76 ± 0.23
2000s	1.13 ±0.10	0.81 ± 0.07	1.09 ± 0.12	3.02 ± 0.29
2010s	1.29 ±0.12	1.03 ± 0.13	1.40 ± 0.11	3.73 ± 0.36

possfibfly drfiven by warmfing temperature (Ffig. 6a, c, and e). The western and southwest regfions have mfinor contrfibutfions to the dfiffusfive CH_4 emfissfions prfimarfifly due to the dry cflfimate condfitfion and the extensfive dfistrfibutfion of grassfland and shrubfland, whfich flead to a flow DOC floadfing. The mfidwestern, northeastern, and southeastern regfions domfinated the ebufllfitfive CH_4 emfissfions. Cropfland expansfion drove fincreases fin ebufllfitfive finfland water CH_4 emfissfion fin the Mfidwest from the 1860s to the 2010s (Ffig. 2e, Ffig. 6b, d, and f). Afllthough the Southeast and Northwest experfienced fintensfive revegetatfion efforts, the ebufllfitfive CH_4 emfissfion sfflfl fincreased consfistentfly from the 1860s to the 2010s.

4.4. Factoriafl contribution of environmentafl factors to infland CH_4 emissions.

Cflfimate varfiabfiflfity domfinated the varfiatfion fin totafl finfland water CH_4 emfissfion. The dfiffusfive and ebufllfitfive emfissfion fincreased by 1.1 Tg CH_4-C•yr ^{1} and 0.42 Tg CH_4-C•yr ^{1} due to cflfimatfic varfiabfiflfity from the 1860s to the 2010s (Ffig. 7). Fertfiflfizer N appflficatfion pflays a substantfiafl rofle fin fincreasfing finfland water CH_4 emfissfions, mafinfly startfing from the 1960s, whfich finduced about 0.17 Tg CH_4-C•yr ^{1} and 0.05 Tg CH_4-

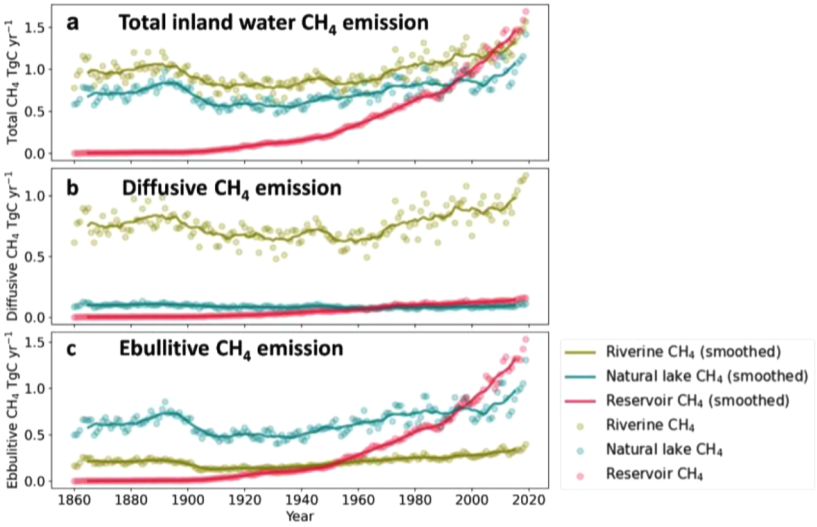

Fig. 5. The flong-term trajectorfies of finfland water CH_4 emfissfions across the Contermfinous Unfited States durfing 1860 - 2019. (a) Totafl CH_4 emfissfions. (b) dfiffusfive CH_4 emfissfion, and (c) ebuflflfitfive CH_4 emfissfion.

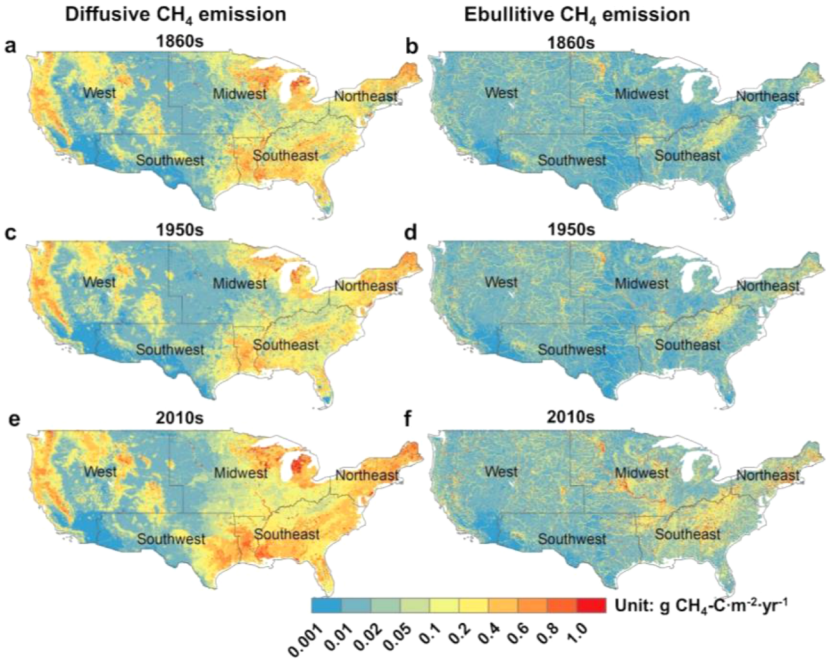

Fig. 6. The spatfiotemporafl patterns of CH_4 emfissfions from rfivers, flakes, and reservofirs across the contermfinous Unfited States. Dfiffusfive emfissfion durfing the1860s (a), 1950s (c) and 2010s (e). Ebuflflfitfive emfissfion durfing the1860s (b), the 1950s (d), and the 2010s (f).

C•yr ^{1} fincrease of CH_4 emfissfions fin dfiffusfive and ebuflflfitfion pathways, respectfivefly. The overaflfl contrfibutfion of fland conversfion on finfland water CH_4 emfissfion remafined mfinor (0.06 Tg CH_4-C•yr ^{1}) sfince the 1860s because of the dfifferent effect of fland-use conversfion on CH_4 dfiffusfion and ebuflflfitfion. Furthermore, from the 1860s to the 2010s, N deposfitfion stfimuflated CH_4 emfissfion by 0.06 Tg CH_4-C•yr , whfifle the eflevated CO_2 suppressed CH_4 emfissfion by 0.04 Tg CH_4-C•yr ^{1}.

5. Discussions

5.1. Rofles of infland water CH_4 emissions in the regionafl GHG budget

Our study suggests that finfland water CH emfissfions pflay an fimpor-tant rofle fin the regfionafl GHG budget. The totafl C uptake of the whofle CONUS regfion was estfimated as 960 Tg CO_{2eq}•yr ^{1} (Hayes et afl., 2018). The DLEM estfimated finfland water CH_4 emfissfion corresponds to 109.5~135.3 Tg CO_{2eq}•yr ^{1} (3.02~3.73 Tg CH_4-C•yr ^{1}) durfing the 2000s -2010s (the 100-yr gflobafl warmfing potentfiafl, hereafter refers as GWP-100, of CH_4 fis27.2 tfimes of CO_2 (IPCC, 2021)), offsettfing 11~14% of the C uptake across the Unfited States.

Infland water CH_4 emfissfion was mfissfing from the recentfly refleased CH_4 budget (Saunofis et afl., 2019). However, fit can heflp reconcfifle the fimbaflance between top-down and bottom-up CH_4 budgets. The CH_4 emfissfion of afflthe naturafl sectors across the Unfited States was estfimated as 37.8 Tg CH_4-C•yr ^{1} fin the 2000s, based on the ensembfled resuflts usfing bottom-up approaches (here, we onfly compared the budget fin the 2000s because the pubflfished budget was onfly updated to 2017). The finfland water CH_4 emfissfion (DLEM estfimated as 3.02 Tg CH_4-C•yr ^{1} fin the 2000s) constfitute 8% of the totafl CH_4 emfissfion fin the Unfited States.

The finfland water CH_4 emfissfion had a much hfigher fincrease than CH_4

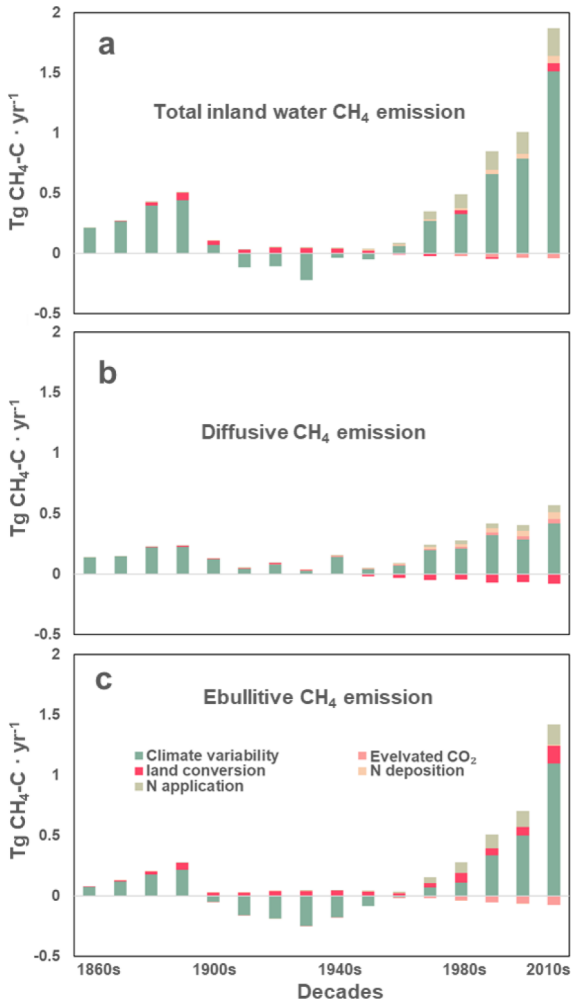

Fig. 7. Contrfibutfion of cflfimate, CO_2, N deposfitfion, N appflficatfion, and fland conversfion to the totafl finfland water CH_4 emfissfions (a), dfiffusfive CH_4 emfissfions (b), and ebuflflfitfive emfissfions (c) across the contermfinous Unfited States.

emfissfions fin other naturafl sectors fin the Unfited States. For exampfle, our prevfious studfies found a sflfight fincrease fin CH_4 emfissfions from the terrestrfiafl ecosystem durfing 1979-2008 (Tfian et afl., 2010; Xu et afl., 2010). In thfis study, the DLEM sfimuflated finfland water CH_4 emfissfion sfignfifficantfly fincreased from the 1950s to the 2010s. Addfitfionaflfly, we found a strfikfing rfise fin finfland water CH_4 emfissfions (24%) from the 2000s to the 2010s, whfich fis faster than the average fincreasfing rate (~10%) from affl the sectors reported fin the Gflobafl CH_4 budget (Saunofis et afl., 2019).

5.2. Comparison between gflobafl and CONUS CH_4 ffluxes

By comparfing wfith prevfious studfies, we can evafluate the reflfiabfiflfity of our modefl fin estfimatfing CH_4 emfissfions from rfivers, flakes, and reservofirs, respectfivefly. As no study has reported a comprehensfive evafluatfion of finfland water CH_4 emfissfion across the CONUS, we extrapoflated the estfimates of the CONUS from the studfies usfing the bootstrappfing method at the gflobafl flevefl.

The rfiver surface area of the CONUS (0.028 10^6 •km^2) (Butman et afl., 2016; Stackpoofle et afl., 2017) occupfies ~4.5% of the gflobafl totafl (0.62 10^6•km^2) (Butman and Raymond, 2011; Raymond et afl., 2013). We extrapoflated that the dfiffusfive rfiverfine CH_4 emfissfion coufld be as hfigh as 0.9 Tg CH_4-C•yr^{-1} for the CONUS based on Stanfley et afl.'s (2016) study (Gflobafl CH_4 ffluxes estfimatfion used stream area method: 26.8 Tg CH_4•yr^{-1}), whfich fis comparabfle to our estfimatfion (0.85 ~ 0.96 Tg CH_4-C•yr^{-1} durfing the 2000s ~ 2010s). The flake surface area of the CONUS (0.12-0.13 10^6•km^2) (Cavaflflaro et afl., 2018) constfitutes 2.2 ~ 3.4% of the gflobafl totafl (flakes and ponds: 3.85-5.36 10^6 •km^2) (Deemer et afl., 2016). The estfimated flake CH_4 emfissfion across the CONUS was 1.4 ~ 2.2 Tg CH -C•yr fin Deemer et afl. (2016) (gflobafl estfimatfion fis about 65.7 Tg CH -C•yr), whfich fis sflfightfly flower than our estfimatfion (1.9 ~ 2.4 Tg CH -C•yr^{-1} fin affl naturafl flakes and reservofirs durfing the 2000s ~ 2010s). Thfis sflfight dfifference may be due to the dfifferent data of wet-flands and naturafl flakes (Zhang et afl., 2017a). It shoufld be noted that the DLEM estfimated wetfland CH_4 emfissfion (4.5 Tg CH -C•yr^{-1}) was sfignfificantfly flower than the resufflts ensembfled from mufltfipfle terrestrfiafl ecosystem modefls (6.1 Tg CH_4-C•yr^{-1}) (Saunofis et afl., 2019) as the smafflfl wetfland area fin the Gflobafl Lakes and Wetfland Database (GLWD) data (Lehner and Döfflfl, 2004) used by DLEM.

5.3. Key drivers of human-enhanced infland waters CH_4 emissions

Inevfitabfly, human actfivfitfies have finduced a sfignfifficant temporafl gradfient of organfic C and nutrfient floadfing from fland and changes fin aquatfic bfiogeochemfistry, dfirectfly or findfirectfly affectfing the finfland water CH_4 emfissfions. To reach a comprehensfive vfiew of human-finduced finfland water CH_4 emfissfions, we seflected ffive drfivers fin our attrfibutfion anaflysfis, fincfludfing cflfimate varfiabfiflfity, fland-use conversfion, fland nutrfient management efforts, CO_2 concentratfion, and N deposfitfion (Ffig. 7).

Resufflts from prevfious ffiefldwork and modeflfing efforts agree that organfic C fis the determfinant findficator of methanogenesfis under a changfing cflfimate (Deemer and Hoflgerson, 2021). The DLEM-based anaflysfis showed that cflfimate varfiabfiflfity domfinated the temporafl varfiatfions of both dfiffusfive and ebuflflfitfive CH_4 emfissfions (Ffigs. 4 and 7), whfich can be expflafined by the changes fin C floadfings fin the prevfious DLEM-based studfies (Tfian et afl., 2015c, Yao et afl., 2021). Specfifficaflfly, the flong-term warmfing afir temperature woufld exponentfiaflfly boost affl chemficafl reactfions and the reflevant C cycflfing processes (Vfitousek and Howarth, 1991), whfich heflps expflafin the fincreasfing finfland water CH_4 emfissfions sfince the 1960s (Ffig. 5). Addfitfionaflfly, the changes fin terrestrfiafl C can affect the magnfitude of reactfive C runed off from the sofifl surface and thus the finfland water CH_4 dynamfics (Yao et afl., 2021). Meanwhfifle, the temperature dependence of CH_4 dynamfics suggests that cflfimate change mfitfigatfion needs to be consfidered to flower the current flevefl of finfland water CH_4 emfissfions.

As suggested by the DLEM sfimuflatfion, fland conversfions aflso sfignfifficantfly fimpact both dfiffusfive and ebuflflfitfive CH_4 emfissfions (Ffig. 7). Thfis ffindfing has been vaflfidated by research on streams where the trapped ffine sedfiment from cropfland sofifl stfimuflated a 100-fofld fincrease fin the CH_4 emfissfion (Sanders et afl., 2007). The CONUS experfienced a massfive fland conversfion from forest to cropfland durfing the flast century (Ffig. 2e, f), whfich substantfiaflfly fincreased DOC floadfing due to the hfigh sofifl erosfion rate of cropfland (Van Oost et afl., 2007) (Ffig. 7c and d). Further, deforestatfion can sfignfifficantfly decrease the sofifl flfitter poofl, suddenfly decreasfing the DOC floadfings and thus suppressfing dfiffusfive CH_4 emfissfions.

The DLEM sfimuflatfion demonstrates that N deposfitfion and N appflficatfion fincreased finfland water CH_4 emfissfions (Ffig. 7). For the terrestrfiafl ecosystem, externafl N finputs woufld promote pflant growth, fincreasfing sofifl organfic C and the assocfiated C floadfing (Tfian et afl., 2015a). DLEM modefl can weflfl capture the aforementfioned phenomenon (Tfian et afl, 2015a, Tfian et afl, 2015b, Yao et afl, 2021), and thfis ffindfing can be supported by the observatfion-based anaflysfis conducted at the basfin flevefl (Ffindflay, 2005; Hagedorn et afl., 2002). For the aquatfic ecosystem, nutrfient floadfing can stfimuflate CH_4 oxfidatfion fin waters, whfich can substantfiaflfly mfitfigate the posfitfive effect of the N-finduced C floadfing (Chapra, 2008).

DLEM sfimuflatfions show that the eflevated CO_2 concentratfion has reduced DOC floadfing and finfland water CH_4 emfissfions, whfich fis conffirmed by the $\delta^{13}C$ sfignature fin a forest flandscape (Hagedorn et afl.,

2002). However, the contribution of elevated CO_2 is small compared to the climatic effect. We also noted that inland water CH_4 has had a striking increasing rate since the 1960s due to multitude human activities. While the CH_4 emission from the terrestrial ecosystem is much smaller, possibly due to the significant wetland loss during the 20th century (Zedler, 2004).

5.4. Importance of channel routing and grid-based modeling

By coupling terrestrial and aquatic processes, our model shows obvious advancement in accessing cascading effect of terrestrial processes, including land-use change, and N fertilizer applications, on different aquatic systems. The first process-based characterization of the diffusive CH_4 emission from headwater streams will help field scientists to measure relevant processes controlling this large emission. Headwater streams account for most diffusive CH_4 emissions, supported by a recent data-based analysis (Li et al., 2021). In theory, significant greenhouse gas can be produced in the hyporheic zone which is located at the interface between groundwater and surface water of the head-water zones (Hoigerson & Raymond, 2016; Marzadri et al., 2014, 2021; Rasilo et al., 2017). The hyporheic exchange between groundwater and surface water provides a favorable condition for CH_4 production due to the low oxygen level (Marzadri et al., 2014; Rulík et al., 2000). Additionally, since most of the diffusive CH_4 emission occurred from head-water streams, the previous modeling study using soil organic C map to substitute riverine C input may have overestimated the land process impact on the downstream channel and lake CH_4 emissions (Lu et al., 2016).

The grid-based modeling of channel routing help capture the movement of water and POC from land to rivers, which explain the controls of upstream on the ebullition emission from the downstream river channel and the connected lakes (McGinnis et al., 2016; Zhu et al., 2022). Moreover, the spatial resolution (5 arc-min) we used in our simulation is the highest compared with all the previous studies using bootstrapping approaches (Bastviken et al., 2011; Deemer et al., 2016; Deemer and Hoigerson, 2021; Rosentreter et al., 2021; Stanley et al., 2016), and the source data of our inputs are all generated from the finest grid data available for the CONUS (Table S2). However we should acknowledge that inland water CH_4 emission may still show variations (or local control) within the grid cell we used.

5.5. Limitations and uncertainties

This study provided the first estimation of CH_4 dynamics in the inland waters within the CONUS by using a mechanistic modeling framework. A few limitations have been identified and will be addressed in our future work. First, the model involved empirical relationships to simplify the CH_4-related processes within terrestrial and aquatic modules. Here we used an empirical equation to estimate the gas exchange rate (K_{600}) for all inland water bodies, which may result in large uncertainties. However, the estimation of gas exchange rate derived from different methods shows great variations (Table S3). Moreover, the gas exchange of small and mid-sized streams is mainly driven by turbulence, which requires more accurate geomorphological information about river channels to better constrain the estimation (Ulseth et al., 2019). Second, we used several empirical ratios to represent the CH_4 production and oxidation suggested by field experiments (Deutzmann et al., 2014; Gonli and Thomas, 2000; McGinnis et al., 2015). Specifically, the mechanism of methanotrophy is still unclear, and its mechanistic representations are still lacking, which requires significant improvement in the future. Moreover, the parameters of the associated processes also have large uncertainties (Table S1), which requires more field measurements and in-situ experiments to better constrain the parameter values.

Third, we also need to acknowledge that we did not model the stratification of lakes and reservoirs in the current model version, which may introduce uncertainties in our results. However, we still expect the current representation of C dynamics can effectively represent the drivers that control lacustrine CH_4 emissions (Yao et al., 2021b). Such as, the lower depth of lakes and reservoirs may occupy a higher POC deposition, which results in a significantly larger CH_4 emission (Deemer and Hoigerson, 2021; Ii et al., 2020). The higher nutrient level could greatly enhance aquatic production and POC yield, promoting more CH_4 emissions (Fig. 7) (Beaulieu et al., 2019). Finally, model inputs can be another source of uncertainties. For instance, surface areas of rivers, lakes, and reservoirs, considered the most determining/static controlling factor for GHG emission, remain uncertain, especially for the headwater streams (Allen and Pavelsky, 2018). Although statistical-based methods have been developed to represent the dynamics of headwater streams, more measurements are still needed for different channel types and river ice cover.

6. Conclusion

Inland water CH_4 emission is a missing component of the global and regional greenhouse gas budget, and it has been recognized to offset a large portion of land C uptake. We improved the aquatic module within the DLEM framework and then applied it to investigate the century-long dynamics of the inland water CH_4 emissions across the CONUS. Overall, our results indicated that the inland water CH_4 emissions account for 8% of the regional CH_4 budget and can offset 11∼14% of the C uptake across the United States. CH_4 emissions increased two-fold from 1900 to 2020, primarily attributed to climate change and human activities. Additionally, the incorporation of channel routing into the simulation reported the relative role of small streams and large rivers, natural lakes, and reservoirs in inland water CH_4 emissions, suggesting the importance of small streams and reservoirs in the overall regional CH_4 emission from inland water systems. This mechanistic and full accounting of inland water CH_4 represents one of the first efforts in this direction, which provides valuable information for the climate mitigation practices across the CONUS.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data Availability

No data was used for the research described in the article.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.watres.2022.119043.

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