Science of the Total Environment 917 (2024) 170560

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Historical trends and drivers of the laterally transported terrestrial dissolved organic carbon to river systems

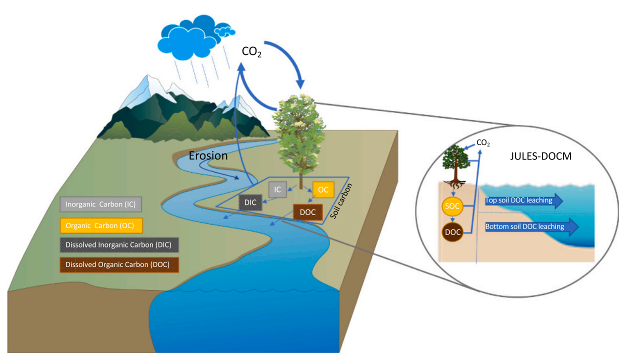
Mahdi (Andre) Nakhavali[a,*], Ronny Lauerwald[b], Pierre Regnier[c], Pierre Friedlingstein[d,e]

[a] International Institute for Applied Systems Analysis (IIASA), Laxenburg, Austria
[b] Université Paris-Saclay, INRAE, AgroParisTech, UMR ECOSYS, 78850 Thiverval-Grignon, France
[c] Biogeochemistry and Modelling of the Earth System, Department Geoscience, Environment and Society, Université Libre de Bruxelles, Bruxelles, Belgium
[d] LMD/IPSL, ENS, PSL Université, École Polytechnique, Institut Polytechnique de Paris, Sorbonne Université, CNRS, Paris, France
[e] University of Exeter, College of Engineering, Mathematics and Physical Sciences, Exeter EX4 4QE, UK

HIGHLIGHTS

- Historical DOC leaching trends highlight CO2 and climate influence.
- Terrestrial DOC inputs rose 17 % to 292 Tg C yr-1 due to CO2 fertilization.
- Tropics are primary DOC exporters, but boreal growth rates surpass.
- Land use change has limited impact on DOC leaching trajectory.

GRAPHICAL ABSTRACT



ARTICLE INFO

Editor: Ashantha Goonetilleke

Keywords:
Dissolved organic carbon (DOC)
Terrestrial carbon flux
Historical carbon attribution
Climate-driven changes
CO2 fertilization
Soil-to-River carbon transfer

ABSTRACT

Dissolved organic carbon (DOC) represents a critical component of terrestrial carbon (C) cycling and is a key contributor to the carbon flux between land and aquatic systems. Historically, the quantification of environmental factors influencing DOC leaching has been underexplored, with a predominant focus on land use changes as the main driver. In this study, the process-based terrestrial ecosystem model JULES-DOCM was utilized to simulate the spatiotemporal patterns of DOC leaching into the global river network from 1860 to 2010. This study reveals a 17 % increment in DOC leaching to rivers, reaching 292 Tg C yr^{-1} by 2010, with atmospheric CO_2 fertilization identified as the primary controlling factor, significantly enhancing DOC production and leaching following increased vegetation productivity and soil carbon stocks. To specifically quantify the contribution of CO_2 fertilization, a factorial simulation approach was employed that isolated the effects of CO_2 from other potential drivers of change.

The research highlights distinct regional responses. While globally CO2 fertilization is the dominant factor, in boreal regions, climate change markedly influences DOC dynamics, at times exceeding the impact of CO_2. Temperate and sub-tropical areas exhibit similar trends in DOC leaching, largely controlled by CO_2 fertilization, while climate change showed an indirect effect through modifications in runoff patterns. In contrast, the tropics

* Corresponding author.
E-mail address: nakhavali@iiasa.ac.at (M.(A. Nakhavali).

https://doi.org/10.1016/j.scitotenv.2024.170560
Received 30 August 2023; Received in revised form 26 January 2024; Accepted 28 January 2024
Available online 1 February 2024

show a relatively low increase in DOC leaching, which can be related to alterations in soil moisture and temperature.

Additionally, the study re-evaluates the role of land use change in DOC leaching, finding its effect to be considerably smaller than previously assumed. These insights emphasize the dominant roles of CO_2 fertilization and climate change in modulating DOC leaching, thereby refining our understanding of terrestrial carbon dynamics and their broader implications on the global C budget.

1. Introduction

Quantification of changes in soil carbon (C) stocks and their feedback to climate change are crucial for better understanding the perturbation of the global C cycle (Jobbágy and Jackson, 2000). Despite its importance in the global C cycle (Battin et al., 2009; Regnier et al., 2013a; Tranvik et al., 2009), the amount of C exported from terrestrial ecosystems into the inland water network has so far been estimated only coarsely by budget closure based on observed fluvial C exports to the coast and (the still poorly constrained) estimates of inland water CO_2 evasion and C burial in aquatic sediments (Battin et al., 2009; Drake et al., 2018; Raymond et al., 2013; Regnier et al., 2013b). Recent work suggests that the representation of lateral C exports in land surface schemes of Earth system models will arguably help to improve the representation of soil C cycling and its response to atmospheric CO_2 increase, climate change, and land-use change (Lauerwald et al., 2017; Nakhavali et al., 2018; Tian et al., 2015). Ignoring those exports has so far likely been compensated by an overestimation of heterotrophic soil respiration and/or C accumulation in the soil (Jackson et al., 2002; Janssens et al., 2003; Regnier et al., 2013b), potentially introducing a bias in future projections of CO_2-C and climate-C cycles feedbacks (Cox, 2019).

Dissolved organic C (DOC) represents about 20 % of the fluvial C export to the oceans (Dai et al., 2012), but its proportion in the terrestrial C inputs discharging into inland waters is likely higher (Nakhavali et al., 2021) because of its higher reactivity compared to particulate organic C (POC) and inorganic C mobilized by chemical alteration of rocks. DOC is thus a major contributor to the net-heterotrophy of inland waters and related CO_2 evasion (Battin et al., 2008). An increase in the export of DOC from soils to inland waters over the past decades has been observed for the UK (Freeman et al., 2001), Northern and Eastern United States (Stoddard et al., 2003), Canada (Bouchard, 1997), Norway (Hongve et al., 2004) and Czech Republic (Hejzlar et al., 2003). Several potential drivers of this increase were suggested, which affect soil DOC production as well as soil organic C (SOC) stocks as the primary source of soil DOC. Those include increase of DOC leaching due to the increase in temperature (Freeman et al., 2001; Rind et al., 1990), soil acidification (Funakawa et al., 2014; Pschenyckyj et al., 2020), CO_2 fertilization intensity (Clair et al., 1999), increase in precipitation (Hongve et al., 2004) and increased runoff and river discharge that have potentially led to changes in the fraction of soil DOC being laterally displaced through the river network (Ledesma et al., 2012), land use (Brye et al., 2001) and burning biomass (Clutterbuck and Yallop, 2010).

Although recent studies have utilized empirical models to map global dissolved organic (Guo et al., 2020), as highlighted in previous studies (Lauerwald et al., 2017; Regnier et al., 2013b), the limited amount of empirical data (global coverage and historical time series) and poorly documented available temporal and spatial data, leaves process-based models as the only tool to quantify the magnitude and temporal evolution of DOC exported flux from soil to rivers at regional to global scales. This is especially true when assessing these fluxes at global scale, describing their long-term trends, and attributing those changes to the main environmental drivers, which is virtually impossible to achieve via observations alone. Hence, recently the land surface model JULES-DOCM was developed (Nakhavali et al., 2018), which represents production and cycling of DOC within the soil column, and leaching of DOC from the soil column. JULES-DOCM has been calibrated and successfully validated at global scale for present day conditions (Nakhavali et al., 2021). In this study, the model simulates spatio-temporal trends in soil-river DOC fluxes on a global scale over the historical period from 1860 to 2010. The study emphasizes variations in temporal patterns and the influence of environmental drivers across boreal, temperate, tropical, and sub-tropical climatic zones. It is hypothesized that DOC leaching flux has risen during the study period, predominantly due to elevated atmospheric CO_2 levels and its fixation by vegetation. This research specifically explores to what extent increased DOC leaching counterbalances the C sink prompted by increased primary production, which is crucial for the temporal assessment of land C budgets.

2. Materials and methods

2.1. Model development

The historical patterns of DOC leaching from soils were studied using the novel extension of the JULES land surface model version 4.4, JULES-DOCM (Nakhavali et al., 2018). Vegetation dynamics within this model are represented through the TRIFFID model for nine different plant functional types (PFTs), as described by Harper et al. (2016). The representation of SOC is managed by the RothC model (Jenkinson et al., 1990; Jenkinson and Coleman, 2008), which defines four C pools. These pools are decomposable plant material (DPM) and resistant plant material (RPM), both receiving direct inputs from plant litter. During decomposition of the DPM and RPM pools, not all of the decomposed carbon is respired back to the atmosphere. The microbial biomass (BIO) and humified material (HUM) pools are also included, which are allocated a fraction of the decomposed C from DPM and RPM, that is not released as CO_2 into the atmosphere. All four soil organic matter pools leach DOC to the soil solution, where it may undergo decomposition, diffusion, ad/sorption or is leached from the soil column with soil drainage and subsurface runoff (Nakhavali et al., 2018) (Fig. S1, Table S1).

2.2. Model calibration and evaluation

In the previous study (Nakhavali et al., 2021), soil DOC dynamics were characterized through two fundamental equations for production and decomposition of DOC. DOC production $\left(F_{P_{n,i}}\right)$ in each soil layer (i) for each pool (n) was represented as:

$$F_{P_{n,i}} = S_{C_n} \times \left(1 - e^{\left(-K_p \times RM_i\right)}\right) \quad (1)$$

where SCn is the SOC content, K_p is DOC production basal rate, and RM_i is a rate modifier accounting for temperature, moisture, vegetation, and soil texture influences in each soil layer.

The DOC decomposition $\left(F_{D_{n,i}}\right)$ was formulated as follows:

$$F_{D_{n,i}} = S_{DOC_{n,i}} \times \left(1 - e^{\left(-K_{DOC_n} \times Ft_i\right)}\right) \quad (2)$$

where $S_{DOC_{n,i}}$ is the soil DOC content, K_{DOC_n} is the DOC decomposition basal rate, and Fti is a temperature-dependent rate modifier. During calibration, JULES-DOCM was updated to make K_p and K_{DOC} sensitive to the dominant plant functional types. The calibration was performed using the Latin hypercube sampling method to select random values across and beyond the observed ranges for comprehensive parameter

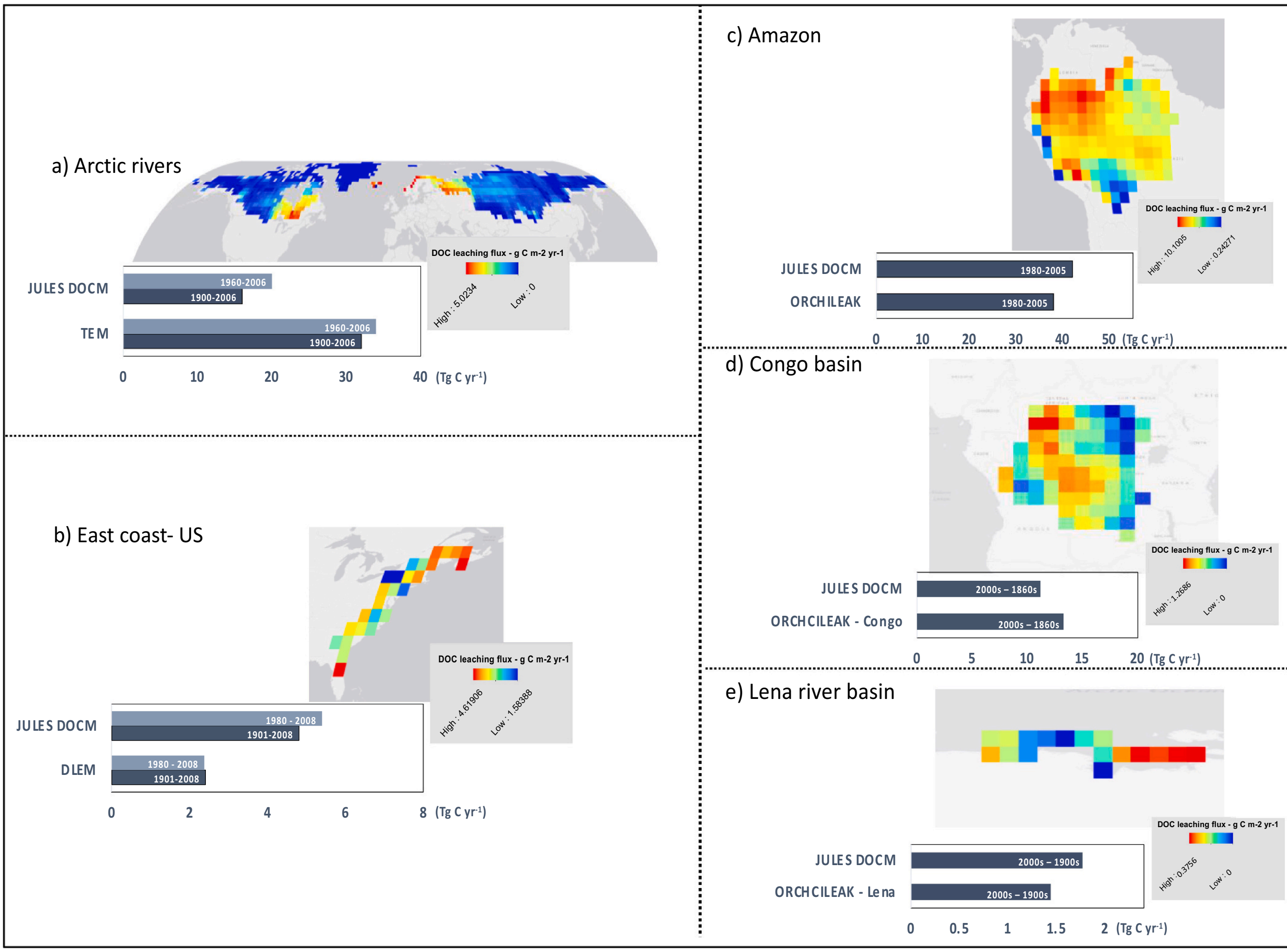


Fig. 1. Model comparison. Historical and present-day DOC leaching flux simulated by JULES at a) artic rivers b) east coast US and c) Amazon d) Congo basin (Hastie et al., 2021) and e) Lena River basin (Bowring et al., 2019) compared to relative models and periods.

space coverage. Specifically, for each PFT, 25 random combinations of K_p and K_{DOC} within the observed ranges and 5 additional values outside these ranges were selected. For cross-validation purposes, the soil DOC observations for each PFT were splitted between a calibration and a validation dataset. For PFTs with sparse data, all site combinations were used, allocating three-quarters for calibration and one-quarter for validation. For PFTs with more substantial data, a selection of random combinations was made. The root mean square error (RMSE) was computed for each K_p and K_{DOC} pairing across calibration and validation datasets, selecting the pair with the lowest RMSE for final calibration.

For further validation, our global version of JULES-DOCM was evaluated against a newly compiled, extensive database of measured DOC concentrations ($N = 109$), which we classified into the four main biomes of boreal, temperate, and tropical forests, and grassland. All the measurements falling within the same grid-cell (1.25° latitude, 1.875° longitude) were aggregated and the resulting grid-cell averaged concentration of 39 grid cells were used for model evaluation. Moreover, we also evaluated the simulated DOC leaching fluxes against observations from headwater streams taken from the GloRiCh database (Nakhavali et al., 2021).

2.3. Historical simulations

The historical simulation adhered to the TRENDY protocol (Sitch et al., 2015) with settings adapted from JULES (Harper et al., 2016) at an N96 resolution (1.875° longitude×1.25° latitude). The applied climate forcing was CRUNCEP version 4 (Harris et al., 2014) spanning from 1860 to 2010. Additionally, the model was forced by data on historical atmospheric CO_2 (Dlugokencky and Tans, 2013) and land cover data from HYDE v. 3.1 (Goldewijk et al., 2011). To achieve a pre-industrial steady state for simulated SOC and DOC pools, an accelerated spin-up method was employed, necessitating only 200 to 300 years of spin-up instead of several thousand years (Harper et al., 2016). In this method the decomposition rate of the most labile litter pool is used for all C pools. This was achieved by scaling the humus, biomass and resistant plant material decomposition rates, based on the labile plant material pool, by a factor of 33, 15 and 500, respectively. Then the simulated C pools were rescaled by the same scaling factors and another 300 years of spin up with the actual decomposition rate for each pool was performed to reach a steady-state corresponding to the “pre-industrial” 1860 conditions of climate, CO_2 and land-cover. Land use change was integrated into the model using a method that accounts for the conversion between natural vegetation and agricultural land (Harper et al., 2016). These transitions, along with their impacts on Plant Functional Type (PFT) fractions, were tracked, which in turn influenced NPP and subsequent DOC dynamics.

2.4. Identification of drivers

For transient simulation over the historical period, the initial condition was defined by using the final results from the steady state spin up. A transient simulation, S_{ALL}, was executed with time-variant climate,

Table 1
Global and regional DOC leaching changes. Dissolved organic carbon leaching fluxes changes, relative increase and attribution to environmental drivers (sum = 100 %).

	Average 1860s (Tg C yr-1)	Average 2000s (Tg C yr-1)	Difference 1860s:2010s (Tg C yr-1)	Increase (%)	CO_2 (%)	CLM (%)	LUC (%)
Global	250	292 ± 21	42	17	64	23	13
Boreal	19	24 ± 4	5	28	30	65	5
Temperate	41	49 ± 7	8	20	48	42	10
Tropics	152	173 ± 5	21	14	75	10	15
Sub-tropics	38	46 ± 5	8	20	60	28	12

land use, and CO_2 forcing data. To attribute DOC leaching variations to environmental drivers (CO_2 fertilization, land use change, climate change), three additional experiments were conducted: one with fixed land use (S_{LUC}), another with constant atmospheric CO_2 (S_{CO2}), and the last with static climate (S_{CLM}) forcing. The impact of land use change was then calculated as the difference S_{ALL}-S_{LUC}, that of atmospheric CO_2 increase as S_{ALL}-S_{CO2}, and that of climate change as S_{ALL}-S_{CLM}. To assess temporal trends, 10-year running means of simulation results were computed, suppressing inter-annual variability and making the long-term trends easier to analyse (Fig. S2).

2.5. Inter-model comparisons

For inter-model comparisons, only three prior studies were identified that examined historical DOC flux changes at the terrestrial-aquatic system interface on a regional scale. Those include Arctic catchments simulated with the land-surface TEM model over the period 1900–2006 (Kicklighter et al., 2013), catchments along the east coast of the US simulated with DLEM and covering the period 1901–2008 (Tian et al., 2015), and the Amazon basin simulated with ORCHILEAK for the 1861–2005 period (Lauerwald et al., 2020). JULES-DOCM was applied over identical study areas and timeframes, with results subsequently compared.

3. Results and discussion

3.1. Model validation and comparison

In our previous study (Nakhavali et al., 2021), the model was successful in replicating dissolved organic carbon concentrations in both topsoil and subsoil, showing strong correlations with measured data across different biomes, and it accurately captured the controlling leaching processes into river systems; for more details, see Supplementary Information and Nakhavali et al. (2021).

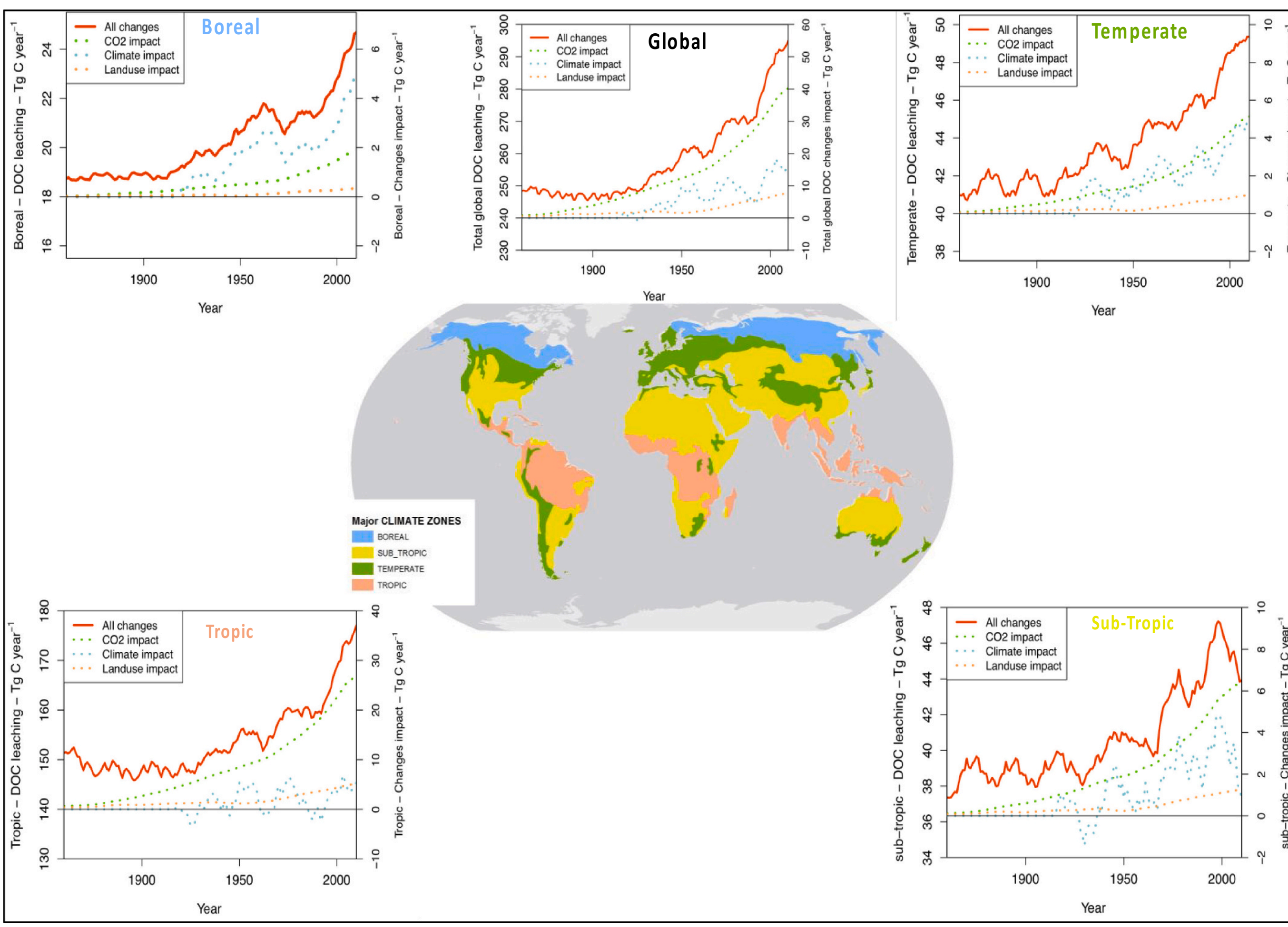


Fig. 2. DOC leaching controllers per major climate zones. Simulated historical trend of global and regional DOC leaching flux and the impact of CO_2, climate and land-use change.

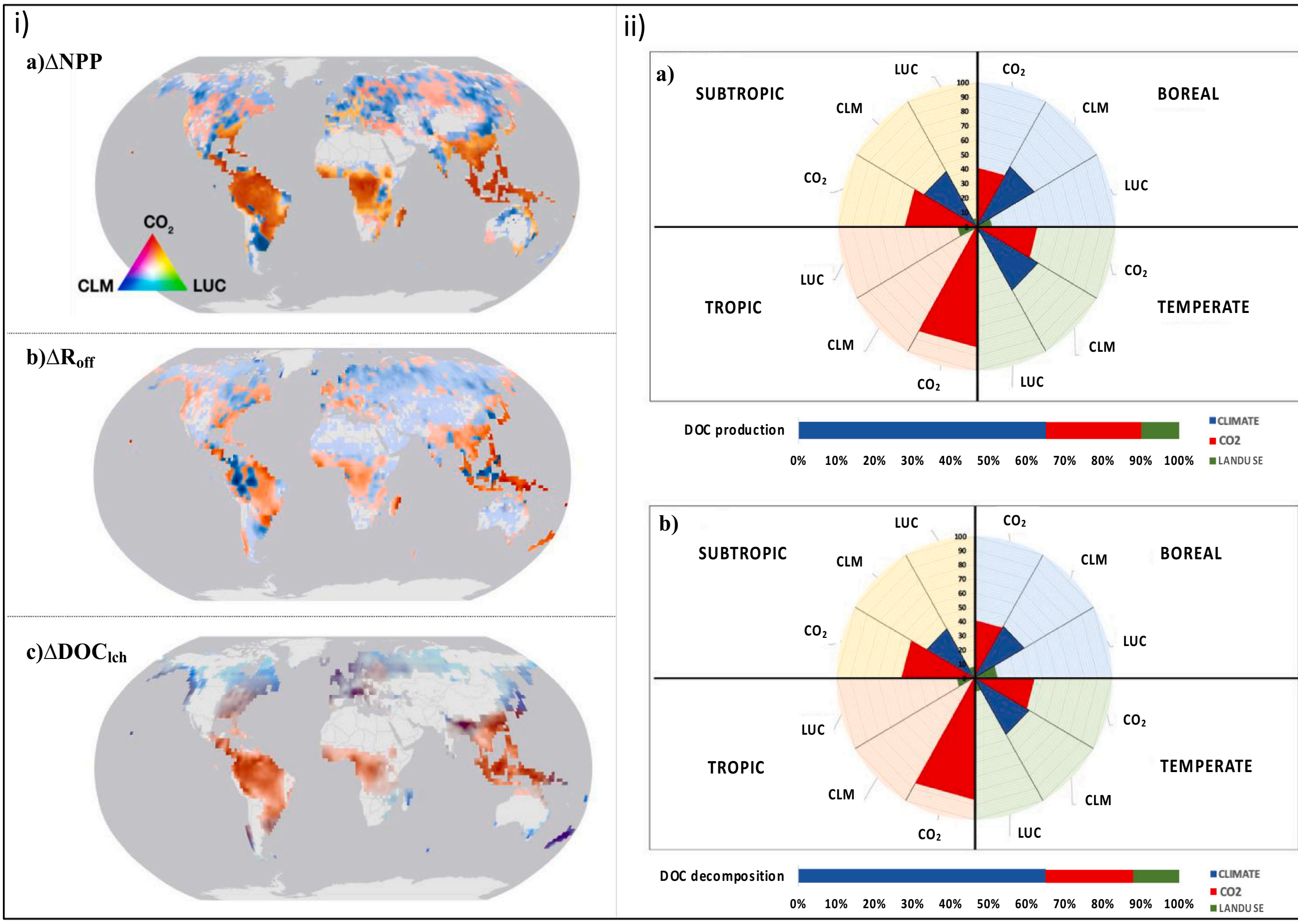


Fig. 3. Global and regional environmental drivers of changes in NPP, runoff and DOC leaching. Effect of CO_2 fertilization, climate (CLM) and land use (LUC) changes on i) NPP, Runoff (R_{off}), DOC leaching (DOC_{lch}), and ii) soil DOC production and decomposition fluxes.

The challenge of evaluating simulated trends against measurements, particularly in low latitudes, persists due to the limited availability and scattered observational data. Thus, a comparison was made with five regional-scale models, previously evaluated across various biomes and scales, to assess their DOC leaching simulation changes against the findings of this study (Fig. 1). The simulated average DOC leaching fluxes in Arctic region rivers indicate an increase attributed to climate change, consistent with the TEM model. However, the fluxes for the periods 1900–2006 and 1960–2006 are less than those of the TEM model (Fig. 1-a). This could be due to the missing wetland mechanism representation in JULES, hence lower flux from this area (Nakhavali et al., 2021). Moreover, in TEM model the production and decomposition of DOC is fairly simplified compared to JULES-DOCM and does not account for impact of some parameters such as vegetation, clay and depth on production and decomposition of DOC. Additionally, TEM does not include the diffusion and vertical transport of DOC within the soil layers. This results in higher DOC concentration available at the top layers for leaching. However, TEM includes the potential effect of permafrost on DOC dynamics such as production and leaching. Therefore, the production and leaching of DOC varying seasonally that is different than JULES-DOCM.

Our findings for the US East Coast, based on averages for the periods 1901–2008 and 1980–2008, indicate a minor increase. These results are marginally higher than those from the DLEM model, which shows a slight decrease. This discrepancy might be attributed to the negative effects of land-use on DOC leaching flux as represented in the DLEM model (see Fig. 1-b). In DLEM model the land use has changed from cropland to forest which has influenced the DOC leaching by altering DOC production and hydrology of region. However, in JULES-DOCM, the coarser resolution relative to DLEM prevents the inclusion of these fine-scale land use changes. Nevertheless, it is acknowledged that the DOC leaching from soils to rivers could be higher than what is simulated by DLEM, due to the potential decomposition of DOC within the river system. This suggests that other sources of DOC, not included in JULES-DOCM such as those from land management practices or organic soils, might be less significant than the in-transit decomposition of DOC, which is accounted for in DLEM but not in the JULES-DOCM model.

Our analysis of the leaching flux from terra firme Amazon soils (averaged for 1980–2005) and across both the Congo basin (averaged for 1860–2000) and the Lena River basin (averaged for 1900–2000) aligns closely with the ORCHILEAK model (see Fig. 1-c, 1-d and 1-e). It's important to note, however, that the ORCHILEAK model also accounts for inundated soils, leading to a slightly elevated total leaching flux. This highlights that wetlands play a significant role in contributing DOC to rivers (Lauerwald et al., 2020).

3.2. Global and regional DOC fluxes and drivers

3.2.1. Current and historical DOC leaching

An average global terrestrial DOC leaching is estimated at 292 ± 21 Tg C yr^{-1} for present day (2000–2010), 24 ± 4 Tg C yr^{-1} of which in the boreal, 49 ± 7 Tg C yr^{-1} in the temperate, 46 ± 5 Tg C yr^{-1} in the subtropical and 173 ± 5 Tg C yr^{-1} in the tropical zone (Table 1). Simulation results indicate that the average global DOC leaching from 1861 to 1870 was 250 Tg C yr^{-1} and increased since then by 17 %. The estimates, when detailed across major climate zones, reveal that the boreal zone experienced the highest relative increase of 28 %. This is followed by a 20 % increase estimated for both the temperate and sub-tropical zones. The tropical zone saw the smallest relative increase of 14 %, as shown in Fig. 2. Over the last three decades, however, the increase per climate zone has been linear, with the boreal and tropical zones each showing a 15 % and 11 % increase, respectively. There was a slight decrease during 1985–1987, after which DOC leaching in the temperate zone increased by 8 %. The sub-tropical zone had the smallest relative increase of 2 %, as indicated in Fig. S4.

3.2.2. Drivers and impacts on DOC leaching

The attribution analysis shows that CO_2 fertilization is the strongest driver of the global increase in DOC leaching over the historical period, contributing 64 % (39 Tg C yr^{-1}) of total changes. Climate and land use change are respectively responsible for 23 % (16 Tg C yr^{-1}) and 13 % (7 Tg C yr^{-1}) of the total change, and thus have a lower impact on the increasing leakiness of the terrestrial C cycle (Table S2; Fig. S5). Analysing the impact of these drivers in each major climate zone nevertheless reveals significant differences.

For the boreal zone, climate change is by far the dominant driver of the increase in DOC leaching, contributing 65 % of the total increase. This finding aligns with a recent experimental study from Sweden, which identified rising runoff as the primary cause for the observed uptick in DOC leaching (Nydahl et al., 2017). Correspondingly, our simulations indicate that the highest DOC concentrations are found in the topsoil, particularly within the boreal zone. This zone also has the highest ratio of surface runoff (effective runoff contributing to topsoil leaching) to total runoff, of about 80 %, when compared to other climate zones (Fig. S6). The remaining increase is almost entirely attributed to the enhanced terrestrial productivity (30 %) while land-use has virtually no effect on the terrestrial DOC leaching flux. In the temperate zone, CO_2 fertilization accounts for 48 % and climate change for 42 % of the DOC leaching increase, while land use change contributes only 10 % of total changes. For the tropical zone, CO_2 fertilization was identified as the main control, contributing 75 % of the increase in simulated DOC leaching. Land use and climate changes show a lower impact on tropical zone DOC leaching of respectively 15 % and 10 % of total changes. The results for the sub-tropical zone are similar to those of the tropical zone, with CO_2 fertilization as main control explaining 60 % of the simulated increase in DOC leaching, climate and land use change contributing 28 % and 12 %, respectively.

3.2.3. Mechanisms and correlations

To comprehend the primary mechanisms through which CO_2, climate, and land use changes influence DOC leaching across major climate zones, an analysis was conducted on how these factors affected terrestrial NPP and runoff (Fig. 3). This analysis allows to elucidate whether the anthropogenic perturbations mostly impact the DOC leaching fluxes through their influence on the carbon cycle, the hydrological cycle, or a combination of both.

On a global scale, an average present-day NPP of 92 Pg C yr^{-1} was simulated, which compared to the period 1861–1870 has increased by 21 Pg C yr^{-1} (+30 %) (Fig. S7) and highly temporal and spatial correlation of DOC leaching with NPP at global scale ($r^2 = 0.53$ and $r^2 = 0.9$ respectively) (Fig. S8–9). The findings obtained through the empirical approach also indicate a significant positive spatial correlation between the DOC concentration and NPP at a global scale (Guo et al., 2020). The temperate zone exhibits the greatest relative increase in NPP (+38 %), as listed in Table S2. This is succeeded by the sub-tropical and boreal zones with simulated NPP increases of 36 % and 33 %, respectively, and the tropical zone with a 24 % increase. These figures agree with the increases reported in observational studies for these biomes (Peterson and Lajtha, 2013; Rizinjirabake et al., 2019). In terms of drivers, CO_2 fertilization accounts for about 74 % of the NPP increase globally and is also the main driver of change in each climate zone with the exception of boreal zone (Nydahl et al., 2017). Land use and climate changes contribute 14 % and 12 % of the global NPP increase, respectively (Fig. 3-i-a, Table S2) which is lower than previously reported regional rates (Chu et al., 2016; Xiao et al., 2019). The model simulations indicated that land use changes, particularly the conversion between natural vegetation and agricultural land, had quantifiable impacts on PFT fractions. These alterations were reflected in the NPP values, which subsequently influenced the DOC dynamics observed during the simulation period. The sensitivity of PFT fractions to land use change was found to be minor; however, it accounted for a non-negligible influence on the overall DOC leaching flux (Fig. S10).

Overall, runoff increased by 4900 kg water m^{-2} yr^{-1} (+13.5 %) over the historical period (Table S3). This trend is consistent with the observed global runoff increment of 4 % during the period of 1900 to 1996 (+5 % in this study) (Labat et al., 2004), with a high temporal and spatial correlation between runoff and DOC leaching ($r^2 = 0.75$ and $r^2 = 0.49$ respectively) (Fig. S8–9). These findings align with previous empirical studies indicating that the primary inter-annual variability and spatial pattern of DOC leaching correspond to runoff (Gielen et al., 2011; Michalzik et al., 2001; Neff and Asner, 2001). The boreal zone shows the highest runoff increase of 31 % followed by temperate, subtropic and tropical zones (17 %, 15 % and 13 % increases respectively, Table. S2). The attribution analysis shows that climate change is the main driver of runoff increase contributing 50 % of the total change, followed closely by the CO_2 fertilization effect (41 %), land use changes only contributing 9 % of total changes (Fig. 3-i-b, Table. S1). These results indicate that the fertilization effect of CO_2 may have a notable impact on the increment of leaf area index and the rate of transpiration, which in turn can lead to alterations in resulting runoff as indicated by prior studies (Zhang et al., 2022a). Additionally, modifications in land use patterns can bring about changes in evapotranspiration and infiltration rates, which can subsequently lead to slight alterations in runoff (Piao et al., 2007). However, in the boreal zone climate change is responsible for 75 % of total runoff change. CO_2 fertilization and land use have less impact with 21 % and 4 %, respectively (Table S2). In comparison to other biomes, the tropical zone exhibits a lesser increase in runoff. However, in the temperate, tropical, and sub-tropical zones, the impacts of CO2 fertilization and climate change on runoff change are similar. In contrast, the influence of land use change on runoff change is less pronounced across all the major climatic zones. In contrast to NPP changes that are at first order dominated by the CO_2 fertilization, the relative influence of the drivers of runoff changes reveal significant differences among major climate zones.

The global map of the impacts of temperature and runoff on DOC leaching flux and its change between 1860 and 2010 are presented in Fig. S11. These resources indicate that the hotspots of changes in DOC leaching are largely corresponding to the hotspots of changes in runoff (r^2: 0.38). On the other hand, there appears to be no meaningful correlation between the changes in DOC leaching and temperature (r^2: 2e-0.4).

Therefore, it can be inferred that the rise in DOC leaching resulting from the impact of climate change is primarily controlled by precipitation and the consequent runoff, rather than temperature. As a result, this forms the justification for certain models that utilize exclusively the DOC concentration and runoff to assess the resulting DOC leaching flux (Kindler et al., 2011).

3.2.4. Ecosystem dynamics and model implications

In addition to the primary mechanism of runoff, it should be noted that the DOC leaching is also influenced by the DOC stock, which is contingent upon both production and decomposition processes (Kalbitz et al., 2000). Our simulated, global DOC production and decomposition fluxes increase by 21 % and 22 % over historical, period respectively (Fig. S12). Notably, while the rate of DOC production increase was lower than that of decomposition, the former flux was observed to be 60 %

larger than the latter, resulting in an overall increase in historical DOC stocks (Table S4). In JULES-DOCM two rate modifiers of soil temperature and moisture are controlling the DOC production (both temperature and moisture) and decomposition (temperature) (Nakhavali et al., 2018). These rate modifiers are determined based on the prior observations of the biodegradability of plants in various biomes, which are associated with temperature and moisture levels (Johnson et al., 2000; Kalbitz et al., 2003; Nakhavali et al., 2021; Yule and Gomez, 2009).

Our results show the highest DOC production and decomposition increase in boreal zone (28 % and 32 % respectively). However, the boreal zone shows the highest rate of leached DOC production (21.81 %) compared to the other biomes (Table S4). The main controller of the DOC production and decomposition in boreal zone is climate change (48 % and 42 % of total changes respectively) (Fig. 3-ii-a and b). The climate change in boreal zone results the highest increase of temperature and moisture modifiers (7 % and 10 % respectively) which control the DOC production. However, the decomposition is only controlled by temperature which shows a lower increase compared to the soil moisture (Table S4). Therefore, the present-day DOC decomposition against production in boreal zone is the lowest compared to other biomes (37 %). Together with the highest runoff increase, mainly due to the climate change, the highest relative increase of DOC leaching is observed in the boreal zone. This is in line with a recent study in Sweden showed that increasing runoff is the main reason for the observed increase in DOC leaching (Nydahl et al., 2017). Therefore, in the boreal zone, DOC leaching is transport-limited, meaning it is constrained by runoff. This accounts for the greater sensitivity of changes in DOC leaching to runoff rather than temperature, as shown in Fig. S6. Temperature primarily affects reaction rates (production and decomposition of DOC), as noted by Nakhavali et al. (2021).

Temperate and sub-tropical zones show the similar absolute increase in DOC leaching due to the similar NPP increase (38 %), controlled mainly by CO_2 fertilization (temperate: 67 %, subtropical: 68 % of total changes) (Table S4). However, the soil moisture and temperature increase in temperate is lower than in the boreal zone (by 7 % and 5 %, respectively), hence the lower relative increase in DOC leaching. Moreover, similar to the boreal zone, the runoff increase in the temperate zone is mainly due to climate change. However, opposed to the temperate zone, CO_2 fertilization is identified as the main controller of the DOC production and decomposition (52 % and 53 % of total changes respectively) (Fig. 3-ii-a and b). The increase of soil moisture and temperature are slightly lower than in the temperate zone (both 5 %) and the runoff increase is mainly controlled by climate change (42 %). Therefore, both temperate and sub-tropic zones are both substrate (i.e., limited by DOC availability) and transport limited (i.e., limited by runoff).

Despite tropical terrestrial ecosystems exhibiting the highest efficiency of carbon sequestration and a greater capacity for carbon storage in vegetation (Hirano et al., 2009; Sitch et al., 2003), the zone demonstrates the least relative increase in DOC leaching. This is attributed to a minimal increase in DOC production resulting from a lower increase in soil moisture and temperature, both of which control DOC production. Additionally, although the tropical zone displays the highest overall NPP increase (Fujii et al., 2009; Zhou et al., 2016), the comparatively lower rate of DOC production can be attributed to the estimated 2 % and 5 % increases in soil moisture and temperature, respectively.

Overall, the relative increase of DOC leaching is close to that in runoff, suggesting that at global scale, the increase in DOC flux through the terrestrial-aquatic system's interface is largely transport-limited. However, in some parts of the boreal zone such as Northern Europe, the increase in the DOC stock also contributes to the increase of DOC leaching flux (Fig. S13–14). Moreover, in some part of the tropical zone such as Amazon, where there is no significant increase in the DOC stocks, the DOC leaching flux is not increasing as well (Fig. S14).

Over the simulation period, top soil (here defined as the upper 35 cm) leaching contributes about 90 % to total DOC leaching from soil. However, as it was shown in a previous study (Nakhavali et al., 2021), the top soil layer contains only ~40 % of total soil DOC stocks. The subsoil DOC stocks receive significant amounts of the organic matter through diffusion (Braakhekke et al., 2013). The magnitude of diffusion flux is 0.2 % of DOC leaching flux (averaged total diffusion for period 1860 to 2010 is 0.5 Tg C yr-1), indicating a low mobility of the diffused DOC from the subsoil (Worrall et al., 2008). This limited mobility is the reason why variations in the total SOC and DOC stocks may not correspond directly with changes in DOC leaching (Fig. S6). Leaching is primarily associated with the topsoil DOC cycling, whereas the majority of total DOC stocks are found in the bottom soil. The transfer of DOC between the top and bottom soil layers occurs at a slow pace. NPP increase drives increase in biomass, litterfall, SOC and finally DOC leaching. The increase in NPP entrains a stronger increase in DOC concentrations in the topsoil compared to the subsoil (Guggenberger and Kaiser, 2003), and thus a stronger increase in the DOC leaching flux than in the total soil DOC stock.

The total percentage of NPP lost by "*terra firme*" ecosystems has decreased from 0.35 % on average during 1861–1870 to 0.32 % under present day conditions (Fig. S15). For the boreal zone overall, this ratio is not decreasing significantly with 0.31 % for 1861–1870 to 0.30 % for present day. In the temperate zone the overall ratio is changing from 0.30 % to 0.28 % and in the tropic zone the overall ratio is changing by 0.03 % from 0.4 % for 1861–1870 to 0.37 % for present day. Lastly, for sub-tropic zone this ratio decreases from 0.27 % to 0.25 % for present day. The decline in the proportion NPP lost through dissolved DOC leaching can be attributed to two factors: firstly, the relatively lower increase in runoff flux compared to NPP, with a disparity of 15.5 % versus 30 %, and secondly, the quantity of NPP that has yet to be translated into an equivalent rise in the DOC reservoir (Hensgens et al., 2020). This explains the lowest decrease of the DOC leaching/NPP ratio for the boreal and the highest decrease of DOC leaching/NPP for the tropical zone. Nevertheless, although NPP to DOC leaching ratios across these biomes are quantitatively small, they are ecologically significant, reflecting substantial changes in the C sequestration dynamics and the responsiveness of different ecosystems to global change drivers.

A previous study by Regnier et al. (2013a) suggests a significant impact of the anthropogenic perturbation on the leached C to the aquatic ecosystem without attempting an attribution analysis to CO_2 fertilization, climate change or land use change. While our simulation omits factors like soil erosion and harvesting that could affect SOC stocks, along with minor compartments which were included in Regnier et al. (2013a), the estimation indicates a minor contribution of land use change on the changes in DOC leaching flux. Our results show a minor contribution of land use change on the NPP changes as well as the highest impact of LUC in the tropical zone (15 %) and lowest impact in the boreal zone (6 %) (Table. S2). Nevertheless, the attribution of land use impact on the production of labile DOC is slightly different (Table S5) with the highest land use impact on tropical (14 %) and the lowest in sub-tropical zone (4 %) in line with studies showing that land use change shifts the soil C to more labile forms (Zhang et al., 2020). Nevertheless, the minor role of land use change highlighted in our study contrasts with previous research, which focused solely on carbon leaching affected by land use changes due to agricultural conversion (Guggenberger and Kaiser, 2003; Kindler et al., 2011). These studies did not consider other factors such as CO_2 fertilization and climate change, which our study finds to be significant influencing factors of DOC leaching.

3.3. Model limitations

The effect of increasing atmospheric CO_2 concentrations may be debatable, as other limiting factors of plant productivity, in particular nutrients (Nakhavali et al., 2022), are not considered in this version of JULES. Climate change can be considered a reliable driver, if we assume that drivers of spatial differences can act in a similar way as drivers of temporal change, since the model has been found to well reproduce present-day large scale spatial patterns in our previous study (Nakhavali et al., 2021). While our model captures the predominant factors of climate change, atmospheric CO2 increase, and land use change, we recognize the absence of other potential driving factors in our analysis. Despite incorporating widely-studied DOC controls like temperature, precipitation, and vegetation into the model, it still lacks representations for pH, acid deposition, and nutrients. pH has an impact on the concentration and leaching of soil DOC (Michalzik et al., 2001) by affecting the adsorption capacity (Kalbitz et al., 2000).

On the other hand, acid deposition controls the ionic strength (IS) and soil acidity which affects concentration and leaching of soil DOC (Monteith et al., 2007).

The nutrients impact on soil DOC, for instance, N deposition can modify the enzymatic activity (such as ß- glycosidase) which are controlling the C release from organic matters and increases DOC release (Bragazza et al., 2006). Additionally, it also impacts the microbial activities which are important controller of organic matters processing (Pregitzer et al., 2004).

Furthermore, within JULES-DOCM, only mineral soils are represented, excluding organic-rich soils, constituting around 6–8 % of the total land area (Lehner and Döll, 2004) and wetlands that could contribute up to about 20 % of fluvial DOC export to the coast (Harrison et al., 2005; Lauerwald et al., 2012). Hence, including these drivers and processes can improve the ability of JULES-DOCM to fully represent and predict the temporal and spatial dynamics of soil DOC.

Additionally, the JULES-DOCM model currently calculates DOC consumption by factoring in decomposition within the soil before it enters the streams and overlooks in-stream processes such as microbial degradation or photodegradation, which can significantly consume DOC. These processes are known to reduce DOC concentrations and alter their composition, potentially leading to an overestimation of the actual DOC fluxes that reach aquatic systems (Battin et al., 2009; Drake et al., 2018; Raymond et al., 2013; Regnier et al., 2013b). The exclusion of these consumption pathways from the model framework means that the fluxes represented may not fully correspond to the net DOC available for transport to oceanic reservoirs. Hence, future improvement of the model would benefit from incorporating these dynamics to enhance the accuracy of DOC flux predictions and to provide a more nuanced understanding of carbon cycling within the terrestrial-aquatic interface.

Regarding the land use change effect, this study only accounts for vegetation transitions and does not represent processes such as increased soil erosion following deforestation, which impacts SOC stocks. Furthermore, land use change can alter SOC production, movement and consumption and impact the bioavailable DOC and water extractable OC (Boyer and Groffman, 1996). Moreover, the labile fraction of DOC is dependent on land use (e.g. higher in cropland than in forest soils) (Boyer and Groffman, 1996). This suggests a likely underestimation of the global-scale impact of land use change on DOC leaching. It is however virtually impossible to test these hypotheses from observations as their spatio-temporal coverage is way too scarce to support a global analysis, especially with regard to historical trends.

Land use change is limited to updated vegetation cover following the competition between PFTs and prescribed agricultural masks (Harper et al., 2016) which impact the DOC dynamics by mapping out the chain of effects from changes in vegetation cover that affect plant productivity, cascading to NPP and altering SOC stocks, and which in turn impacts the SOC and DOC production, decomposition and leaching (Fig. S16; see supporting document for detail). The model estimates a 14 % contribution of LUC on NPP, indicating a sensitive relationship between vegetation productivity and the carbon cycle (Table S6). Subsequently, LUC influences SOC by altering organic matter input through litter deposition, with the model capturing a 16 % contribution of LUC on SOC stocks. This change in SOC stock affects DOC production, and subsequently decomposition, and leaching, with the model estimating a 13 % contribution of LUC on DOC pools and leaching. However, it is important to note that JULES-DOCM does not fully account for all processes, such as how LUC can alter soil hydrological properties, which in turn might influence lateral water flow and carbon advection. Other studies indicate that the impact of LUC on these aspects is relatively minor compared to other factors and their estimated effects of LUC on regional and global DOC leaching align with the findings of our study (Tian et al., 2023; Zhang et al., 2022b). Despite its less pronounced role, the effect of land use change on DOC cycling is still a vital element for a more complete understanding and accurate predictions of the model.

However, it is important to note that JULES-DOCM does not fully account for all processes. Our model only focusses on DOC and thus does not incorporate the additional effects of erosional fluxes of soil and particulate organic carbon (POC), which are known to be significantly accelerated by the conversion of forested areas into agricultural land (Van Oost et al., 2012; Wang et al., 2017). Note, however, that our results concur with other very recent studies that report minor impacts of LUC on regional and global DOC leaching, including with models explicitly representing POC (Tian et al., 2023; Zhang et al., 2022b).

Nevertheless, despite the missing processes and the required future improvement, JULES-DOCM performed satisfactory comparing against all metrics and provides for the first time a detailed historical assessment of contribution of the soil DOC production, decomposition and leaching controllers over the historical period. This study lays groundwork for improving the representation of controllers and trends of terrestrial C cycling in ESMs, particularly focusing on the laterally transported fraction across both historical and future periods.

4. Conclusion

Dissolved Organic Carbon (DOC) holds a pivotal role within terrestrial carbon dynamics, influencing both terrestrial and oceanic carbon frameworks. Using the globally calibrated JULES-DOCM ecosystem model, this research reveals a 17 % increase in terrestrial DOC contributions to global river systems between 1860 and 2010, leading to a current flux of 292 Tg C yr^{-1}.

The atmospheric CO_2 fertilization emerges as the primary impetus behind this surge, through enhanced vegetation growth and consequent rises in biomass and soil carbon pools. Although globally CO_2 remains the dominant factor, regional variances emerge. In temperate and boreal zones, climate change significantly impacts DOC levels, even surpassing CO_2 influences in boreal regions. The tropics, while being a major DOC source, record a moderate relative growth. This study challenges the previously overstated role of land use change, emphasizing the need for its reassessment in shaping DOC leaching dynamics.

CRediT authorship contribution statement

Mahdi (Andre) Nakhavali: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Data curation, Conceptualization. **Ronny Lauerwald:** Writing – review & editing, Writing – original draft, Methodology. **Pierre Regnier:** Writing – review & editing, Writing – original draft, Supervision. **Pierre Friedlingstein:** Writing – review & editing, Writing – original draft, Supervision.

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

Global version of JULES-DOCM modified for this paper can be found at: https://code.metoffice.gov.uk/svn/jules/main/branches/dev/mahdinakhavali/vn4.4_JULES_DOCM_GLOBAL_NAKHAVALI/ (registration is required).

Acknowledgments

This study has received funding from European Commission under the scope of the ForestNavigator project (grant agreement No 101056875). P.F received funding from the European Union's Horizon 2020 research and innovation programme under grant agreement No 821003 (4C project). RL received funding from the French state aid managed by the ANR under the "Investissements d'avenir" programme [ANR-16-CONV-0003]. P.R. received funding from the European Union's Horizon 2020 research and innovation programme under Grant Agreement no. 101003536 (ESM2025 – Earth System Models for the Future).

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2024.170560.

References

Battin, T.J., Kaplan, L.A., Findlay, S., Hopkinson, C.S., Marti, E., Packman, A.I., Newbold, J.D., Sabater, F., 2008. Biophysical controls on organic carbon fluxes in fluvial networks. Nat Geosci 1 (8), 95–100. https://doi.org/10.1038/ngeo602.

Battin, Tom J., Luyssaert, S., Kaplan, L.A., Aufdenkampe, A.K., Richter, A., Tranvik, L.J., 2009. The boundless carbon cycle. Nat Geosci 2 (9), 598–600. https://doi.org/10.1038/ngeo618.

Bouchard, A., 1997. Recent lake acidification and recovery trends in southern QUEBEC. Canada 5, 225–245.

Bowring, S.P.K., Lauerwald, R., Guenet, B., Zhu, D., Guimberteau, M., Tootchi, A., Ducharne, A., Ciais, P., 2019. ORCHIDEE MICT-LEAK (r5459), a global model for the production, transport, and transformation of dissolved organic carbon from Arctic permafrost regions - part 1: rationale, model description, and simulation protocol. Geosci Model Dev 12 (8), 3503–3521. https://doi.org/10.5194/gmd-12-3503-2019.

Boyer, J.N., Groffman, P.M., 1996. Bioavailability of water extractable organic carbon fractions in forest and agricultural soil profiles. Soil Biol Biochem 28 (6), 783–790. https://doi.org/10.1016/0038-0717(96)00015-6.

Braakhekke, M.C., Wutzler, T., Beer, C., Kattge, J., Schrumpf, M., Ahrens, B., Schöning, I., Hoosbeek, M.R., Kruijt, B., Kabat, P., Reichstein, M., 2013. Modeling the vertical soil organic matter profile using Bayesian parameter estimation. Biogeosciences 10 (1), 399–420. https://doi.org/10.5194/bg-10-399-2013.

Bragazza, L., Freeman, C., Jones, T., Rydin, H., Limpens, J., Fenner, N., Ellis, T., Gerdol, R., Hajek, M., Hajek, T., Iacumin, P., Kutnar, L., Tahvanainen, T., Toberman, H., 2006. Atmospheric nitrogen deposition promotes carbon loss from peat bogs. Proc Natl Acad Sci 103 (51), 19386–19389. https://doi.org/10.1073/pnas.0606629104.

Brye, K.R., Norman, J.M., Bundy, L.G., Gower, S.T., 2001. Nitrogen and carbon leaching in agroecosystems and their role in denitrification potential. J Environ Qual 30 (1), 58. https://doi.org/10.2134/jeq2001.30158x.

Chu, C., Bartlett, M., Wang, Y., He, F., Weiner, J., Chave, J., Sack, L., 2016. Does climate directly influence NPP globally? Global Change Biol 22 (1), 12–24. https://doi.org/10.1111/gcb.13079.

Clair, T.A., Ehrman, J.M., Higuchi, K., 1999. Changes in freshwater carbon exports from Canadian terrestrial basins to lakes and estuaries under a 2xCO2 atmospheric scenario. Global Biogeochem Cycles 13 (4), 1091–1097. https://doi.org/10.1029/1999GB900055.

Clutterbuck, B., Yallop, A.R., 2010. Land management as a factor controlling dissolved organic carbon release from upland peat soils 2: changes in DOC productivity over four decades. Sci Total Environ 408 (24), 6179–6191. https://doi.org/10.1016/j.scitotenv.2010.08.038.

Cox, P.M., 2019. Emergent constraints on climate-carbon cycle feedbacks. Curr Clim Change Rep 5 (4), 275–281. https://doi.org/10.1007/s40641-019-00141-y.

Dai, M., Yin, Z., Meng, F., Liu, Q., Cai, W.J., 2012. Spatial distribution of riverine DOC inputs to the ocean : an updated global synthesis. *Current Opinion in Environmental {…}*. http://www.sciencedirect.com/science/article/pii/S1877343512000358.

Dlugokencky, E., & Tans, P. (2013). Observed atmospheric CO2 - http://www.esrl.noaa.gov/gmd/ccgg/ trends.

Drake, T.W., Raymond, P.A., Spencer, R.G.M., 2018. Terrestrial carbon inputs to inland waters : A current synthesis of estimates and uncertainty. Limnol Ocean Lett 132–142. https://doi.org/10.1002/lol2.10055.

Freeman, C., Evans, C.D., Monteith, D.T., Reynolds, B., Fenner, N., 2001. Export of organic carbon from peat soils. Nature 412 (6849), 785. https://doi.org/10.1038/35090628.

Fujii, K., Uemura, M., Hayakawa, C., Funakawa, S., Sukartiningsih, Kosaki, T., & Ohta, S., 2009. Fluxes of dissolved organic carbon in two tropical forest ecosystems of East Kalimantan. Indonesia Geoderma 152 (1–2), 127–136. https://doi.org/10.1016/j.geoderma.2009.05.028.

Funakawa, S., Fujii, K., Kadono, A., Watanabe, T., Kosaki, T., 2014. Could soil acidity enhance sequestration of organic carbon in soils?. In: Soil Carbon. Springer International Publishing, pp. 209–216. https://doi.org/10.1007/978-3-319-04084-4_22.

Gielen, B., Neirynck, J., Luyssaert, S., Janssens, I.A., 2011. The importance of dissolved organic carbon fluxes for the carbon balance of a temperate scots pine forest. Agric For Meteorol 151 (3), 270–278. https://doi.org/10.1016/j.agrformet.2010.10.012.

Goldewijk, K.K., Beusen, A., Van Drecht, G., De Vos, M., 2011. The HYDE 3.1 spatially explicit database of human-induced global land-use change over the past 12,000 years. Global Ecol. Biogeogr. 20 (1), 73–86. https://doi.org/10.1111/j.1466-8238.2010.00587.x.

Guggenberger, G., Kaiser, K., 2003. Dissolved organic matter in soil : challenging the paradigm of sorptive preservation. Geoderma 113, 293–310. https://doi.org/10.1016/S0016-7061(02)00366-X.

Guo, Z., Wang, Y., Wan, Z., Zuo, Y., He, L., Li, D., Yuan, F., Wang, N., Liu, J., Song, Y., Song, C., Xu, X., 2020. Soil dissolved organic carbon in terrestrial ecosystems: global budget, spatial distribution and controls. Global Ecol Biogeogr 29 (12), 2159–2175. https://doi.org/10.1111/geb.13186.

Harper, A.B., Cox, P.M., Friedlingstein, P., Wiltshire, A.J., Jones, C.D., Sitch, S., Mercado, L.M., Groenendijk, M., Robertson, E., Kattge, J., Bönisch, G., Atkin, O.K., Bahn, M., Cornelissen, J., Niinemets, Ülo, Onipchenko, V., Peñuelas, J., Poorter, L., Reich, P.B., Van Bodegom, P., 2016. Improved representation of plant functional types and physiology in the Joint UK Land Environment Simulator (JULES v4.2) using plant trait information. Geosci Model Dev 9 (7), 2415–2440. https://doi.org/10.5194/gmd-9-2415-2016.

Harris, I., Jones, P.D., Osborn, T.J., Lister, D.H., 2014. Updated high-resolution grids of monthly climatic observations - the CRU TS3.10 dataset. Int J Climatol 34 (3), 623–642. https://doi.org/10.1002/joc.3711.

Harrison, J.A., Caraco, N., Seitzinger, S.P., 2005. Global patterns and sources of dissolved organic matter export to the coastal zone: results from a spatially explicit, global model. Global Biogeochem Cycles 19 (4). https://doi.org/10.1029/2005gb002480.

Hastie, A., Lauerwald, R., Ciais, P., Papa, F., Regnier, P., 2021. Historical and future contributions of inland waters to the Congo Basin carbon balance. Earth Syst Dyn 12 (1), 37–62. https://doi.org/10.5194/esd-12-37-2021.

Hejzlar, J., Dubrovský, M., Buchtele, J., & Růžička, M. (2003). The apparent and potential effects of climate change on the inferred concentration of dissolved organic matter in a temperate stream (the Malše River, South Bohemia). Sci Total Environ, 310(1–3), 143–152. doi:https://doi.org/10.1016/S0048-9697(02)00634-4.

Hensgens, G., Laudon, H., Peichl, M., Gil, I.A., Zhou, Q., Berggren, M., 2020. The role of the understory in litter DOC and nutrient leaching in boreal forests. Biogeochemistry 149 (1), 87–103. https://doi.org/10.1007/s10533-020-00668-5.

Hirano, T., Jauhiainen, J., Inoue, T., Takahashi, H., 2009. Controls on the carbon balance of tropical peatlands. Ecosystems 12 (6), 873–887. https://doi.org/10.1007/s10021-008-9209-1.

Hongve, D., Riise, G., Kristiansen, J.F., 2004. Increased colour and organic acid concentrations in Norwegian forest lakes and drinking water - A result of increased precipitation? Aquat Sci 66 (2), 231–238. https://doi.org/10.1007/s00027-004-0708-7.

Jackson, R., Banner, J., Jobbágy, E., 2002. Ecosystem carbon loss with woody plant invasion of grasslands. Nature 277 (July), 623–627. https://doi.org/10.1038/nature00952.

Janssens, I.A., Freibauer, A., Ciais, P., Smith, P., Nabuurs, G., Folberth, G., Schlamadinger, B., Hutjes, R.W.A., Ceulemans, R., Schulze, E., Valentini, R., Dolman, A.J., 2003. Europe' s terrestrial biosphere anthropogenic CO 2 emissions. Science 300 (June), 1538–1542. https://doi.org/10.1126/science.1083592.

Jenkinson, D.S., Coleman, K., 2008. The turnover of organic carbon in subsoils. Part 2. Modelling carbon turnover. Eur J Soil Sci 59 (2), 400–413. https://doi.org/10.1111/j.1365-2389.2008.01026.x.

Jenkinson, D.S., Andrew, S.P.S., Lynch, J.M., Tinker, M.J.G., P. B., 1990. The turnover of organic carbon and nitrogen in soil. Philos Trans R Soc Lond B Biol Sci 329 (1255), 361–368. https://doi.org/10.1098/rstb.1990.0177.

Jobbágy, E.G., Jackson, R.B., 2000. The vertical distribution of soil organic carbon and its relation to climate and vegetation. Ecol Appl 10 (2), 423–436. https://doi.org/10.1890/1051-0761(2000)010[0423:TVDOSO]2.0.CO;2.

Johnson, C.E., Driscoll, C.T., Siccama, T.G., Likens, G.E., 2000. Position and landscape in a northern hardwood watershed ecosystem. Gene 3 (2), 159–184.

Kalbitz, K., Schmerwitz, J., Schwesig, D., Matzner, E., 2003. Biodegradation of soil-derived dissolved organic matter as related to its properties. Geoderma 113, 273–291.

Kalbitz, Karsten, Solinger, S., Park, J.-H., Michalzik, B., Matzner, E., 2000. Controls on the dynamics of dissolved organic matter in soils a review. Soil Sci 165 (4), 277–304.

Kicklighter, D.W., Hayes, D.J., Mcclelland, J.W., Peterson, B.J., Mcguire, A.D., Melillo, J. M., 2013. Insights and issues with simulating terrestrial DOC loading of Arctic river networks. Ecol Appl 23 (8), 1817–1836. https://doi.org/10.1890/11-1050.1.

Kindler, R., Siemens, J., Kaiser, K., Walmsley, D.C., Bernhofer, C., Buchmann, N., Cellier, P., Eugster, W., Gleixner, G., Grũnwald, T., Heim, A., Ibrom, A., Jones, S.K., Jones, M., Klumpp, K., Kutsch, W., Larsen, K.S., Lehuger, S., Loubet, B., Kaupenjohann, M., 2011. Dissolved carbon leaching from soil is a crucial component of the net ecosystem carbon balance. Global Change Biol 17 (2), 1167–1185. https://doi.org/10.1111/j.1365-2486.2010.02282.x.

Labat, D., Goddéris, Y., Probst, J.L., Guyot, J.L., 2004. Evidence for global runoff increase related to climate warming. Adv Water Resour 27 (6), 631–642. https://doi.org/10.1016/j.advwatres.2004.02.020.

Lauerwald, R., Hartmann, J., Ludwig, W., Moosdorf, N., 2012. Assessing the nonconservative fluvial fluxes of dissolved organic carbon in North America. J Geophys Res 117 (G1). https://doi.org/10.1029/2011jg001820.

Lauerwald, R., Regnier, P., Camino-Serrano, M., Guenet, B., Guimberteau, M., Ducharne, A., Polcher, J., Ciais, P., 2017. ORCHILEAK: A New Model Branch to Simulate Carbon Transfers along the Terrestrial-Aquatic Continuum of the Amazon

Basin. Geoscientific Model Development, April, pp. 1–58. https://doi.org/10.5194/gmd-2017-79.

Lauerwald, R., Regnier, P., Guenet, B., Friedlingstein, P., Ciais, P., 2020. How simulations of the land carbon sink are biased by ignoring fluvial carbon transfers: A case study for the Amazon Basin. One Earth 3 (2), 226–236. https://doi.org/10.1016/j.oneear.2020.07.009.

Ledesma, J.L.J., Köhler, S.J., Futter, M.N., 2012. Long-term dynamics of dissolved organic carbon: implications for drinking water supply. Sci Total Environ 432, 1–11. https://doi.org/10.1016/j.scitotenv.2012.05.071.

Lehner, B., Döll, P., 2004. Development and validation of a global database of lakes, reservoirs and wetlands. J Hydrol 296 (1–4), 1–22. https://doi.org/10.1016/j.jhydrol.2004.03.028.

Michalzik, B., Kalbitz, K., Park, J., Solinger, S., Matzner, E., 2001. Fluxes and concentrations of dissolved organic carbon and nitrogen–a synthesis for temperate forests. Biogeochemistry 52, 173–205. http://link.springer.com/article/10.1023/A:1006441620810.

Monteith, D.T., Stoddard, J.L., Evans, C.D., de Wit, H.A., Forsius, M., Høgåsen, T., Wilander, A., Skjelkvåle, B.L., Jeffries, D.S., Vuorenmaa, J., Keller, B., Kopácek, J., Vesely, J., 2007. Dissolved organic carbon trends resulting from changes in atmospheric deposition chemistry. Nature 450 (7169), 537–540. https://doi.org/10.1038/nature06316.

Nakhavali, M., Friedlingstein, P., Lauerwald, R., Tang, J., Chadburn, S., Camino-Serrano, M., Guenet, B., Harper, A., Walmsley, D., Peichl, M., Gielen, B., 2018. Representation of dissolved organic carbon in the JULES land surface model (vn4.4_JULES-DOCM). Geosci Model Dev 11 (2), 593–609. https://doi.org/10.5194/gmd-11-593-2018.

Nakhavali, M., Lauerwald, R., Regnier, P., Guenet, B., Chadburn, S., Friedlingstein, P., 2021. Leaching of dissolved organic carbon from mineral soils plays a significant role in the terrestrial carbon balance. Global Change Biol 27 (5), 1083–1096. https://doi.org/10.1111/gcb.15460.

Nakhavali, M.A., Mercado, L.M., Hartley, I.P., Sitch, S., Cunha, F.V., di Ponzio, R., Lugli, L.F., Quesada, C.A., Andersen, K.M., Chadburn, S.E., Wiltshire, A.J., Clark, D. B., Ribeiro, G., Siebert, L., Moraes, A.C.M., Schmeisk Rosa, J., Assis, R., Camargo, J. L., 2022. Representation of the phosphorus cycle in the joint UK land environment simulator (vn5.5_JULES-CNP). Geosci Model Dev 15 (13), 5241–5269. https://doi.org/10.5194/gmd-15-5241-2022.

Neff, J.C., Asner, G.P., 2001. Dissolved organic carbon in terrestrial ecosystems: synthesis and a model. Ecosystems 4 (1), 29–48. https://doi.org/10.1007/s100210000058.

Nydahl, A.C., Wallin, M.B., Weyhenmeyer, G.A., 2017. No long-term trends in p CO_2 despite increasing organic carbon concentrations in boreal lakes, streams, and rivers. Global Biogeochem Cycles 31 (6), 985–995. https://doi.org/10.1002/2016GB005539.

Peterson, F.S., Lajtha, K.J., 2013. Linking aboveground net primary productivity to soil carbon and dissolved organic carbon in complex terrain. J Geophys Res Biogeo 118 (3), 1225–1236. https://doi.org/10.1002/jgrg.20097.

Piao, S., Friedlingstein, P., Ciais, P., de Noblet-Ducoudré, N., Labat, D., Zaehle, S., 2007. Changes in climate and land use have a larger direct impact than rising CO 2 on global river runoff trends. Proc Natl Acad Sci 104 (39), 15242–15247. https://doi.org/10.1073/pnas.0707213104.

Pregitzer, K.S., Zak, D.R., Burton, A.J., Ashby, J.A., Macdonald, N.W., 2004. Chronic nitrate additions dramatically increase the export of carbon and nitrogen from northern hardwood ecosystems. Biogeochemistry 68 (2), 179–197. https://doi.org/10.1023/B:BIOG.0000025737.29546.fd.

Pschenyckyj, C.M., Clark, J.M., Shaw, L.J., Griffiths, R.I., Evans, C.D., 2020. Effects of acidity on dissolved organic carbon in organic soil extracts, pore water and surface litters. Sci Total Environ 703, 135585. https://doi.org/10.1016/j.scitotenv.2019.135585.

Raymond, P.A., Hartmann, J., Lauerwald, R., Sobek, S., McDonald, C., Hoover, M., Butman, D., Striegl, R., Mayorga, E., Humborg, C., Kortelainen, P., Durr, H., Meybeck, M., Ciais, P., Guth, P., 2013. Global carbon dioxide emissions from inland waters. Nature 503 (7476), 355–359. https://doi.org/10.1038/nature12760.

Regnier, P., Friedlingstein, P., Ciais, P., Mackenzie, F.T., Gruber, N., Janssens, I.A., Laruelle, G.G., Lauerwald, R., Luyssaert, S., Andersson, A.J., Arndt, S., Arnosti, C., Borges, A.V., Dale, A.W., Gallego-Sala, A., Goddéris, Y., Goossens, N., Hartmann, J., Heinze, C., Thullner, M., 2013b. Anthropogenic perturbation of the carbon fluxes from land to ocean. Nat Geosci 6 (8), 597–607. https://doi.org/10.1038/ngeo1830.

Regnier, Pierre, Arndt, S., Goossens, N., Volta, C., Laruelle, G.G., Lauerwald, R., Hartmann, J., 2013a. Modelling estuarine biogeochemical dynamics: from the local to the global scale. Aquat Geochem 19 (5–6), 591–626. https://doi.org/10.1007/s10498-013-9218-3.

Rind, D., Goldberg, R., Hansen, J., Rosenzweig, C., Ruedy, R., 1990. Potential evapotranspiration and the likelihood of future drought. J Geophys Res 95 (D7), 9983–10004. https://doi.org/10.1029/JD095iD07p09983.

Rizinjirabake, F., Pilesjö, P., Tenenbaum, D.E., 2019. Dissolved organic carbon leaching flux in a mixed agriculture and forest watershed in Rwanda. J Hydrol Region Stud 26 (February), 100633. https://doi.org/10.1016/j.ejrh.2019.100633.

Sitch, S., Smith, B., Prentice, I.C., Arneth, A., Bondeau, A., Cramer, W., Kaplans, J.O., Levis, S., Lucht, W., Sykes, M.T., Thonicke, K., Venevsky, S., 2003. Evaluation of ecosystem dynamics, plant geography and terrestrial carbon cycling in the LPJ dynamic global vegetation model. Global Change Biol 9, 161–185. https://doi.org/10.1046/j.1365-2486.2003.00569.x.

Sitch, S., Friedlingstein, P., Gruber, N., Jones, S.D., Murray-Tortarolo, G., Ahlström, A., Doney, S.C., Graven, H., Heinze, C., Huntingford, C., Levis, S., Levy, P.E., Lomas, M., Poulter, B., Viovy, N., Zaehle, S., Zeng, N., Arneth, A., Bonan, G., Myneni, R., 2015. Recent trends and drivers of regional sources and sinks of carbon dioxide. Biogeosciences 12 (3), 653–679. https://doi.org/10.5194/bg-12-653-2015.

Stoddard, J.L., Kahl, J.S., Deviney, F.A., DeWalle, D.R., Driscoll, T.C., Herlihy, A.T., Kellogg, J.H., Murdoch, P.S., Webb, J.R., Webster, K.E., 2003. *Responses of Maine Surface Waters to the Clean Air Act Amendments of 1990* (Issue January). Environmental Protection Agency, U.S.

Tian, H., Yang, Q., Najjar, R.G., Ren, W., Friedrichs, M.A.M., Hopkinson, C.S., Pan, S., 2015. Anthropogenic and climatic influences on carbon fluxes from eastern North America to the Atlantic Ocean: A process-based modeling study. J Geophys Res Biogeo 120 (4), 752–772. https://doi.org/10.1002/2014JG002760.

Tian, H., Yao, Y., Li, Y., Shi, H., Pan, S., Najjar, R.G., Pan, N., Bian, Z., Ciais, P., Cai, W., Dai, M., Friedrichs, M.A.M., Li, H., Lohrenz, S., Leung, L.R., 2023. Increased terrestrial carbon export and CO 2 evasion from global inland waters since the preindustrial era. Global Biogeochem Cycles 37 (10). https://doi.org/10.1029/2023GB007776.

Tranvik, L.J., Downing, J.A., Cotner, J.B., Loiselle, S.A., Striegl, R.G., Ballatore, T.J., Dillon, P., Finlay, K., Fortino, K., Knoll, L.B., Kortelainen, P.L., Kutser, T., Larsen, S., Laurion, I., Leech, D.M., Mccallister, S.L., Mcknight, D.M., Melack, J.M., Overholt, E., Weyhenmeyer, G.A., 2009. Lakes and reservoirs as regulators of carbon cycling and climate. Limnol Oceanogr 54 (1), 2298–2314. https://doi.org/10.4319/lo.2009.54.6_part_2.2298.

Van Oost, K., Verstraeten, G., Doetterl, S., Notebaert, B., Wiaux, F., Broothaerts, N., Six, J., 2012. Legacy of human-induced C erosion and burial on soil–atmosphere C exchange. Proc Natl Acad Sci 109 (47), 19492–19497. https://doi.org/10.1073/pnas.1211162109.

Wang, Z., Hoffmann, T., Six, J., Kaplan, J.O., Govers, G., Doetterl, S., Van Oost, K., 2017. Human-induced erosion has offset one-third of carbon emissions from land cover change. Nat Clim Chang 7 (5), 345–349. https://doi.org/10.1038/nclimate3263.

Worrall, F., Gibson, H.S., Burt, T.P., 2008. Production vs. solubility in controlling runoff of DOC from peat soils - the use of an event analysis. J Hydrol 358 (1–2), 84–95. https://doi.org/10.1016/j.jhydrol.2008.05.037.

Xiao, X., Li, X., Jiang, T., Tan, M., Hu, M., Liu, Y., Zeng, W., 2019. Response of net primary production to land use and climate changes in the middle-reaches of the Heihe River basin. Ecol Evol 9 (8), 4651–4666. https://doi.org/10.1002/ece3.5068.

Yule, C.M., Gomez, L.N., 2009. Leaf litter decomposition in a tropical peat swamp forest in peninsular Malaysia. Wetlands Ecol Manage 17 (3), 231–241. https://doi.org/10.1007/s11273-008-9103-9.

Zhang, H., Lauerwald, R., Ciais, P., Van Oost, K., Guenet, B., Regnier, P., 2022b. Global changes alter the amount and composition of land carbon deliveries to European rivers and seas. Commun Earth Environ 3 (1). https://doi.org/10.1038/s43247-022-00575-7.

Zhang, Q., Jia, X., Wei, X., Shao, M., Li, T., Yu, Q., 2020. Total soil organic carbon increases but becomes more labile after afforestation in China's loess plateau. For Ecol Manage 461 (January), 117911. https://doi.org/10.1016/j.foreco.2020.117911.

Zhang, X., Jin, J., Zeng, X., Hawkins, C.P., Neto, A.A.M., Niu, G., 2022a. The compensatory CO 2 fertilization and stomatal closure effects on runoff projection from 2016–2099 in the Western United States. Water Resour Res 58 (1). https://doi.org/10.1029/2021WR030046.

Zhou, W.J., Lu, H.Z., Zhang, Y.P., Sha, L.Q., Allen Schaefer, D., Song, Q.H., Deng, Y., Deng, X.B., 2016. Hydrologically transported dissolved organic carbon influences soil respiration in a tropical rainforest. Biogeosciences 13 (19), 5487–5497. https://doi.org/10.5194/bg-13-5487-2016.