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Biological lability of terrestrial DOM increases CO2 outgassing across Arctic shelves

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Response to Reviewers:	Answers to the reviewer/editor comments and criticisms have been attached in a dedicated file

Biological lability of terrestrial DOM increases CO_2 outgassing across Arctic shelves

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Abstract

Arctic shelf seas receive greater quantities of river runoff than any other ocean region and are experiencing increased freshwater loads and associated terrestrial matter inputs since recent decades. Amplified terrestrial permafrost thaw and coastal erosion is exposing previously frozen organic matter, enhancing its mobilization and release to nearshore regions. Changing terrestrial dissolved organic matter (terr-DOM) loads and composition may alter shelf primary productivity and respiration, ultimately affecting net regional CO_2 air-sea fluxes. However, the future evolution of Arctic Ocean climate feedbacks are highly dependent upon the biological degradability of terr-DOM in coastal waters, a factor often omitted in modelling studies. Here, we assess the sensitivity of CO_2 air-sea fluxes from East Siberian Arctic Shelf (ESAS) waters to changing terr-DOM supply and degradability using a biogeochemical model explicitly accounting for bacteria dynamics and shifting terr-DOM composition. We find increasing terr-DOM loads and degradability trigger a series of biogeochemical and ecological processes shifting ESAS waters from a net sink to a net source of CO_2, even after accounting for strengthening coastal productivity by additional land-derived nutrients. Our results suggest that future projected inputs of labile terr-DOM from peat and permafrost thaw may strongly exacerbate the CO_2 efflux from the Arctic shelf sea, causing currently unquantified positive feedbacks to climate change.

1. Introduction

Pan-Arctic river discharge to the Arctic Ocean has increased over recent decades (Peterson et al. 2002; Haine et al. 2015) with climate model simulations forecasting this to continue and potentially accelerate during the 21st century (Ahmed et al. 2020; Wang et al. 2021). Hydrologic models informed with climate projections estimate increases of ~25 to 50 % in freshwater discharge to the

Laptev and East Siberian Shelf by 2100 (Wang et al. 2021). Ongoing terrestrial permafrost thaw and enhanced rates of coastal erosion in response to climate warming will additionally alter the source and loads of dissolved organic matter (DOM) exported from watersheds to coastal waters (Wang et al. 2021; Frey and McClelland 2008). An intensifying hydrologic cycle will therefore cause greater quantities of terrestrial dissolved organic carbon (terr-DOM) to reach coastal shelf waters, with changes in its overall composition due to redistribution of aged peat and permafrost derived materials (Mann et al. 2022; McGuire et al. 2009; Vonk and Gustafsson, 2013).

The shallow Laptev Sea (50 m) receives the greatest quantities of freshwater across the ESAS (~ 745 km^3 yr^{-1}), predominantly supplied by the Lena River (566 km^3 yr^{-1}; Cooper et al. 2008). Shelf waters of the Laptev Sea are characterized by high DOM concentration (up to 500 μM, Juhls et al 2019) comprised mostly of terr-DOM, evidenced by stable isotope carbon composition and ratios consistent with terrestrial sources (Alling et al. 2011; Salvadó et al. 2016). Arctic shelf DOM also comprises DOM derived from coastal erosion and marine production, and is influenced by the contrasting fate (export, mineralization, flocculation, and burial) of different DOM fractions (e.g., non-humic vs. humic DOC) across the shelf. Overall losses of up to 10-20 % of the riverine DOC pool have been estimated across the Lena River estuary before export onto the ESAS shelf, likely due to processes such as photodegradation, flocculation and sedimentation (Alling et al. 2011; Gustafsson et al. 2000) which mainly remove humic DOM fractions in the lower salinity zone (between 4 and 6 psu, Forsgren et al. 1996; Eckert and Sholkovitz, 1976; Stubbins et al. 2016). By contrast, moving offshore, where salinity is higher, DOC losses in the non-humic fraction of the DOM pool become increasingly more important, indicating a dominant role for bacteria in degrading terr-DOM in the outer shelf regions (Anderson et al. 2019, Alling et al. 2010; Amon & Benner, 2003; Lobbes et al. 2000).

The fate and impact of terr-DOM on shelf biogeochemistry is, therefore, influenced by the capacity of heterotrophic bacteria to utilize terr-DOM to fulfil their carbon and nutrient requirements i.e**.,** on the biological lability of terr-DOM, a key yet highly uncertain parameter (Alling et al. 2010, Holmes et al. 2008, Manizza et al. 2009). Despite this, the sensitivity of air-sea CO_2 flux estimates to contemporary or future terr-DOM biological degradability has yet to been addressed. However, mounting evidence suggests future changes to coastal terr-DOM composition with permafrost thaw and reduced freshwater residence times (Mann et al. 2022) may increase the biological lability of terr-DOM promoting enhanced DOC losses (Catalan et al. 2016; Mann et al. 2015; Vonk et al. 2013) and supply of nutrients to support coastal productivity across Arctic shelves (Theraar et al. 2019, 2021). Here, we use a complex marine biogeochemical model – the European Regional Seas Ecosystem Model (ERSEM, Butenschon et al. 2016) specifically augmented with a new formulation accounting for terr-DOM to assess the impact of terr-DOM concentration and its biological reactivity on the CO_2 exchange between the ocean and the atmosphere.

2. Materials and Methods

2.1 The biogeochemical model

ERSEM is a biomass and functional type based marine biogeochemical model describing carbon and nutrient cycling within the planktonic and benthic environment. Heterotrophic prokaryotes

(DOM consumers) are modelled through a single functional type, hereafter generically called "bacteria". DOM-bacteria interactions, including terr-DOM are summarized in Fig 1.

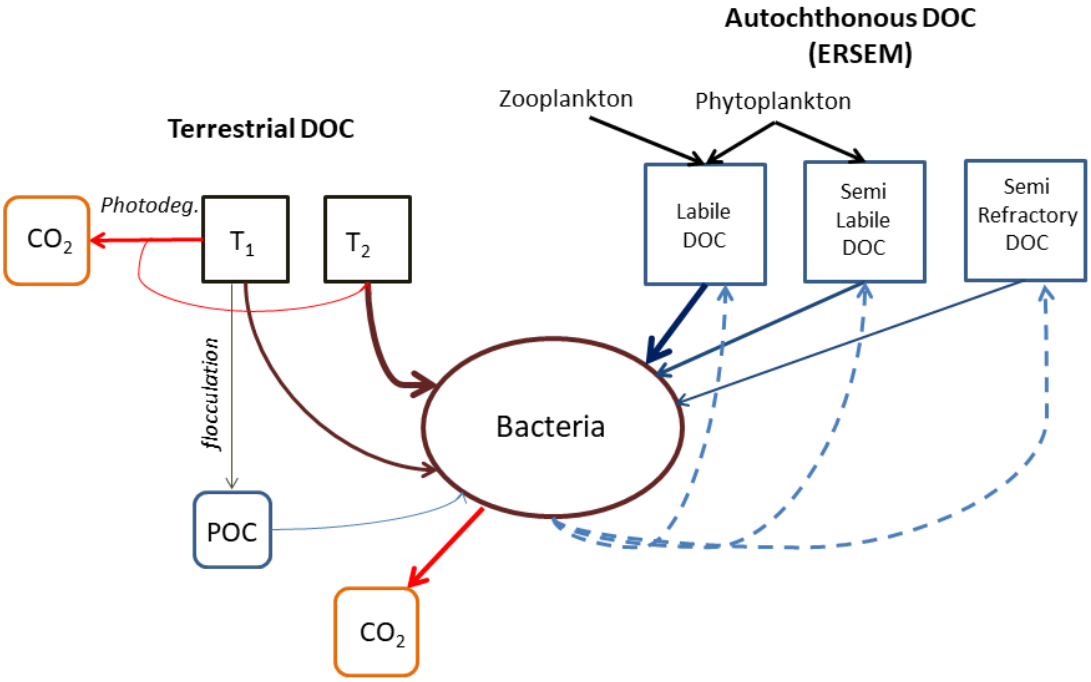


Fig 1. Model schematic describing the interactions between bacteria and organic carbon. On the right side is described the dynamics of autochthonous DOC as modelled in ERSEM (Butenschön et al., 2016). On the left side is described the addition of terr-OC modelled based on Anderson et al. (2019)

ERSEM is fully described in Butenschön et al. (2016), which includes a detailed description of model assumptions and mathematical equations. The current implementation is based on the Butenschön et al. (2016) description with 4 phytoplankton and 3 zooplankton functional groups. Here we limit our description to the equations accounting for terr-DOM which have been newly developed in this work partially based on Anderson et al. (2019) and to bacterial respiration which is a key variable for the purpose of this study. terr-DOM is described through two state variables: T_1 and T_2 (both given in mg C m^{-3}). T_1 represents a coloured, light-absorbing 'humic' fraction which is susceptible to both microbial and photochemical degradation. T_2 by contrast, accounts

for the non-light absorbing 'non-humic' fraction and is susceptible only to degradation by bacteria. Following Anderson et al. (2019), the T_1 fraction is also assumed to be prone to flocculation processes. terr-DOM is assumed to have a fixed C:N and C:P molar ratio equal to 22 and 950, respectively (Cauwet and Sidorov 1996; Kutscher et al. 2017; Sanders et al. 2022). Model parameters, other than those explicitly described here, are taken from Butenschön et al. (2016).

The evolution of terr-DOM can be summarized as:

$$\frac{dT_{1,2}}{dt} = \boldsymbol{uptake} - \boldsymbol{photodeg} - \boldsymbol{flocc} - \boldsymbol{diffusion} - \frac{1}{R}\left(\boldsymbol{T_{1,2}} - \boldsymbol{T_{obs}}\right) \quad (1)$$

Where $\boldsymbol{T_{1,2}}$ is the terr-DOM fraction concentration, $\boldsymbol{uptake}$ is the bacterial consumption (see below) and $\boldsymbol{flocc, photodeg\ and\ diffusion}$ are the terr-DOM changes due to flocculation, photodegradation and turbulent diffusion, respectively. R is a relaxation constant (10 days). The

purpose of R is to enable the introduction of a source of terr-DOM ($\boldsymbol{T_{obs}}$, driven by the seasonality of the Lena outflow) in our 1D model setup where horizontal advection is not resolved

The term $\boldsymbol{uptake}$ (mg C m^{-3} d^{-1}) is equal to:

$$\boldsymbol{uptake} = \frac{\boldsymbol{\alpha_{1,2} \cdot T1,2}}{\boldsymbol{Total\ substrate}} \cdot \boldsymbol{min(Pot, Total\ substrate)} \quad (2)$$

Where $\boldsymbol{Pot}$ (mg C m^{-3} d^{-1}) is the potential bacterial uptake (i.e. the uptake under non-limiting carbon conditions) and is given by:

$$\boldsymbol{Pot = r \cdot B \cdot T^{10} \cdot O2lim} \quad (3)$$

Where $\boldsymbol{r}$ (d^{-1}) is the max specific uptake rate of bacteria, $\boldsymbol{B}$ is the bacterial concentration (mg C m^{-3}) and $\boldsymbol{T^{10}}$ and $\boldsymbol{O2lim}$ are adimensional functions describing temperature and O_2 dependency, respectively (Butenschön et al. 2016). $\boldsymbol{\alpha_{1,2}}$ are non-dimensional parameters describing the degree of lability of T_1 and T_2 with respect to the degradability of the labile DOC fraction ($\boldsymbol{\sigma}$, 1 d^{-1}, Butenschön et al. 2015, Polimene et al. 2006).

$\boldsymbol{Total\ substrate}$ (mg C m^{-3} d^{-1}) is the sum of all the carbon sources available to bacteria multiplied by the relative lability coefficient $\boldsymbol{\alpha_n}$ and $\boldsymbol{\sigma}$:

$$\boldsymbol{Total\ substrate = T_1 \cdot \alpha_1 \cdot \sigma + T_2 \cdot \alpha_2 \cdot \sigma + \sum_1^n DOM_n \cdot \alpha_n \cdot \sigma + \sum_1^m POM_m \cdot \alpha_m \cdot \sigma} \quad (4)$$

Where DOM and POM are the dissolved and particulate organic matter produced by the marine planktonic system respectively, and $\boldsymbol{\alpha_n}$ and $\boldsymbol{\alpha_m}$ the relative lability coefficients (Butenschon et al

2016). The number of DOM and POM fractions (n and m) described in ERSEM map onto the different functional groups considered (Butenschon et al. 2016).

Note that if $\boldsymbol{Total\ substrate} < \boldsymbol{Pot_{upt}}$, $\boldsymbol{\alpha_{1,2}}$ is a first order degradation rate of $T_{1,2}$.

The term $\boldsymbol{photodeg}$ (mg C m^{-3} d^{-1}) is only applied to T1 and is given by:

$$\boldsymbol{photodeg} = \boldsymbol{\varphi} \cdot \frac{\boldsymbol{I}}{\boldsymbol{I_{ref}}} \cdot \boldsymbol{T1} \quad (5)$$

Where $\boldsymbol{\varphi}$ is the reference photodegradation rate (assumed to be 0.03 d^{-1}, Mann et al. 2012), $\boldsymbol{I}$ the incident irradiance (W m^{-2}) and $\boldsymbol{I_{ref}}$ the reference irradiance assumed to be 130 W m^{-2}. According to Anderson et al. (2019), byproducts of photodegradation are partially directed into the inorganic carbon and nutrient pools, while the remaining fraction (0.2) is redirected into the T2 component. It should be stressed that we assumed photodegradation to be a function of the total irradiance (Ultraviolet or UV light is not modelled) and this could lead to an overestimation of this process as UV absorption by water is significantly higher than at larger wavelengths. However, since photodegradation parameters were not altered during our experiments, this does not affect the quality of our results. Loss due to flocculation (which only applies to T1) is described by the term $\boldsymbol{flocc}$ (mg C m^{-3} d^{-1})

$$\boldsymbol{flocc} = \boldsymbol{\vartheta} \cdot \boldsymbol{e}^{-\frac{(\ln \boldsymbol{S} - \boldsymbol{S_x})^2}{2 \cdot \boldsymbol{\beta}^2}} \cdot \boldsymbol{T1^2} \quad (6)$$

Where $\boldsymbol{S}$ is salinity in psu, $\boldsymbol{\vartheta}$ is the maximum flocculation rate [$2 \cdot 10^{-6} \cdot d^{-1}$ (mg C)$^{-1}$, Anderson et al., 2019], $\boldsymbol{S_x}$ is the ln of salinity at which flocculation is maximum (~2 psu) and $\boldsymbol{\beta}$ is the parameter

of the bell-shaped curve. We have used $\boldsymbol{\beta}$ = 1.35**,** implying that the max flocculation rate decreases by one order of magnitude at 35 psu.

Bacterial respiration ($\boldsymbol{RESP}$**,** mg C m^{-3} d^{-1}) is given by the sum of two terms describing activity and rest respiration, respectively:

$$\boldsymbol{RESP} = \boldsymbol{uptake} \cdot [\mathbf{1} - \boldsymbol{Eff} - \boldsymbol{EffO2} \cdot (\mathbf{1} - \boldsymbol{O2lim})] + \boldsymbol{R_r} \cdot \boldsymbol{B} \cdot \boldsymbol{T^{10}} \quad (7)$$

Where $\boldsymbol{Eff}$ (unitless) is the maximal bacterial growth efficiency (0.6, Butenschön et al. 2016), $\boldsymbol{EffO2}$ the bacterial growth efficiency under oxygen limitation (0.2, Butenschön et al. 2016) and $\boldsymbol{R_r}$ the daily carbon specific rest respiration rate (0.1 d^{-1}, Butenschön et al. 2016)

2.2 Model physical set up

We coupled the model described above with a one-dimensional hydrodynamic model, the General Ocean Turbulence Model (Burchard et al. 1999) and implemented it at the shelf-ocean break of the Siberian Laptev Sea (Lat 75, Lon 130, depth ~38 m). This site is well documented to receive significant quantities of terr-DOM from the Lena River and coastal erosion (Bauch et al. 2013; Juhls et al. 2019).

The model was forced with tidal data (Egbert and Svetlana 2002) and reanalysis meteorological data, including net short wave radiation [hourly ERA-5, generated using Copernicus Climate Change Service information (2018); Hersbach et al. (2018)] and initialized with vertically uniform

values of nutrients (mmol m^{-3} of NO_3, PO_4 and SiO_2) estimated from literature (NO_3= 4, PO_4=0.8 and SiO_2=8, Kattner et al. 1999; Sorokin and Sorokin 1996; Bauch and Cherniavskaia 2018).

To reproduce the seasonal water column structure evolution, temperature and salinity profiles were constrained between artificially created summer (August) and winter (January) profiles (Fig S2 and S3). Summer temperature and salinity profiles range from 4 degrees and 10 psu, respectively at the surface to -1.5 degrees and 34 psu at the bottom (Bauch et al. 2013) while winter values were constant through the water column and equal to the summer bottom values (i.e. -1.5 and 34 psu, respectively). These profiles were linearly interpolated to generate seasonal time series which were assimilated by the model. Resulting temperature and salinity values, averaged over the light season, were consistent with the data reported in Semiletov et al. (2016) (SI, Fig S1 and S2).

Light extinction through the water column is dependent on the concentrations of organic particulates (Butenschön et al. 2016) and the coloured fraction of terr-DOM (T1). Light absorption by inorganic particles (which are not explicitly modelled) has been simulated by using the mass specific light absorption for inorganic suspended solid matter reported in Butenschön et al. (2016) and a concentration of 800 mg m^{-3}. The latter was estimated from the total suspended matter measured in the proximity of the model location (~1 g m^{-3}, Wegner et al. 2003) assuming a 20% contribution of living and detrital organic particles. T_1 has been added to the light absorbing substrates assuming a specific absorption coefficient of 0.0002 (m^2 mg C^{-1}). This value is at the lower range of those reported in Matsuoka et al. (2014) for the ESAS. At the lower boundary of the water column, a simple remineralization closure is applied (Polimene et al. 2014). According

to this, sinking organic particles are exported from the water column and re-injected into the bottom waters as dissolved nutrients and inorganic carbon.

Ice coverage reduction due to global warming is expected to affect PP in the Arctic Seas (Lewis et al. 2020) with potential consequence on CO_2 fluxes. To account for this, we used a conservative approach by not considering ice coverage in the model setup. This implies model phytoplankton experience the longest productive season theoretical possible i.e. coinciding with the light season. The light season was here defined based on modelled non-zero values of daily photosynthetically active radiation simulated from March to October. Simulations were run for 9 years (2010-2018) to check for model stability (i.e. absence of drift in state variables). After 2 years of simulation, a stable repeating cycle for the main biogeochemical variables was achieved and the year 2012 was taken for the sensitivity exercise. The physical setup of the model has been kept constant in all the scenarios tested.

2.3. Terr-DOM scenarios

The starting terr-DOM concentration (present day scenario) was estimated from late summer DOC observations (on average ~200 μmol L^{-1}, Juhls et al. 2019) and assuming a 60% terr-DOM content (Kattner et al. 1999). Spring and Winter terr-DOM concentrations were then estimated from the seasonal evolution of the Lena River DOC discharge (Cauwet and Sidorov, 1996, Juhls et al. 2020). terr-DOM loads during May to June and over winter months (January-March) are approximately double and half (respectively) that of late summer concentrations (August). These values were linearly interpolated to construct a terr-DOM seasonal time series ($\boldsymbol{T_{obs}}$) which was assimilated by the model into the T_1 and T_2 state variables (50% each) as described in eq. 1. Present day terr-DOM fluxes (SI, Fig S3) were increased by 25, 50, 75 and 100 % covering the range of

increases estimated for Arctic Shelf Seas (20-46%, Frey and Smith 2005; Frey et al. 2007) and model scenarios (up to 100%, Terhaar et al. 2019).

Terr-DOM biological lability is expressed as life time i.e. the time by which a terr-DOM pool [X] is degraded to a value equal to [X]/e (Hansell 2013). A range of life time parameters were used based upon decomposition rates reported in literature: 0.7 years (river and estuarine water incubations; Holmes et al. 2008; Vonk et al. 2013), 3.3 years (field observations; Alling et al. 2010) and 10 years (modelling estimate; Manizza et al. 2009). To examine more extreme future changes in terr-DOM lability (e.g. enhanced permafrost thaw; Mann et al. 2022), we widened the life times examined to include 0.3 and 20 years. The inverse of these life times (degradation rates) have been used as input values for the model parameters $\boldsymbol{\alpha_1}$ and $\boldsymbol{\alpha_2}$. Total terr-DOM is assumed to be composed by the same amount of T_1 and T_2 (i.e. 50-50%) while, according to Anderson et al. (2019), T_2 is considered to be ~ three times more labile than T_1. The values of $\boldsymbol{\alpha_1}$ and $\boldsymbol{\alpha_2}$ used are: $\boldsymbol{\alpha_1}$=[0.004, 0.002, 0.0004, 0.0001, 0.00005] and $\boldsymbol{\alpha_2}$ = [0.012, 0.006, 0.0012, 0.0003, 0.00015].

The average of each pair of these values corresponds to the life times described above (from 0.3 to 20 years).

The model was run under four configurations each combining the terr-DOM concentrations and labilities described above (i.e. up to 25 runs, SI Table S1):

1) Core experiment with parameters and initial conditions as above

2) Experiment S1 assuming terr-DOM was totally refractory to bacteria (i.e. $\boldsymbol{\alpha_1}$ and $\boldsymbol{\alpha_2}$ set to zero)

3) Experiment S2 with doubled nutrient initial conditions (nitrate, phosphate and silica)

4) Experiment S3 with double N and P content in terr-DOM (i.e. with double N:C and P:C ratios).

Model performance in the core experiment was assessed by comparing simulated PP with satellite and field estimates presented in literature (SI, Fig S4).

2.4. Model caveats and limitations

The presented modelling approach has caveats and limitations mainly due to the simulation of the physical features of the system. The effect of climate changes on the physical structure of the Arctic shelf is likely to be complex (Timmermans and Marshall 2020; Arthum et al. 2020) and our theoretical model setup was not meant to reproduce such complexity. Consequently, the simulation of the biogeochemical and ecological dynamics we have highlighted is quantitatively affected by the uncertainty associated with our simplified physical setup. However, we stress that our goal was to investigate potential trends in CO_2 fluxes rather than providing quantitative estimates**.**

3. Results and Discussion

Air-sea CO_2 fluxes as a function of terr-DOM concentration and life time were examined using our model (Fig 2). In an idealized water column as here, we could expect a CO_2 balance close to zero in the absence of allochthonous DOM due to the close interdependence and balance between CO_2 consumption (driven by primary productivity) and production driven by community respiration. Positive, seasonally averaged air to sea CO_2 fluxes (i.e. CO_2 uptake) are possible representing produced carbon that is buried in sediments and/or respired over longer time scales. Addition of terr-DOM in such a system should alter this balance potentially leading to dissolved inorganic carbon accumulation in the upper part of the water column.

Assuming initial conservative life time estimates of 10 - 20 years and present day terr-DOM supply, our model estimates a small annual CO_2 uptake (~ 4 mmol CO_2 m^{-2} d^{-1}) in good agreement with recent estimates for the Arctic basin (4 ± 4 mmol CO_2 m^{-2} d^{-1}, Yasunaka et al. 2016). Air-sea CO_2 fluxes were sensitive to both increased terr-DOM supply and/ or reductions in average life times (Fig 2). Increased terr-DOM supply alone only shifted shelf waters toward a net CO_2 source under the doubled scenario (+100% terr-DOM) when life times were higher (3.3, 10 and 20 years). However, air-sea fluxes were highly responsive to terr-DOM life times, with shelf waters shifting to net CO_2 sources under all terr-DOM supply scenarios- including under present

day terr-DOM loads, when shorter life times of 0.7 and 0.3 years were examined. Concurrent changes in both terr-DOM load and life times, caused more rapid shifts toward net CO_2 emissions.

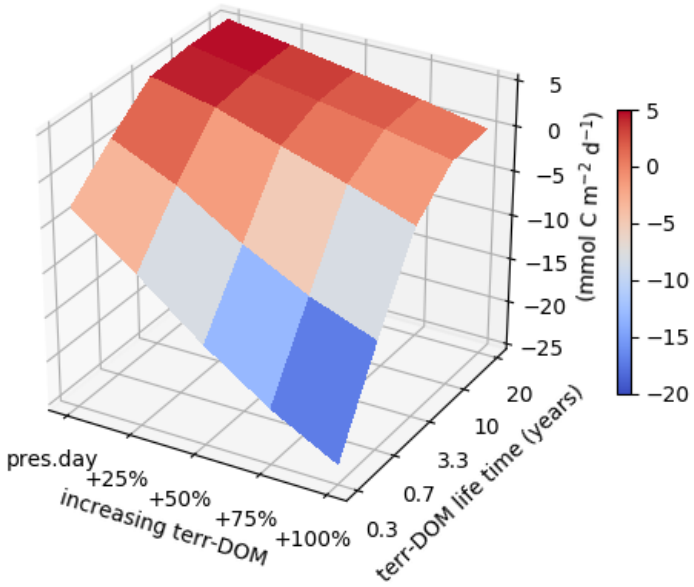


Fig 2. CO_2 air-to-sea flux as function of terr-DOM concentration and life time. Positive values indicate CO_2 sinks, and negative values net sources. Values have been averaged over the light season.

Increased CO_2 air sea emissions were proportional to increased bacterial terr-DOM uptake, which in turn varied across the terr-DOM scenarios tested (Fig 3a). Our model indicates that < 10% of total DOC uptake is subsidized by terr-DOM when life times > 3.3 yrs, whereas terr-DOM contributed up to 60% of the DOC assimilated when life times were < 1 year (Fig 3a). Increased future terr-DOM subsidies to marine bacteria may therefore be expected to drive greater quantities of CO_2 production through respiration (del Giorgio and Cole, 1998).

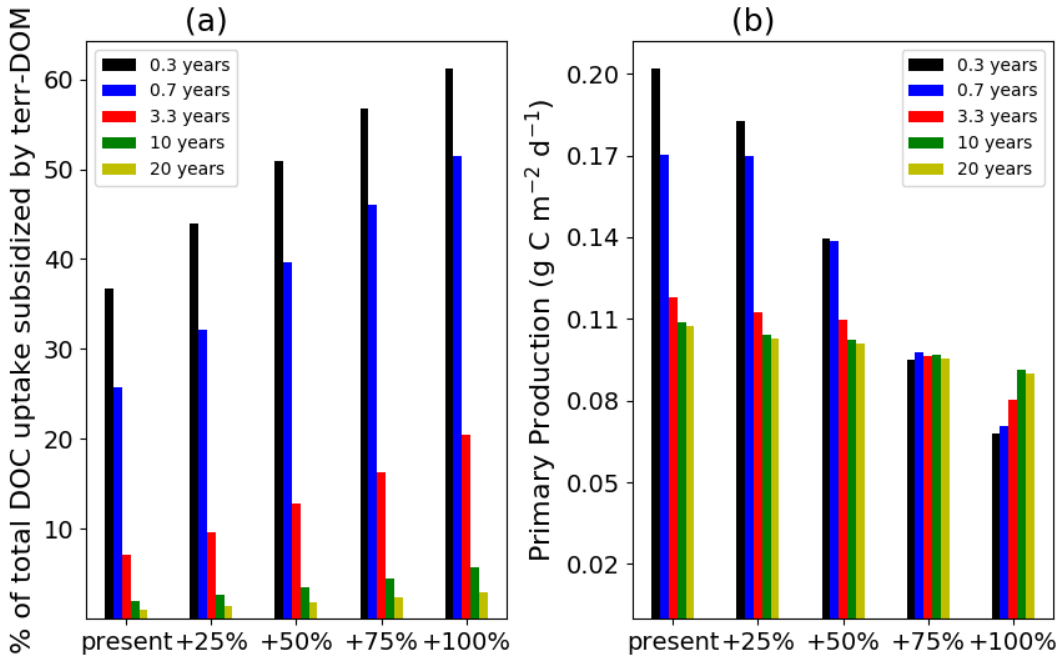


Fig 3. (a) Percentage of bacterial DOC uptake subsidized by terr-DOM as function of terr-DOM concentration (x axis) and lifetime (colors) and (b) PP as a function of terr-DOM concentration (x-axis) and lifetime (colours). Fluxes have been depth-integrated and averaged over the light season.

Future increases in bacterial respiration may be partially balanced by concomitant increases in PP, due to the fertilization effect of nutrients associated with terr-DOM. However, although incorporating the impacts of nutrient enrichment from terr-DOM (Fig S5), our model simulated decreased PP under all scenarios of increasing terr-DOM concentrations (Fig 3b) causing a positive feedback to CO_2 outgassing (Fig 2).

Patterns of decreasing PP were due to the interplay of two factors: light limitation and increased grazing pressure on primary producers. When terr-DOM was assumed to have longer life times (> 10 years), declining PP was primarily caused by reduced light penetration caused by greater terr-DOM light absorption in the water column. Model simulations repeated assuming terr-

DOM was completely refractory (acting as a "light screen" alone) demonstrated PP decreases between 4 to 16% with 25 to 100% increases in terr-DOM supply (experiment S1; Fig 4). By contrast, when terr-DOM lifetimes were shorter, increased grazing pressure on primary producers became progressively more important in all terr-DOM scenarios (Fig 5 and S6). When terr-DOM was more biologically available, bacterial biomass became higher during winter and early spring months (i.e. before the phytoplankton bloom, Fig S7), inducing a shift to a more heterotrophic dominated food chain with relatively high zooplankton concentrations (Fig 5 and Fig. S6). Increased zooplankton control phytoplankton through grazing, reducing PP. As such, the combined effects of light reduction and increased zooplankton pressure resulted in more pronounced declines in PP (up to 60% PP reduction in the +100% terrOC scenario), further decoupling CO_2 production from consumption and causing sharp increases in projected CO_2 outgassing (Fig 2). Notably, for each given terr-DOM concentration, decreasing terr-DOM lifetime

strongly enhanced the CO_2 outgassing (up to 20 times in the +100% scenario with 0.3 years terr-DOM lifetime).

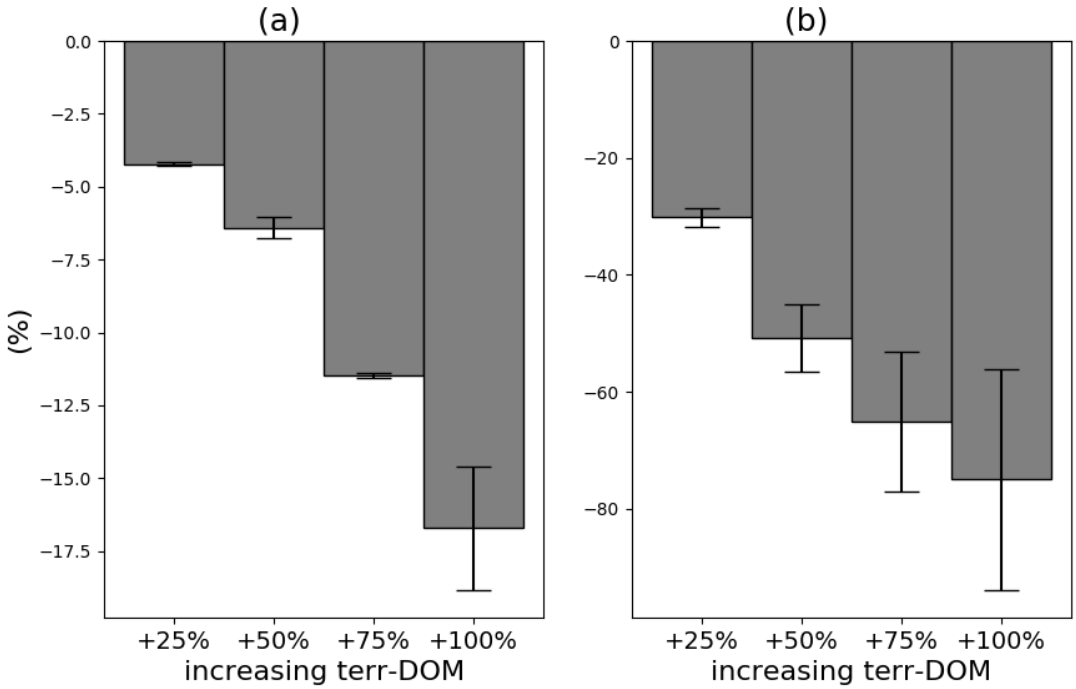
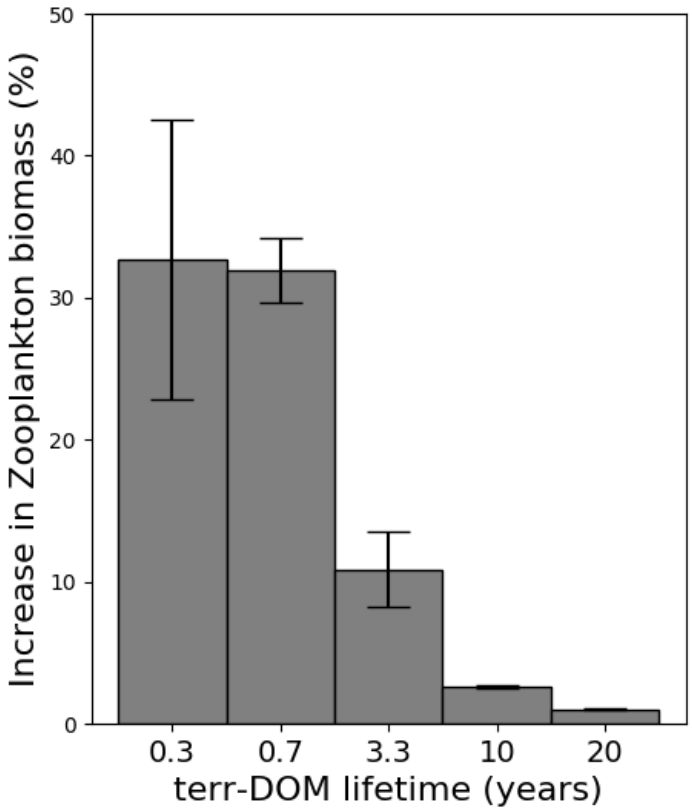


Fig 4. Decrease in average, depth-integrated Primary Production (a) and Photosynthetically Active Radiation simulated at 5-meter depth (b) with respect to the present day scenario in the experiment

299 S1. Uncertainties have been estimated by considering the Coefficients of Variation
300 (CV=(std/mean)*100) in the +100% scenario normalized by the CVs in the present day.



301

302 **Fig 5**. Relative increase in Zooplankton Biomass as function of terr-DOM life times in the
303 +100% scenario with respect to present day. Uncertainties have been estimated by considering
304 the Coefficients of Variation (CV=(std/mean)*100) in the +100% scenario normalized by the
305 CVs in the present day.
306

307 Recent modeling studies suggest nitrogen subsidies from terr-DOM may support up to 51% of
308 contemporary Arctic shelf PP and infer that terrestrial nutrient supplies from land will affect the
309 future evolution of the Arctic Ocean PP (Terhaar et al. 2021). To examine the relative importance
310 of land-derived nutrient supply, we compare PP simulated under present day scenarios assuming
311 high and low terr-DOM life times (Fig 3). Model simulation incorporating high terr-DOM life time
312 (20 years) examine PP when land-derived nutrients are limited in availability for use by primary

producers, whereas low terr-DOM life time (0.3 years) assess PP when nutrients are readily accessible to phytoplankton. In good agreement with Terhaar et al. (2021), simulated PP under short terr-DOM life times (0.3 yr^{-1}) was approximately double of that simulated under 20 years terr-DOM lifetimes (Fig 2b), suggesting terrestrially-derived nutrients remineralised by bacteria and their grazers have potential to sustain up to ~50% of simulated PP under the present day terr-DOM concentrations. However, our model results further suggest that with increasing terr-DOM concentrations, this "fertilization" effect is progressively offset by light limitation imposed by terr-DOM absorption and/or a stimulated zooplankton grazing on phytoplankton populations.

We re-ran our model scenarios (as Fig 1) assuming: 1) a doubling in initial nutrient concentration inputs (experiment S2) and, 2) doubled nutrient content of the terr-DOM pool (e.g. doubled N:C and P:C ratios; experiment S3). Experiment S2 provides insights into nutrient supplies not associated to terr-DOM as, for example, through the additional supply of inorganic nutrients (PO_4, NO_3 and SiO_2) advected from the Atlantic and Pacific Oceans (Lewis et al. 2020). Experiment S3, examines the effect of increased nutrients (organic P and N) within terr-DOM (i.e. assuming more organic nutrients per unit of terr-DOM). Increased nutrient loads only marginally impacted the CO_2 air-sea fluxes in both experiments (Fig 6). However, the response to nutrient enrichments differed between experiments. Ocean CO_2 uptake increased under all scenarios tested in exp. S2

(Fig 6a), whereas increased CO_2 uptake was only simulated at low terr-DOM concentrations and high terr-DOM lifetime in exp. S3 (Fig 6b).

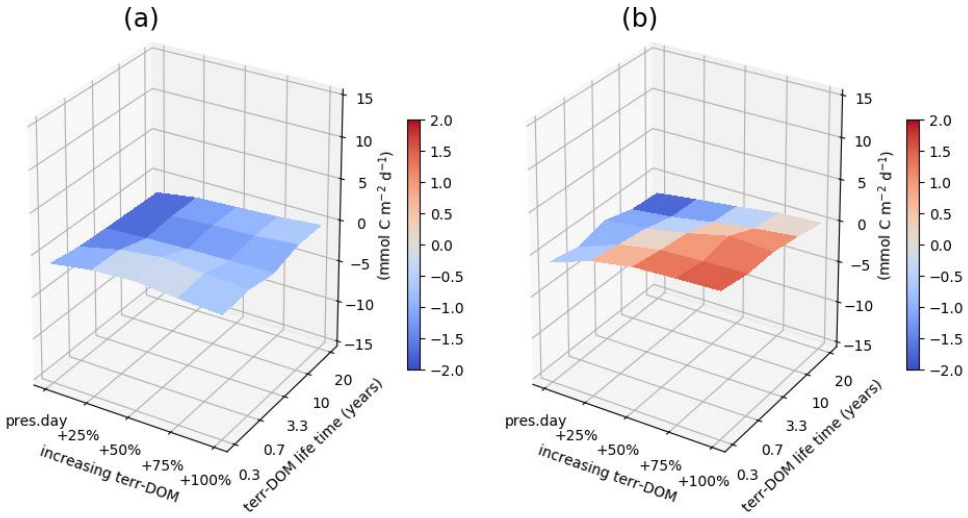


Fig 6. Differences in CO_2 average fluxes between the core experiment (Fig. 1) and, a) Experiment S2 and, b) Experiment S3. Negative differences (blue colour) indicate increased CO_2 ocean uptake, positive values (red colour) increased CO_2 emissions.

In both nutrient enrichment experiments, increases in PP (Fig S7) were partially balanced by increased heterotrophic respiration (bacterial respiration alone increases up to 18%), resulting in limited overall effects on CO_2 uptake (Fig 6 and Fig S8). Large differences between the two experiments were simulated when terr-DOM concentrations and degradability were high; this is due to the dominance of diatoms under these conditions i.e. when smaller phytoplankton groups are top-down controlled by grazers which are enhanced by increased bacteria (Fig S6). Consequently, in exp S3 which provided no SiO_2 enrichment, increasing N and P associated to terr-DOM are less effective in fertilizing phytoplankton since PP is mainly limited by silica.

Together, these results indicate that future increases in nutrients supply or even PP will not necessarily translate to proportional enhancements in net CO_2 Ocean uptake.

Our results question the capacity of the Arctic Ocean to serve as a net sink for atmospheric CO_2 in agreement with prior studies (Semiletov et al. 2016) and may help to explain recent satellite observations indicating decreasing PP in the Laptev Sea over recent decades (Demidov et al. 2020). Contrary to prior studies (e.g. Bates and Mathis 2009), we suggest that Arctic shelf waters are susceptible to shifting from net sinks to net atmospheric sources of CO_2 under future changes in terr-DOM supply and origin. Additional nutrients from land-derived sources, or elsewhere, only marginally offset these trend due to complex concomitant changes in biogeochemical and ecological processes. Our results highlights the need for future studies to incorporate complex biogeochemical models explicitly accounting for bacterial dynamics to simulate emergent properties affecting CO_2 production and consumption in coastal areas.

Terrestrial permafrost thaw and enhanced coastal erosion rates across the Arctic can mobilize peat and permafrost-derived terr-DOM to coastal waters, modifying nutrient and carbon loads and their relative biological degradability (Mann et al. 2015; Vonk et al. 2013). Small subsidies of permafrost-derived terr-DOM to riverine and estuarine waters (~1%) have been shown to result in marked increases in biodegradability rates (20 – 60% dependent on DOM pool), and thus reduced mean life times (Mann et al. 2022). Changes to terr-DOM sources combined with an intensification in Arctic hydrologic cycles (e.g. 25 – 50% freshwater increase to ESAS shelf by 2100, Wang et al. 2021), is therefore likely to deliver greater quantities of more bioavailable terr-DOM to Arctic shelves over coming decades. Declining coastal sea-ice cover will also modify coastal light availability and timing. As our model assumptions implies that ice is absent during the light season, our model estimates already effectively incorporate future ice-free conditions and thus model

simulated PP is likely overestimated with resulting CO_2 fluxes tending to be conservative. To reliably quantify the future evolution of Arctic Ocean climate feedbacks complex biogeochemical models coupled to a three-dimensional hydrodynamic framework will be needed, on which to run climate-scenario simulations. Our study suggests that future inputs of terr-DOM from peat and permafrost thaw may strongly exacerbate net CO_2 efflux from the Arctic shelf causing, currently unquantified, positive feedbacks to global warming.

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Data/code availability

The ERSEM model is freely available at: https://github.com/riquitorres/Lena-1D

Conflict of interests

The authors have no conflicts of interest to declare that are relevant to the content of this article

Ethical approval

Not applicable

Consent to participate

Not applicable

Consent for publication

Not applicable

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629

Dear Editor,

Please find attached a substantially revised version of the manuscript titled: “Biological lability of terrestrial DOM increases CO2 outgassing across Arctic shelves”.

We have carefully addressed all the reviewer’s comments and modified the manuscript according to his/her suggestions. Please note that we have also made a few unsolicited changes to further improve the clarity and readability of the manuscript. For example, we have restructured Fig 4 (Fig 5 in the revised version) and have moved the section “Model caveats and limitations” upward at the end of the model description

Thanks you very much for considering our work for publication in Biogeochemistry Letters

Sincerely,

Luca Polimene

2“this is a stimulating study and interesting for biogeochemists working on C cycling across aquatic systems”.

We thank the reviewer for this positive comment and the constructive suggestions and criticisms which have helped us to improve our manuscript.

Specific comments

L22. Here and throughout the manuscript. The contraction for terrestrial dissolved organic carbon should be Terr-DOC. Otherwise, it is misleading and it can be understood that you are referring to POC+DOC. If that is the case, then keep Terr-OC. Also, consider whether it should be DOC or DOM. In my opinion, you are not just referring to concentration, so it should be DOM. Modify accordingly.

The reviewer makes a good point and accordingly the contraction for terrestrial DOC has been changed to terr-DOM throughout the manuscript.

L 44. What about potential flocculation and sedimentation, wouldn’t that be a potential effect? Clarify.

We agree that flocculation and sedimentation are processes that may reduce the concentration of terr-DOM in the water column. To address this, and later related points in the comments, we significantly restructured the introduction section to clarify that Arctic DOM can undergo a suite of transformations, affecting its fate during transport across the shelf (new lines 48 to 63). We also emphasize how several studies (references added in the revised version), within the region, have demonstrated that contrasting terr-DOM fractions (broadly characterized as “non-humic” and “humic”) undergo contrasting degradation rates by different processes (new lines 58-63). This hopefully improves the readability of the introduction and strengthens our case that future modeling of terr-DOM dynamics requires multiple fractions with different fates (e.g our T1 and T2 fractions).

The great majority of terr-DOM losses to flocculation and subsequent sedimentation has been suggested to take place at lower salinities (< 6 psu) closer to river mouths (e.g. Forsgren et al. 1996, Gustafsson et al. 2000, Alling et al. 2011) and to remove largely humic-like fractions (new lines 56 -58). To more clearly present that our model simulations were conducted further offshore and at higher salinities (10-34 psu, new lines 166-167 and Fig S2), we added a more detailed description at the beginning of the model set up description (Lines 154-158).

L57-58. Also here, this decay only refers to biodegradation? In such a salinity gradient wont flocculation have a relevant role as well?

We agree with the reviewer that flocculation can be an important process for removing terr-DOM from the water column in the presence of strong salinity gradients. We would like to stress that the presented model uses a one dimensional implementation (i.e. only vertical dynamics are represented) meaning that spatial salinity gradients are not represented. Furthermore, we expect flocculation to play a far lesser role at this shelf break site relative to biodegradation. We evidence several studies (new lines 55-63 with references) that conclude DOC losses in non-humic fractions (indicating bacterial degradation) becoming increasingly important on the outer shelf. Please note, that despite our expectations that flocculation rates would be low (Anderson et al. 2019), we still simulated flocculation according to equations 1 & 6. Since here the focus is the interaction between bacteria and terr-DOM, model parameters describing flocculation (Eq. 6) are kept constant throughout scenarios.

L80. Define DOM.

In this revised version DOM has been defined earlier in the text (new line 42)

L87. Is this the only characteristic of T2? Does that imply that it is transparent and not light-absorbing? Improve definition.

We have added more clear definitions for both T1 and T2 (new lines 91-102), using terminology we now introduce and justify in the introduction (i.e. relative fates of humic vs non-humic DOM fractions).

L88. Flocculation is not included in the introduction. Also, algal-derived material, which I guess might be included in T2 is one of the main precursors of transparent exopolymeric particles and thus, of flocs. How do you link this? Clarify here and in the text.

We have now introduced the flocculation processes in the text (see lines 47-, 62). Algal (marine phytoplankton) derived dissolved and particulate material (now mentioned in line 53, see also the new Fig 1 and equation 4) is explicitly modelled in the ERSEM model (Butenschon et al 2015, Polimene et al, 2006). T1 and T2 represent only terrestrial DOM i.e. derived from soils (see Anderson et al., 2019). In the model, only terrestrial material is assumed to flocculate (see the improved definition of T1 and T2, lines 91-102, equation 4 and lines 140-141).

L90. $photo_{deg}$ I found the use of subscripts to be so weird in the formulations used. Usually the subscript refers to a fraction or specificity of the variable. That is not the case in this example. Also, if you take the subscript out of Bac_{upt} or Pot_{upt}, Bacterial or Potential are not variables. Modify accordingly.

We have followed the suggestion of the reviewer and have removed the subscripts in the revised version

L92. total substrate = T1 + T2? Clarify

Total substrate available for bacteria includes T1, T2, DOM and POM derived from the marine planktonic system (i.e. from phytoplankton, zooplankton and bacteria). We have now made this point clear in the text (new lines 121-128, equation 4 and the new Fig 1)

L100. Provide the equation for Total substrate

Equation describing the total substrate available to bacteria has been added (equation 4)

L101. From where did you obtain the degradation rate of labile DOC 1d-1? Provide information.

We have now cited the two papers from which that parameter has been taken (Butenschon et al. 2015, Polimene et al. 2006)

L106. Eff does not have units?

Eff is an efficiency, as such, is a-dimensional. We have clarified this in the text (new line 150)

L109. Photodegradation is a much more complex process that might indeed be poorly estimated here. First, clarify if you are referring to photomineralization or not. If you are, are those previous estimates based on calculations of the apparent quantum yield? if so, clearly estate in which aspects you would be overestimating the process. If UV light is not included, that would lead to an underestimation of the process, wouldn't it?? Do you consider all T1 to absorb on a specific wavelength? What would be

the effect of depth?? AQY has been shown to decrease rapidly when concentrations of colored DOC are important.

We agree that photodegradation is a complex process that can affect the biological degradation of DOC, as well as changing the molecular composition and optical properties of DOM. Few modelling studies represent these processes well largely due to high uncertainty in the apparent quantum yield. This is likely a consequence of heterogeneity in DOM sources and their composition. The approach we used was based on a recently published model (Anderson et al., 2019). According to this formulation, direct photomineralization of T1 (the more humic and aromatic DOM component capable of light absorption) produces CO_2 and dissolved nutrients. In addition, T1 photomineralization also causes the production of a residual DOM fraction – (represented as a production of 20% in T2) which acts as a means of representing photopriming (e.g. Ward et al. 2017 Nat Comm). We added this in the revised text (see lines 133-135).

Since ERSEM does not account for UV radiation (only the total irradiance is simulated) we have assumed that photodegradation is dependent on the total irradiance (SWR). SWR extinction through the water column is calculated starting from the surface value based on the concentration of particulate organic matter, colored terr-DOM (T1) and inorganic sediments. According to this, high phytoplankton biomass and/or colored terr-DOM reduces the light with depth in the water column and thence photo-degradation (as suggested by the reviewer).

SWR penetration through the water column is deeper than just UV radiation. For this reason, the use of the full SWR may lead to an overestimation of the processes. As explained in the text, since photodegradation parameters were not altered in our numerical experiments, this approximation does not affect the quality of our results, which focuses on the relative changes between scenarios.

L116. Flocculation is not mentioned in the introduction. Introduce the different processes there

We have now mentioned flocculation in the introduction (new lines 53 and 56)

L122. so T2 is entirely derived from T1?? By this point of the manuscript, the link between both fractions should have been made clear to the reader. Clarify and see suggestions below on the order of methods.

T2 is provided as terr-DOM input along with T1. In addition, T2 is also enhanced by the photodegradation of T1. We have clarified this point in the revised version (see lines 133-135 and the whole revised description of T1 and T2). We hope this is now clearer in the revised version.

L127. Update this reference and values, there has been much literature on DOM stoichiometry since then

Much additional literature about DOM stoichiometry has been published since the Cauwet and Sidrov paper, however much of these works have continued to largely focus on freshwater/ river ratios, from hundreds of kms upshore. The ratios we use are in a similar range to those recently reported (Kutscher et al. 2017; Sanders et al 2022). We have added theses more recent references to the manuscript (new line 100).

~~)~~

L133. What is “high”? provide numbers. Also, the evidence on why it is considered terrestrial

We have added numbers (500 µM) and explanations on why most of this DOM is considered terrestrial (lines 49-51 in the revised version).

L151. On particulates and dissolved. Modify and see comment above regarding the effect of DOM color on light extinction and photodecay with depth.

We have modified this sentence (see new line 173 in the revised version); see also our previous answer re. photodegradation

L156. What is that 20% based on?

*This percentage has been estimated as follows: starting from the average DOC concentration from Juhls et al 2019 (see also Fig S4) we have considered a POC:DOC ratio of 0.07 (Sanchez-Garcia et al 2011, GBC). We have then assumed a Readfield ratio (106:16:1) for POC:PON:POP and have estimated the total weight of POM in mg m^{-3}. The resulting estimate was roughly 200 mg m^{-3} which is the 20% of the SPM concentration reported in Wegner et al. 2003***.***

L158. Indeed, it is very low. Why is this value selected? If a higher value would have been considered, how would that impact your phorodegradation rate? This should be evaluated. For parameters like this a minimal sensitivity analysis should be shown.

Given the high level of uncertainty associated to these parameters, we have purposely used a conservative approach choosing a value at the lower range of those reported in the literature. Figure 1 below shows that the impact of the absorption parameter is greatest at very high irradiances (> 300 W m-2), with a low absorption parameter leading to an increase in the photolysis flux. In our study, the low absorbance parameter means that the terr-DOC-induced photo-limitation is likely to be underestimated. Therefore photodegradation is likely to be overestimated due to stronger light penetration through the water column (Figure 1). These two effects would: 1) further reduce the PP and 2) further increase the terr-DOM available for bacteria, in both cases reinforcing our conclusions. We argue that this point is already very clear in the manuscript without the need of adding further analyses within the manuscript.

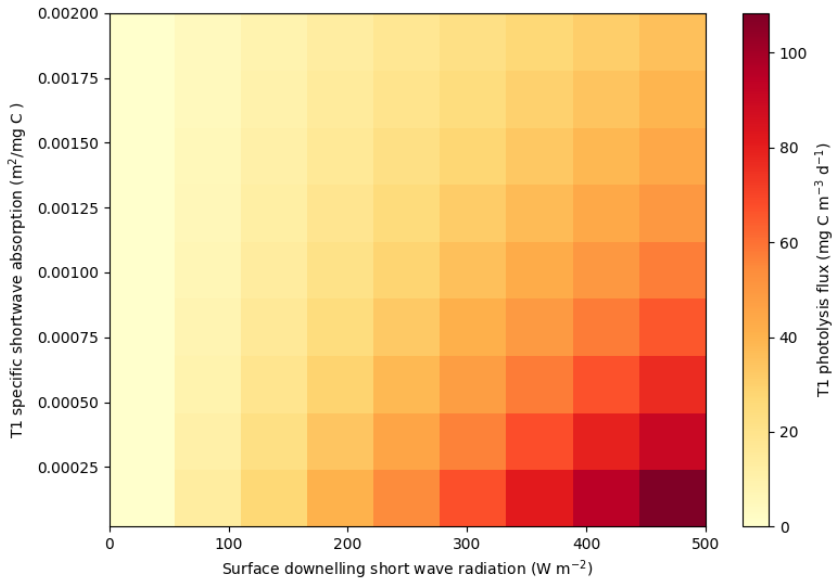

Figure 1: Sensitivity analysis showing the impact of surface downwelling short wave radiation (equivalent of I in equation1) and specific shortwave absorbance of T1 on the photolysis flux. In the model, surface downwelling short wave radiation is prescribed on hourly timescales by an input file, while the absorption parameter is 0.0002 m2/mg C

L174. Provide DOC concentration value range here, to give the reader an idea.

Done, see line 195 in the revised version

L177. Specify months as well and in the line below for late summer.

Done, see lines 198-199 in the revised version

L178. Do you mean the whole Lena river seasonal evolution was interpolated? Clarify.

The concentration of terr-DOM at the site of the model setup was estimated from the late summer values of total DOM in the same area reported in Juhls et al 2019 assuming that 60% of the total DOM is due to terr-DOM (Kettner et al, 1999). Starting from that value, terr-DOM seasonality was estimated by considering the seasonal DOM discharge of the Lena River reported in literature **(***Cauwet and Sidorov, 1996, Juhls et al. 2020***)**. *According to this, organic matter discharge in winter and spring were therefore considered to be half and double (respectively) of the late summer terr-DOM concentration. These three points were interpolated to obtain the seasonal profile displayed in Fig S3.*

Fig.S4. related to that, the result in Fig. S4 looks extremely poor. Is the seasonal time series meaningful in terms of concentration? The trend is almost completely out of the SD of the observations! Could the Lena river trends be provided in that figure? the effect of such uncertainty on the model results should be evaluated.

We thank the reviewer for this question, as it allows us to clarify an important point. The figure shows the model seasonal cycle of terr-DOM (T1+T2, see equation 1) while the data point is the total DOM from which terr-DOM has been estimated (considering that 60% is of terrestrial origin). We have updated the caption for Fig S3 accordingly.

L184. Explain more clearly what is R

R is a relaxation time meaning that it describes how quickly the modelled values of T1 and T2 are adjusted toward Tobs. *Assuming R to be very high would results in no input of terr-DOM on the site*. *This is a common approach in 1D models that do not explicitly model horizontal advection (i.e. external water sources) of variables. We have better described R in the revised version of the manuscript (see lines 108-110)*

L191. Most typically used is half-life. Are you referring to that? Clarify. Additionally, if half-life is 10 years, that would imply that there is no terrestrial OC in the ocean...?

We have used the life-time concept which is widely used to describe marine DOM degradation time scales (Hansel 2013; Polimene et al., 2018). The amount of terr-DOM in the ocean is object of debate, however estimates reported in literature suggest that DOM derived from land is a substantial portion of the Arctic surface ocean (5-33% Opsahl et al. 1999) but is on average very low (<3%) in the Atlantic and Pacific Oceans (Osphal and Benner, 1997)

L197-200. This definition should come much earlier in the manuscript. Start the model description by describing TerrOC and the two pools and then the processes that will affect them and not the other way around.

We have followed the reviewer suggestion and the description of T1 and T2 dynamics have been moved earlier in the manuscript. The whole model description has been revised and made clearer (line 80 onwards)

L204. Provide table summarizing the 25 conditions

A table (Table S1) summarizing the 25 simulations of the core experiment has been added to the supplementary materials

L206. For condition 3) specify which nutrient(s)

Done. See line 224 in the revised version

L210. Provide the details on software, packages, etc. used to develop the model. Provide novel code developed for this contribution. They are a must.

The model code, written in Fortran90, and all the scripts required to run the model (including forcing functions, initial conditions and relative documentation) are freely available at this link https://github.com/riquitorres/Lena-1D (added at the end of the manuscript)

L212. Should be Shelf-sea and sea-air CO2 fluxes (joint estimate)? Clarify.

We thank the reviewer for this comment as we realized that the sentence was actually confusing. We meant the air-sea CO_2 fluxes in the shelf sea. However, since we already stated that the model location is on the shelf we have deleted "Shelf Sea "in the revised version of the manuscript (see line 238).

L215. In the absence of an allochthonous input, shouldn't it be more likely that the balance was as C sink? Clarify

Indeed, as said in the subsequent lines, in absence of external inputs of organic matter, a positive flux of CO_2 (meaning CO2 sink) is possible due to carbon stored in recalcitrant fractions of DOM and/or sediment.

L219. CO_2 accumulation where? As DIC? Pay attention to wording to avoid confusion.

We have now clarified that the C may accumulate as DIC in the upper part of the water column (lines 245-246)

L224. Which are Net CO_2 source conditions? I believe this is another example of what I was talking about regarding wording. Otherwise, if you specifically refer to some initial modelling conditions, which ones? Specify. For that purpose, again, it will come at handy the table I was suggesting. Also, explain why, are the other scenarios considered to be meaningless? Clarify.

We meant the conditions allowing CO_2 source (i.e. a negative balance between CO_2 consumption and CO_2 production). We have made the text clearer (lines 250-257).

L228-229. Does this mean than under any other condition net CO2 outgassing is not simulated, so it is considered to be acting as a sink?Clarify

We have reshaped (and hopefully clarified) the sentence as follow (lines 250-255): "Increased terr-DOM supply alone only shifted shelf waters toward a net CO_2 source under the doubled scenario (+100% terr-DOM) when life times were higher (3.3, 10 and 20 years). However, air-sea fluxes were highly responsive to terr-DOM life times, with shelf waters shifting to net CO_2 sources under all terr-DOM supply scenarios- including under present day terr-DOM loads, when shorter life times of 0.7 and 0.3 years were examined. Concurrent changes in both terr-DOM load and life times, caused more rapid shifts toward net CO_2 shelf emissions".

L236-238. This should be discussed in relation with previous studies on the response of bacteria to changing TerrOC. Add

We have expanded our discussion on the potential future response and influence of our findings on bacterial response. We also place our findings in the context of future change, and a recent paper examining how changing Lena River hydrology and OM source can influence DOM biological reactivity (new lines 358-369).

L239. Won't the Terr-OC inputs have terrestrial bacteria associated? It should be expected, right?

We cannot exclude that this might be the case. However, we do not have specific information about this and the reviewer does not provide any specific reference. We have therefore assumed that once in sea water, fresh water bacteria are taken over by the marine community. Previous studies have shown that transplanting bioreactive DOM into estuarine waters (i.e. including inoculum of terr-DOM bacterial) had no significant impact on DOM turnover rates measured over short periods (e.g. Vonk et al. 2013). Even if terrestrial bacteria were present, we have no evidence to suggest it would fundamentally change DOM dynamics.

L241-242. I fail to see the novelty of this statement, it needs to be discussed under the light of previous studies and the contribution of the present work be made apparent. Add

We agree with the reviewer that that statement per se was not novel and therefore unnecessary. Accordingly we have deleted this sentence in the revised version of the manuscript

L252. Why would the fast-degrading DOC have that effect on PP? it is expected to be the one transparent fraction, so a higher amount should not hamper PP. Again, the manuscript would benefit of a more extense discussion and use of previous literature here (e.g. from L240 to 273 there are no references!).

The effect of terr-DOM on PP is twofold. 1. Terr-DOM does reduce the light available for phytoplankton increasing light-limitation. 2 terr-DOM fuels an heterotrophic food chain (bacteria, heterotrophic nanoflagellates, microzooplankton) which also exert a top down control on phytoplankton, resulting in a net reduction of PP. This latter mechanisms is the main results of the manuscript and is, to the best of our knowledge, novel. This is why we do not cite other papers when describing it. However, the implications of this finding are, we think, adequately discussed later in the text.

L339. However, it is also important to note that flocculation and photodegradation will also be dependent on the type of C...That could have a major impact on your results, as those processes are independent of the foodweb. Discuss.

There are evidences that flocculation is a processes which is only relevant within estuaries i.e. at low salinity (up to 6 psu, e.g. Forsgren et al. 1996 and the others papers cited in our introduction). As

such, it is likely to have marginal effects in the site where the model has been implemented (see also Anderson et al. 2019). While photodegradation is more relevant than flocculation (e.g. Anderson et al., 2019) it is well established that terr-DOM degradation processes become progressively dominated by microbes moving from estuaries onto the shelf (e.g. Goncalves-Araujo et al. 2015 Front. Mar. Sci.). Other studies examining the relationship between CDOM and DOC (Helms et al 2008; Matsuoka et al 2012) have noted that higher CDOM absorbance at 443 nm ($a_{CDOM_{443}}$) and lower absorbance spectral slope ($S_{350–500}$) can be used as a proxy to distinguish between DOM losses from microbial degradation and photodegradation. Using this approach, Juhls et al (2019), observed higher $a_{CDOM_{443}}$ associated with lower $S_{350–500}$ across the salinity gradient observed in the Laptev Sea (2 – 24 psu), pointing towards stronger microbial degradation than photodegradation of terr-CDOM in the area where the model is implemented..

While the above cited papers clearly suggest that photodegradation is a relatively minor process at the location where our model has been implemented we also conducted a sensitivity experiment to be sure that changes in photodegradation parameters would not substantially change our results. The results shown in the figure below (Figure 2 of this document) demonstrate that small uncertainties in the photodegradation parameters (iref and phi, see equation 5) will not greatly impact the photolysis rate (changes mostly <<20%), with the greatest sensitivity occurring only with an approximate order of magnitude increase in phi and very low iref values. This exercise make us confident that even considering that the photodegradation parameters would vary concomitantly with the biological lability, this would not substantially impact the quality of our results.

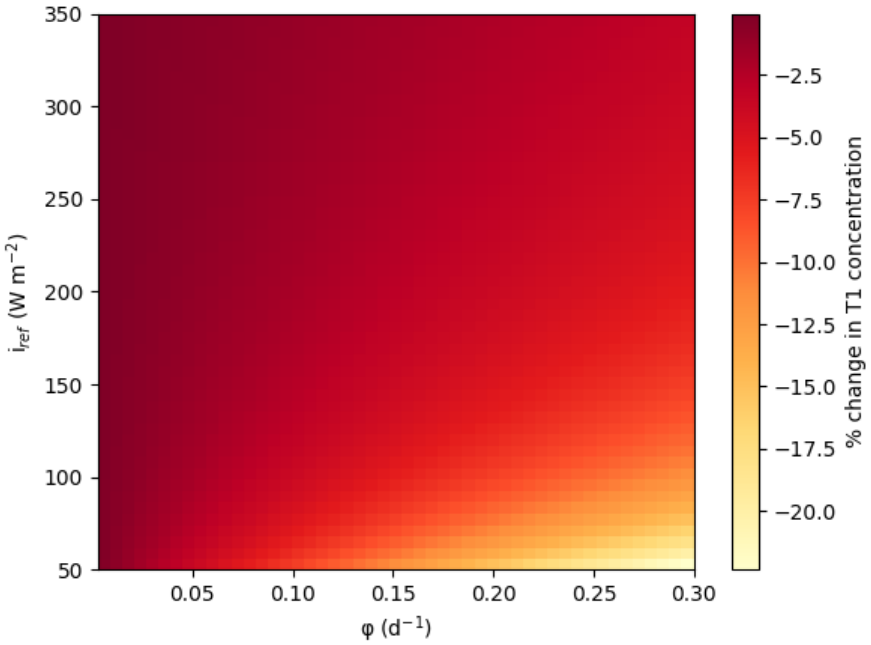

Figure 2: Sensitivity plot showing how photolysis parameters, phi and iref (Eq 5) impact T1 concentration after 1 day. Note the default parameters for phi and iref are 0.03 d-1 and 130 respectively.

FIGURES

Figure 1.

Why there are only 16 polygons?? shouldn't it be 25 conditions in that surface? why representing it with a more "coarse" resolution? it hampers the interpretation.

The 25 simulations are represented by the corners of each polygons and their values are reported on the figure grid. Colors refer to the interpolation between them.

In the caption, CO2 flux means net outgassing? keep the wording in the text and captions consistent. Also, provide the name of the variable in the legend.

We have specified that the variable is air-to-sea CO2 flux. So positive values mean ocean uptake, negative, outgassing. This is the convention used across most marine biogeochemical models. This has been clarified in the text.

The fact that positive values indicate CO2 sink is extremely confusing. The majority of studies on CO2 exchange water-air provide negative values as influx and positive values as efflux. First, clarify in the caption that you refer to the flux to the atmosphere and if so, efflux (net source) should be positive values and sink negative values.

See previous answers

Fig 3. (and results&discussion associated)

No need for different colors, avoid.

We have followed the reviewer's suggestion (See new Fig 4)

Also, what is the uncertainty of the bars? Add.

Uncertainties have been added, see new Fig 4

What about the photodegradation active radiation at 5-meter depth? How would a better accounting of photodegradation impact the results? Include those aspects in the discussion. What is the dependency of the results on the parameters linked to e.g. photodegradation? That would make clear the relevance of the bacterial degradation process as the novelty of the model.

The point of the manuscript is NOT the "relevance of bacteria degradation" which, in our opinion, is already well established (see lines 59 62 and the papers cited there), but how the system reacts to increasing terr-DOM lability. Photodegradation parameters are not altered in our experiments in order to highlight the effect of changing biological lability on the simulated patterns.

See also our previous answers re. photodegradaton and figure 2 in this document

Fig 4.

Provide x axis in correct order (from low to high values).

Also, avoid colors, they do not provide any information.

Additionally, is there no uncertainty linked to those averages? it should be added.

We have revised Fig 4 (Fig 5 in the revised version) following the suggestions of the reviewer

Fig S1.

I would have appreciated having this figure in the main text. Also, the same wording should be used. photo-lysis is not used, POC is not important, T1 and T2 are in subscript.

We have revised this figure and included it in the main text (new Fig 1). We agree that POC is not very important in this context but we have left it in the figures for consistency with equation 4 (Total substrate available for bacteria)

References (not already cited in the main documents)

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Supplementary Materials