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Global changes alter the amount and composition of land carbon deliveries to European rivers and seas

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Lateral carbon transfer along the land-ocean continuum is a key component of global carbon cycle, yet its response to global change is poorly quantified. Here, we use a land-surface model to simulate vertical (soil-plant-atmosphere) and lateral (land-river-ocean) carbon exchanges in Europe between 1901–2014 and investigate the effect of atmospheric carbon dioxide, climate and land use changes on lateral carbon transfer. We find that global change during 1901–2014 led to a significant increase in the total terrestrial carbon delivery to European rivers (33% increase) and to the sea (20% increase). Carbon delivery increased in the dissolved phase and decreased in the particulate phase. Climate change, increased atmospheric carbon dioxide, and land-use change explain 62%, 36% and 2% of the temporal change in European lateral carbon transfer during the study period, respectively. Our findings suggest that redistribution of soil carbon due to lateral carbon transfer induced a 5% reduction in the net land carbon sink in Europe.

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Lateral carbon (C) transfer (LCT) along the land-to-ocean continuum has been a topic of broad interest as it is an important component of the global C cycle[1–6] and can strongly affect the function of aquatic ecosystems including the coastal zone (e.g., oxygen concentrations, nutrients availability, and turbidity)[7,8]. Global soil erosion and leaching release large amounts of C from land to river and the sea every year[5,6]. The subsequent deposition and burial of particulate C in river channels and floodplains contribute to the global C sequestration[9–11]. In contrast, the re-emission of carbon dioxide (CO_2), which is leached from upland soils to rivers or produced in-transit by the mineralization of riverine organic C usually constitutes a closed C loop with the atmosphere[6,12–14]. The fraction that is not buried or recycled back to the atmosphere is ultimately exported to the ocean, partly contributing to the recalcitrant organic C pool, or to trophic status of marine waters and the air-sea CO_2 exchange[8,15–20]. Moreover, previous studies have revealed that the magnitude of global lateral C flux (2.35 ± 0.70 PgC yr^{-1}) is comparable to the net C sink of global terrestrial ecosystems (2.30 ± 1.5 PgC yr^{-1})[6], and ignoring LCT in land surface models results in notable biases (~10%) in the simulated terrestrial C budget[21–23]. Accurately estimating LCTs along the land-to-ocean continuum thus is vital for better constraining the C budget of terrestrial and aquatic ecosystems, as well as their response to fossil fuel emissions, land use and climate changes.

Nonetheless, there are still notable uncertainties in existing estimates of large-scale LCTs, in particular regarding their temporal trends under global change[4–6]. Previous estimates of global C loss from soil to rivers vary from 1.6 to 4.1 Pg ($=10^{15}$ g) C per year (yr^{-1})[5,6] while estimates of global riverine C export to the sea are better constrained and mostly fall in the range 0.7–1.2 Pg C yr^{-1}[5,6,24] (Supplementary Table S1). Estimates of CO_2 emission from global inland waters also vary widely from 0.75 to 2.5 Pg C yr^{-1}[5,6,25]. These assessments are mostly based on inventory and extrapolation of observed riverine organic C and CO_2 concentrations, river discharges, and surface area of inland waters[1,3,8,26]. However, existing observational data are still too sparse to provide a global and unbiased spatial coverage while long-term trends (e.g., before 1980s) are essentially unknown, especially for the particulate organic C (POC)[4]. Due to the scarcity of observational data, only a few studies have investigated the temporal evolution of LCTs over the past decades at large spatial scales[1,6,27].

Process-based land-surface models (LSMs) with explicit representation of LCTs, in conjunction with sparse observational data for model calibration and evaluation, have proven to be a suitable approach to estimate the long-term land-to-ocean C fluxes at large spatial scales[22,28–30]. As LSMs simulate both vertical (atmosphere-plant-soil) and lateral (land-river-ocean) carbon cycles, they can also be used to explore the complex interactions between vertical and lateral C fluxes, from vegetation C uptake to C exports to the ocean. In addition, global changes, most importantly atmospheric CO_2 increase, climate change and land use change, have together resulted in notable alterations of LCTs by impacting not only surface runoff and belowground drainage[31], but also vegetation growth, soil C stocks and the decomposition rate of riverine organic C[4,32–34]. As the few observed changes in LCTs result from the combined effects of different global/regional change factors, it is difficult to attribute the overall temporal evolution to each of these factors. By being able to conduct factorial simulations, LSMs are ideal tools for attribution analyses, allowing to distinguish the effect of each global change factor on past LCTs and predict their future evolution under different climate change or land use change scenarios.

At the European scale, the magnitude of the lateral C flux from land to river (113 Tg C yr^{-1}) has been estimated to be comparable to the carbon accumulation in European forests[1], and the C concentrations in the waters of many European rivers have been observed to have changed drastically over the past decades[33,35–37]. Even so, an integrated view of the magnitude and composition of LCTs through the European river network and how they evolved over the past century as a result of changes in climate, land-use, and atmospheric CO_2 concentration, is still lacking. In the absence of such temporal trends, the river C fluxes cannot be decomposed into natural and anthropogenic perturbation terms, precluding their inclusion in anthropogenic CO_2 budget analyses, as performed by the Global Carbon Project[38] and the IPCC[39]. To fill these gaps, we apply the newly developed LSM ORCHIDEE-Clateral[40] to: (1) estimate the spatiotemporal variations of lateral C (POC, DOC, and CO_2) transfers along the land-to-ocean continuum of the European continent during the period 1901–2014; (2) quantify the respective contributions of three different global and regional change factors (atmospheric CO_2 increase, land use change and climate change (including changes in climatic factors such as temperature, precipitation, radiation, and wind speed)) to the temporal evolution of C fluxes from land to river and the sea over the study period; and (3) estimate the impact of LCTs on the European terrestrial C budget. ORCHIDEE-Clateral is a new branch of the land-surface scheme of the Institute Pierre Simon Laplace (IPSL) Earth system model[40]. In contrast to conventional LSMs, it simulates not only the vertical fluxes of energy, water and C in terrestrial ecosystems, but also represents the lateral transfers of sediment, POC, DOC, and CO_2 from terrestrial ecosystems to rivers and the sea.

Results and discussion

Present-day (2005–2014) lateral C transfers in Europe. Based on the simulation results of LCTs in Europe during the period 2005–2014, the present-day annual total C delivery (including POC, DOC, and dissolved CO_2) from upland soils to rivers is 46.6 ± 3.7 (mean ± standard deviation of annual values during 2005–2014) Tg C yr^{-1}, of which 17.0 ± 0.5 Tg C yr^{-1} is ultimately exported to the sea (Fig. 1). The largest fraction ($53 \pm 2\%$) of C delivered to the river network is in the form of dissolved CO_2 from the decomposition of soil organic matter. DOC and POC account for $35 \pm 1\%$ and $12 \pm 1\%$ of the soil-to-river C flux, respectively (Fig. 1). Moreover, although leaching from upland soil solution is the main source of DOC and CO_2 inputs to European rivers, swamps and floodplains still contribute 16% and 41% of the total DOC and CO_2 inputs, respectively. Annual total C burial and infiltration in sediments of European rivers amounts to 5.8 ± 0.9 Tg C yr^{-1} while CO_2 emissions are much larger and reach 23.8 ± 2.7 Tg C yr^{-1}. Terrestrial CO_2 inputs fuel a large part (~85%) of these emissions, with only a small part (~15%) originating from the in-situ decomposition of riverine organic C (Fig. 1). Because of internal transformation processes within the river network, the composition of the total C exports to the sea are different from the one of terrestrial C inputs, with the DOC fraction in total C exports to the sea being $54 \pm 1\%$, followed by CO_2 ($25 \pm 1\%$) and POC ($21 \pm 2\%$).

Both the magnitude and composition of C delivered from upland soils to rivers and the seas vary drastically in space across Europe (Supplementary Figs. S2 and S3). The total C loss rates from soils to rivers range from <0.1 g C m^{-2} yr^{-1} to more than 10 g C m^{-2} yr^{-1} (Supplementary Fig. S2). Similar to previous studies[32,41–43], the simulated POC loss rate broadly increases from northern to southern Europe while the DOC loss rate in northern Europe and the Alps is overall higher than in the other regions. Different from POC and DOC, the highest loss rates of

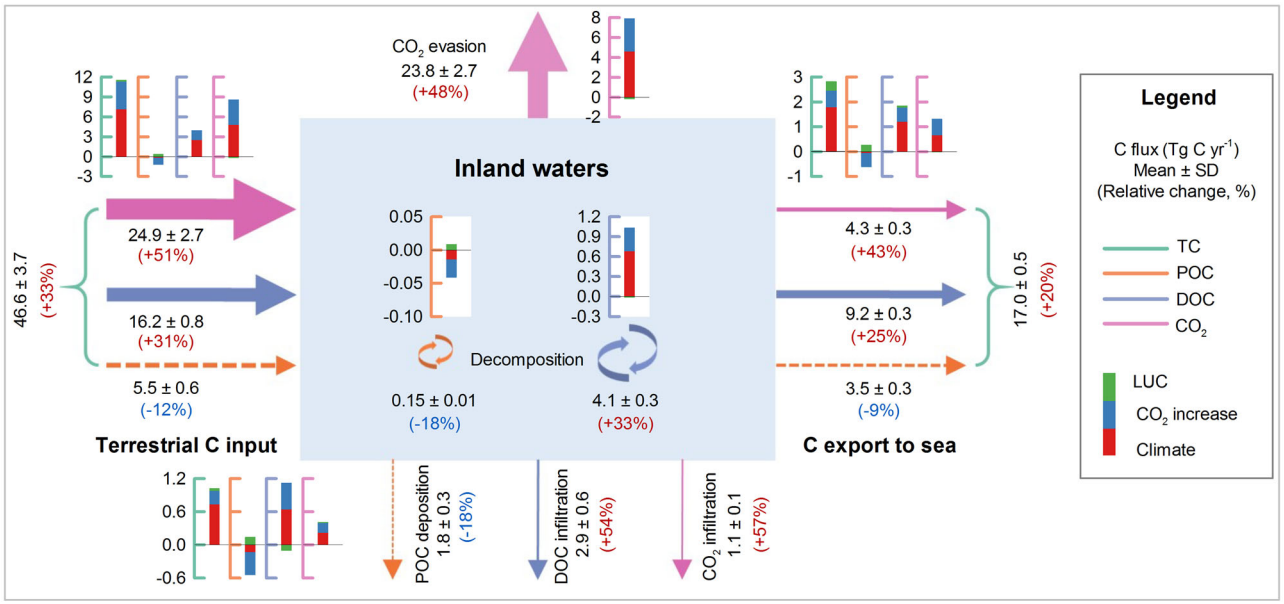


Fig. 1 Present-day (2005–2014) lateral carbon fluxes in Europe and their relative changes from the reference period 1901–1910 to 2005–2014. POC particulate organic carbon (C), DOC dissolved organic C, CO_2 dissolved C dioxide, TC total C flux which is the sum of POC, DOC and CO_2. Percentages in the brackets are the relative changes in C fluxes from the reference period 1901–1910 to present-day (2005–2014), with dashed arrows and percentages in blue representing the decrease and solid arrows and percentages in red representing the increase. Bar charts show the respective contributions of climate change, atmospheric CO_2 increase and land use change (LUC) to the changes in lateral C fluxes from 1901–1910 to 2005–2014.

dissolved CO_2 are mostly found in central and western Europe. Further analysis indicates that the total C delivered to rivers is mainly composed of DOC (53%) in northern Europe (EuN), with POC only accounting for 4% (Supplementary Fig. S3). In middle-western (EuMW) and middle-eastern (EuME) Europe, CO_2 accounts for the largest fraction of total C delivered to river, followed by DOC and POC. In stark contrast from our findings in central and northern Europe where the POC delivery is drastically smaller than the DOC delivery, the fraction of total C delivered in the form of POC (26%) is higher than that of DOC (24%) in Southern Europe (Supplementary Fig. S3).

Investigating the difference in soil C losses under different land uses is helpful to understand the impact of land use change on LCTs. We find that different land use types across Europe exert a strong control on the total C deliveries to rivers (Supplementary Fig. S4). In northern Europe (EuN), forests contribute 67% of the total C delivery to rivers, followed by the contributions of grasslands (30%) and croplands (3%). The overall pattern is different in middle and southern Europe where croplands contribute 30–40% of the total C delivery, a fraction that is comparable to forests. For the whole of Europe, forests contribute the largest fraction (42%) of C delivery to river, followed by grasslands (31%) and croplands (27%). The contribution of forests, grasslands and croplands to the total C loss to European rivers not only depends on the land cover fraction of each vegetation type, but also on the area-averaged C loss rate per vegetation type. Consistent to widespread observations[34,44], our simulation results suggest that soil POC loss rates are generally lower in forests than in croplands and grasslands (Supplementary Fig. S4), as the accumulated aboveground litter and the well-developed tree canopy and root structure can protect soils from erosion. DOC loss rates, however, are generally highest in forests, compared to croplands or grasslands. For the whole of Europe, the average POC loss rate under forest is about 58% and 28% of that on grassland and cropland, respectively. However, DOC loss rate in forest is about 1.7 and 1.9 times that on grassland and cropland, respectively. POC only accounts for 5% of the total C losses from forests to rivers, but accounts for 22% of the total C losses from croplands to rivers. At face value, DOC accounts for 43% of the total C losses from forests and 26% of the total C losses from croplands (Supplementary Fig. S4). In grasslands, POC and DOC accounts for 13% and 35% of the total C loss to rivers, respectively. CO_2 is the largest component of total C loss to rivers regardless of the land use type. In forests, grasslands and croplands, CO_2 all accounts for ~52% of the total C loss to rivers.

Century-scale changes in lateral C transfers (1901–2014). Global change, including climate change, atmospheric CO_2 increase and land use change in this study (Supplementary Figs. S5–S7), has led to significant ($p < 0.05$) changes in the magnitude and composition of C deliveries from land to river and the sea in Europe (Fig. 1 and Supplementary Fig. S3). Our simulation results reveal that the total C delivery from upland soils to European rivers and seas has increased by 33% and 20%, respectively, from the reference period 1901–1910 to present-day (period 2005–2014, Fig. 1). These increases are mainly attributed to enhanced DOC and CO_2 deliveries. In fact, the POC delivery actually shows an overall decreasing trend from 1901 to 2014. As a result, the POC fraction in the total C delivery to rivers decreased from 18 ± 2% in 1901–1910 to 12 ± 1% in 2005–2014, while the CO_2 fraction concomitantly increased from 44 ± 4% in 1901–1910 to 52 ± 2% in 2005–2014 (Fig. 1). The change in the relative contribution of DOC is insignificant ($p > 0.05$) over the simulation period. A similar pattern is also observed for the total C exported to the sea, with the fraction of POC decreasing from 27 ± 2% in 1901–1910 to 21 ± 2% in 2005–2014 and the fractions of DOC and CO_2 increasing from 52 ± 1% to 54 ± 1% and from 21 ± 1% to 25 ± 1%, respectively (Fig. 1). Moreover, as riverine DOC and CO_2 generally have a much shorter turnover time than POC, the stability of riverine C has declined from 1901 to 2014, due to its compositional change. As a result, the ratio of total C exported to the sea to the total C delivered from land to river has decreased by 10% from 1901–1910 to 2005–2014.

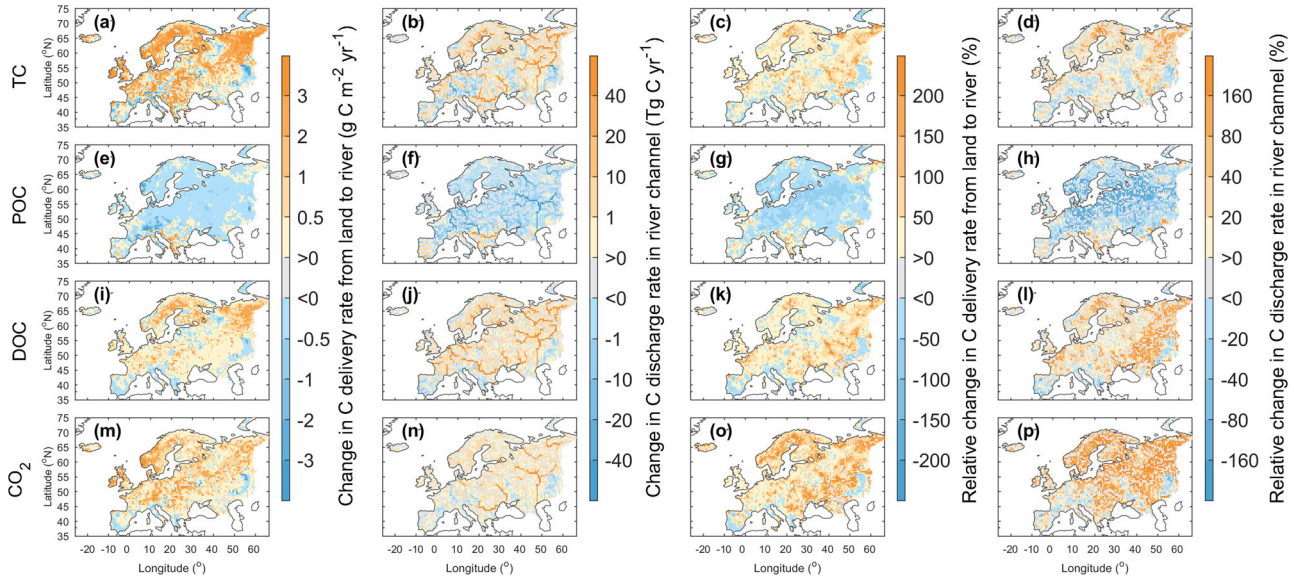


Fig. 2 Spatial distribution of the changes in the lateral carbon deliveries in Europe from the reference period 1901–1910 to 2005–2014. POC particulate organic C, DOC dissolved organic C, CO_2 dissolved C dioxide, TC total C flux, which is the sum of POC, DOC, and CO_2. **a**, **e**, **i**, **m** The changes in delivery rates of TC, POC, DOC, and CO_2 from land to river, respectively. **b**, **f**, **j**, **n** The changes in riverine discharge rates of TC, POC, DOC, and CO_2, respectively. **c**, **g**, **k**, **o** The relative changes in delivery rates of TC, POC, DOC, and CO_2 from land to river, respectively. **d**, **h**, **l**, **p** The relative changes in riverine discharge rates of TC, POC, DOC, and CO_2, respectively. Grid cell with no significant change ($p > 0.05$) is colored in gray.

Due to the combined effects of enhanced C delivery to rivers and rising temperature, CO_2 evasion from European inland waters (including river flows and flooding waters) has increased significantly ($p < 0.01$) from 1901 to 2014 (Fig. 1 and Supplementary Fig. S3). In 2005–2014, the average annual total CO_2 evasion in Europe was higher by 48% compared to the reference period 1901–1910. Not surprisingly, the spatial patterns of changes in CO_2 emission from inland waters follow that of changes in C delivery to rivers (Supplementary Fig. S8). In central and northern Europe, the CO_2 emissions from most rivers increased significantly ($p < 0.01$) from 1901–1910 to 2005–2014, while in southern Europe, they declined significantly ($p < 0.01$). The relative changes in many regions across Europe are larger than 50% and can even reach 150%.

The simulated trends in lateral C deliveries over Europe are overall consistent with previous studies based on observations[33,36,37,45]. Several studies have reported a substantial and widespread increase (>60%) in total organic C concentrations in European rivers during the past decades, mainly due to enhanced terrestrial DOC exports to freshwaters[35–37,46,47]. A significant ($p < 0.01$) increase in CO_2 emission has also been observed in many boreal aquatic ecosystems caused by the increased export of terrestrial dissolved C to inland waters[13,48]. Different from DOC and CO_2, several publications have reported a declining trend in sediment and POC discharges in some European rivers (e.g., the Rhine and Ebro) during the past decades[49,50]. Our model overall captures the divergent temporal trends in dissolved and particulate C lateral fluxes, although their magnitude of change might be different from observations[33,36,37,48].

Our results also highlight that due to the marked spatial variation in climate[51] and land use changes[52] (Supplementary Figs. S6 and S7), trends in LCTs in Europe may also show drastical spatial variation over the time period considered (Fig. 2). Overall, the total C delivery to rivers increased significantly ($p < 0.01$) in most regions of Europe and the spatial patterns of changes in DOC and CO_2 deliveries to rivers follow the same pattern (Fig. 2). Their relative changes are generally smaller than 50%, but in some regions with substantial increase in rainfall (Supplementary Fig. S6), the relative changes can exceed 150% (Fig. 2). The spatial pattern of changes in POC delivery is essentially opposite to that of DOC and CO_2 (Fig. 2), with a decline in most regions of Europe, except in areas with strong increase in rainfall or with substantial conversion from forests or grasslands to croplands (Fig. 2, Supplementary Figs. S6). Similar to the changing terrestrial C inputs to rivers, the POC discharges to the sea decreased in most European rivers, while DOC and CO_2 discharges increased during the period 1901–2014 (Fig. 2).

Impacts of different global change factors on lateral C transfer. Drivers of changes in lateral POC, DOC, and CO_2 transfers from the reference period 1901–1910 to the period 2005–2014 are different (Fig. 1 and Supplementary Fig. S3). The lateral POC delivery from land to river is mainly determined by the soil erosion rate. Climate change (including rising temperature and precipitation) and atmospheric CO_2 increase in Europe (Supplementary Fig. S6) induced a significant ($p < 0.01$) rise in net primary production (NPP), plant biomass and litter stocks[53,54] (Supplementary Fig. S9). These increases in plant biomass (especially leaf and root stocks) and litter have enhanced the protection of the soil and its associated C content against erosion (Supplementary Fig. S10a), although the rising (+14%) rainfall and runoff (Supplementary Fig. S9i) have partly mitigated this effect. In the model, the protection effect of vegetation and litter on soil loss from uplands to rivers is represented by the cover and management factor (C-factor, Supplementary Fig. S9g). Different from climate change and atmospheric CO_2 increase, land use change in Europe induced a significant ($p < 0.01$) increase in the total POC delivery to European rivers (Fig. 1), mainly due to the expansions of cropland in southeastern Europe[34] (Supplementary Fig. S7). In stark contrast from the behavior of POC, climate change and atmospheric CO_2 increase enhanced the DOC and CO_2 deliveries to river (Fig. 1). The increasing rainfall in Europe from 1901–1910 to 2005–2014 led to an increase of about 14% in the total water discharge from headwater basins to rivers (Supplementary Fig. S9j). In parallel, the atmospheric CO_2 increase

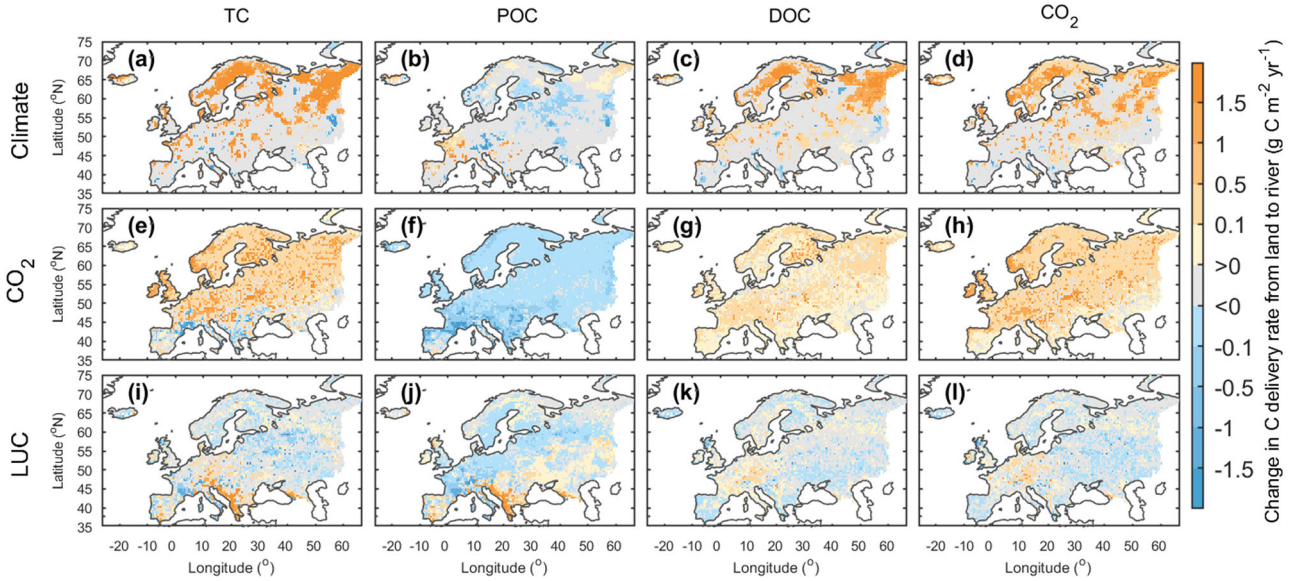


Fig. 3 The respective impacts of climate change, atmospheric CO_2 increase, and land use change on lateral carbon transfers in Europe. Climate (**a**–**d**), CO_2 (**e**–**h**) and LUC (**i**–**l**) denote the changes in lateral carbon transfers due to climate change, atmospheric CO_2 increase and land use change (LUC) from the reference period 1901–1910 to 2005–2014, respectively. POC particulate organic C, DOC dissolved organic C, CO_2 dissolved C dioxide, TC total C flux, which is the sum of POC, DOC and CO_2. Positive value means an increase in the lateral C delivery rate, and vice versa. Grid cell with no significant change ($p > 0.05$) is colored in gray.

induced a slight but significant ($p < 0.01$) increase in soil C stocks by increasing plant primary production and litterfall (Supplementary Fig. S9). Both of these effects have stimulated the leaching of soil DOC and dissolved CO_2. Compared to climate change and atmospheric CO_2 increase, land use change only had a slight influence on the total DOC or CO_2 delivery to the European river network (Fig. 1). Moreover, we find that the impacts of the different global change factors on C exported to the sea are overall similar to their impacts on the C delivery from upland soils to rivers (Fig. 1).

In quantitative terms, integrated over the whole of Europe, climate change (including changes in temperature, precipitation, radiation, and wind speed), atmospheric CO_2 increase, and land use change explain 62%, 36% and 2% of the change in total C delivery to rivers, respectively (Fig. 1). The drivers of changes in DOC and CO_2 are broadly consistent, with relative contributions amounting to 64%, 35% and 1% for DOC and 55%, 43% and 2% for CO_2. However, the attribution reveals that the change in POC delivery is driven by distinct factors, climate change only contributing to 13%, while the relative shares of atmospheric CO_2 increase (61%) and land-use change (26%) are significantly ($p < 0.01$) larger. As the river CO_2 outgassing mainly originates from terrestrial CO_2 inputs and the in-situ decomposition of riverine DOC, changes in riverine CO_2 emissions are also mainly caused by climate change and atmospheric CO_2 increase (Fig. 1 and Supplementary Fig. S3).

The contribution of each factor in explaining trends in LCTs nevertheless shows substantial spatial variation (Fig. 3, Supplementary Figs. S3 and S11). The decreasing trend in POC and increasing trends in both DOC and CO_2 deliveries to rivers induced by atmospheric CO_2 increase are overall consistent across Europe (Fig. 3 and Supplementary Fig. S11). In most regions of Europe, atmospheric CO_2 increase explains 30–60% of the overall change in the lateral transfer of total C (Supplementary Fig. S12). Yet the trends resulting from climate and land use change vary drastically across Europe. Overall, climate change dominates the changes in LCTs in Northern Europe and in some scatteredly distributed areas in middle Europe (Supplementary Fig. S12). Land use change mainly induces an increasing trend in LCT in Southern Europe (most notably in the Apennines and the Balkans), due to the conversion from forests to croplands and grasslands (Supplementary Figs. S7 and S11). In many areas of the Great European Plain, reforestation induces a strong (>40%) decline in the POC delivery from land to the river, however, a slight increase in the deliveries of DOC and dissolved CO_2. In addition, land use change mainly affects the lateral transfer of POC, and its effects on DOC and CO_2 transfers is limited (mostly <10%, (Supplementary Fig. S11).

Our knowledge on the drivers of the long-term change in LCT over Europe is still very limited, due to the scarcity of large-scale and long-term observation data. Several hypotheses have been proposed to explain the observed increases in riverine DOC concentrations[55]. For example, based on local observations, Evans et al.[35] and Monteith et al.[36] have proposed that the decrease in the acidity of the soil solution and stream waters resulting from changes in atmospheric deposition chemistry was the main driver of the increase of riverine DOC concentration in Europe. Using regression approaches, Hruska et al.[37] concluded that the changes in ionic strength of soil and stream waters are the primary drivers of rising DOC concentration in central European streams, rather than climate change. Our simulation results do not account for these complex effects but nonetheless support the hypothesis[56] that climate change (warming and increasing precipitation) and atmospheric CO_2 increase can already explain a certain part of the rising DOC concentrations in European rivers (Fig. 1, Supplementary Figs. S3 and S13), as these global change drivers affect vegetation production, soil organic matter decomposition and, ultimately, the DOC concentration in soil solution and the leaching rate of soil DOC (Supplementary Fig. S9). We also note that the simulated total DOC discharge from European rivers to the sea in the global study by Li et al.[57] declined by ~30% between 1951 and 2015. This declining trend is opposite to our simulation results. Li et al.[57] proposed that global warming might have resulted in a decline in DOC discharge by increasing the decomposition rate of the riverine DOC and extending the residence time of waters in the European river network. However,

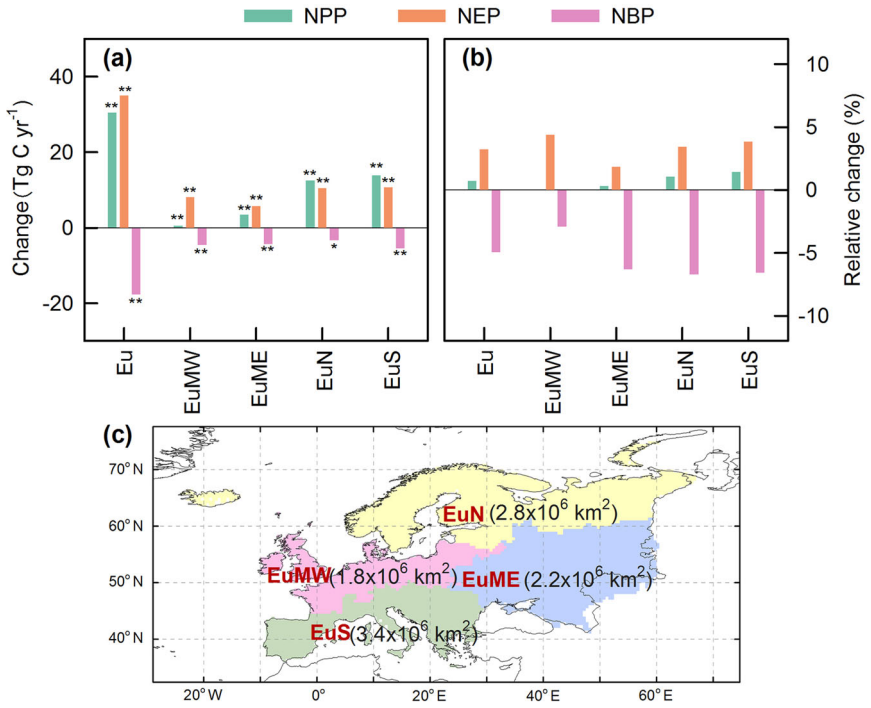


Fig. 4 Impacts of lateral carbon transfer on the terrestrial carbon budget in Europe during the period 2005-2014. NPP, NEP and NBP are net primary production, net ecosystem production and net biome production, respectively. **a** Changes in NPP, NEP and NBP caused by lateral carbon transfer. **b** Relative changes in NPP, NEP and NBP caused by lateral carbon transfer. **c** Locations and areas of the four subregions (i.e., EuN Northern Europe, EuMW Middle-Western Europe, EuME Middle-Eastern Europe, and EuS Southern Europe) of Europe (Eu). ** and * denotes the simulated NPP, NEP or NBP with representation of lateral carbon transfer is very significantly ($p < 0.01$, based on paired-sample t-test) and significantly ($p < 0.05$) different from the simulation result without representation of lateral carbon transfer, respectively.

the temperature increase in Europe from 1901 to 2014 is only 1.6 °C on average (<2 °C in most areas of Europe). According to the empirical temperature-dependent DOC decomposition function with parameters calibrated against observations[29,57], this 1.6 °C warming would only induce a 4% increase in the decomposition rate of riverine DOC[57], which is much smaller than the observed increase (>25%) in total DOC delivery from soils to rivers[36,49,50,58] (Fig. 2).

Implications of lateral C transfer for the terrestrial carbon balance. LCT alters the terrestrial C balance and induces a substantial spatial redistribution of terrestrial C stocks and fluxes in Europe. We estimate that the total annual land-to-river C delivery ($POC + DOC + CO_2$) over Europe during the period 2005-2014 is about 1.1 ± 0.1% of the vegetation primary net production (NPP), 4 ± 1% of the net ecosystem production (NEP, calculated as the difference between Net Primary Production and soil respiration) and 18 ± 10% of the net biome production (NBP, defined as the net C accumulation in terrestrial ecosystems) (Fig. 4). The organic C delivered to rivers can be transported over long distances before finally settling or infiltrating in floodplains or river channels. In these sedimentary settings, the local environmental conditions are typically quite different from those encountered in upland soils from where these C pools originate. These changes in environmental conditions can affect the decomposition rate of the laterally redistributed organic carbon[59,60].

Besides redistributing spatially the terrestrial C, the lateral transport of water, sediment and C also affects the land C budget through several indirect ways. For example, soil erosion and sediment deposition affects the land C budget by altering the vertical distribution of litter and soil organic C, as the two control factors of SOC decomposition rates, soil temperatures and fresh organic matter inputs (through the priming effect[61,62]), vary across soil layers. In addition, the lateral movement of surface runoff affects the terrestrial C budget by modifying soil wetness. Comparison of simulations with (scenarios-1) and without (scenarios-4) representation of LCT (Supplementary Table S2) reveals that this effect can be significant ($p < 0.05$), especially in floodplains (Supplementary Fig. S14g), where the increase in soil wetness can be larger than 10%. Benefiting from the reduced constraints of soil water on plant growth, the NPP in many grid cells with a large floodplain area increases by more than 5% in the simulation with LCT (Supplementary Fig. S14a).

Ignoring LCTs in land-surface models can potentially introduce considerable biases in simulated vegetation and soil C stocks and, thus, in the terrestrial C sink (Fig. 4 and Supplementary Fig. S14). Comparison of simulations with and without representation of LCT suggests that the LCT from 1901 to 2014 induces a slight (within ± 1%) but significant ($p < 0.01$) change in vegetation biomass, litter and SOC stocks (Supplementary Fig. S9 and S14) in Europe. The change in soil DOC is however much larger than that of SOC, with a decrease of 18% when the lateral C fluxes are accounted for (Fig. S9f). For the last decade (2005-2014) of our simulation, we find that the LCT has induced an increase of 1% in NPP (Fig. S9a), yet a decrease of 5% in NBP (Fig. S9b). This finding implies that although the atmospheric C uptake by the terrestrial vegetation increases due to LCT, the annual net C accumulation in European terrestrial ecosystems (NBP) actually decreases. Moreover, the impacts of LCTs on regional C stocks and fluxes vary drastically across Europe (Supplementary Fig. S14). Higher lateral C delivery rates from land to the river network generally result in larger decreases in the NBP of terrestrial ecosystems (Eq. 8). In many areas of Europe with high erosion (e.g., the Alps and Balkans) or leaching (e.g.,

British Island and the north-western part of Scandinavia) rates of soil carbon, the decrease in NBP can exceed 20% (Supplementary Figs. S2 and S14b).

The estimate of Ciais et al.[1] based on a budgeting approach suggested that lateral C fluxes through European rivers could reach 30–60% of the annual C uptake of European terrestrial ecosystems, which is higher than our estimate relying on a process-based modeling approach (18 ± 10%). Using a similar modeling approach, Lauerwald et al.[21] and Hastie et al.[63] estimated that the lateral DOC and CO_2 deliveries in the Amazon and Congo river basins are about 21% and 14% of the terrestrial NBP, respectively. Using the JULES-DOCM model, Nakhavali et al.[22] found that the terrestrial DOC leaching flux alone could reach 10% of the terrestrial NEP in temperate and boreal regions, which is significantly ($p < 0.01$) higher than our estimate for the European continent. Nonetheless, this study, together with previous studies, systematically indicates that large amounts of soil C are transported to inland waters and the sea. Although climate change and atmospheric CO_2 increase clearly dominate the historical dynamics of vegetation production and terrestrial ecosystem C budget (Supplementary Fig. S15), our findings support the integration of LCTs in the assessment of the terrestrial C sink and the anthropogenic C budget.

Limitations and outlook. Although our simulation results capture the observed declining trends in POC and sediment deliveries from land to European rivers and the seas (Fig. 1 and Supplementary Fig. S10), we recognize that our assessment might underestimate the decrease in POC and sediment discharges to the sea in some European rivers, as reservoirs, ponds, and lakes are not accounted for in our simulations. Reservoirs and lakes typically increase the sediment and POC deposition in the river network[49,50,58,64], as well as stimulate the mineralization of riverine organic C and the resulting CO_2 emissions by prolonging the water residence time in the river network[65,66]. The total C burial in European rivers and lakes has been estimated to be 1.2–2.6 Tg C yr^{-1}[65,66], but could be substantially higher if smaller impoundments were taken into account. Estimates using an empirical mass balance model suggest that global reservoirs have lowered the organic C export to seas via rivers by 7–13% during the period 1970–2000[65]. Although Europe is not anymore a hotspot of increased damming compared to several other regions (e.g., China, India and Brazil)[65], it should still have notably reduced the riverine POC discharges in Europe and increased the CO_2 and methane emissions from European inland waters during the second-half of the 20th century[26,67]. In addition, support practices for controlling soil erosion (e.g., contour farming and grass margins) are not represented in our model. Panagos et al.[58] suggests that the support practices for controlling soil erosion (e.g., contour farming and grass margins) can reduce soil erosion in Europe by 3%. Therefore, our model might have slightly overestimated the soil and POC losses to rivers in some regions of Europe.

Besides the non-representation of reservoirs and lakes, the omission of some other processes related to LCT potentially induces additional uncertainties in our simulation results, although it is currently hard to quantify them. First, the effects of lateral redistribution of organic matter and soil nutrients on vegetation productivity have not been represented in our model. Many studies have indicated that soil erosion and sediment deposition can affect vegetation productivity by modifying soil nutrient (e.g., nitrogen (N) and phosphorus (P)) availability[41,68–70]. For example, the loss of soil nutrients at the eroding sites might result in a decrease in vegetation production and the accumulation of nutrients in the deposition areas might induce an increase in vegetation production. Second, the carbon inputs from manure[43], sewage and river-borne phytoplankton[71] and weathering of bedrock (e.g., carbonate and silicate rocks)[64,65] are still omitted in the current version of ORCHIDE-Clateral. All riverine dissolved inorganic C (DIC) in current model is assumed to be in form of dissolved CO_2. Lateral transfer of methane, (bi-) carbonate and the transformation between bicarbonate and dissolved CO_2 have not been considered. In addition, wetlands have been identified as an important source of methane, DOC and CO_2 exports to inland waters[1,43,66]. Although we represented riparian wetlands in the form of temporarily inundated floodplains and groundwater-fed swamps in our model, some other important types of wetlands, in particular peatlands in Northern Europe, are still missing in our model. This might result in a notable underestimation of LCT in Europe, especially in the boreal regions.

To decrease these uncertainties, we first plan to implement the lateral transfer of nutrients, carbonate and bicarbonate, as well as the peatlands, lakes, and the regulation of lateral C transport by dams and reservoirs into the model in the next step of model development. Second, we plan to further calibrate our model against more observation data. We encourage future studies to collect more in-situ observations on the riverine sediment and C discharge rates across the world and carry more measurements on the decay rate of the riverine organic C. These observations and measurements are instrumental to improve the calibration of the parameters controlling LCT in land surface models and to evaluate the simulation results. Besides parameters controlling LCT, the model parameters used for simulating the formation and decomposition also need to be further calibrated, as an accurate estimation of soil C concentration is the precondition for well simulating LCT. Comparing our simulation result to the observation-based soil databases suggests that although the simulated total SOC stock for the whole Europe is comparable to the observation-based estimates, our current model underestimates the SOC stock in southern Europe where the soil erosion rate is highest (see Fig. S7 in Zhang et al.[38]). This might explain why the simulated POC discharges in European rivers is not overestimated, despite the omission of the reservoir and lake effect. Finally, we plan to apply and evaluate our model in more regions of contrasting climate, vegetation and topography, or even conduct a global-scale model application. As shown above, the magnitude and composition of LCT, as well as the impact of LCT on land C budget in different regions of Europe are very different (Supplementary Figs. S2, S3 and S14). We speculate that the LCT in regions with well-growing forests (e.g., in tropical and boreal forests) would be dominated by the leaching of soil DOC and CO_2, yet the erosion of soil POC would dominate the LCT in hilly regions (e.g., the Loess Plateau in China) with large-scale grasslands or croplands. By applying our model to different regions of the world, we can compare the LCTs under different climate and vegetation conditions, and test whether our model would perform satisfactorily in regions with very different climates than Europe such as the tropical region. In addition, as LCT can change regional C balance, in particular in regions with high erosion/leaching rate, a global application of our model will be helpful to better understand the role of LCT in global C cycle and to more accurately estimate the C budget of terrestrial, oceanic and atmospheric C pools.

Methods

Land surface model ORCHIDEE-Clateral. The ORCHIDEE-Clateral used in this study is a new branch of the land surface model ORCHIDEE (Organizing Carbon and Hydrology In Dynamic Ecosystems)[40]. It was developed from two previous branches of the ORCHIDEE model: (1) the ORCHILEAK[28] which simulates the leaching and fluvial transfers of DOC and CO_2; and (2) the ORCHIDEE-MUSLE

which simulates the erosion-induced sediment and POC delivery from upland soil to river network, and the erosion-induced dynamics of vertical distribution of soil organic C (SOC)[30]. By merging the ORCHILEAK and ORCHIDEE-MUSLE, and further implementing the fluvial sediment and POC transport processes in river network into the merged model, the final ORCHIDEE-Clateral can simulate large-scale lateral transport of water, sediment, POC, DOC, and CO_2 from uplands to the sea through river networks, the DOC and CO_2 inputs from submerged floodplains and riparian wetlands (e.g., swamp) to inland waters (including river flows and flooding waters, but not lakes and reservoirs), the decomposition and transformation of POC and DOC during fluvial transport, the exchanges of water, sediment and C between rivers and floodplains, the deposition of sediment and POC in river channels and floodplains, the CO_2 evasion from inland waters to the atmosphere, as well as the changes in soil wetness and vertical SOC profiles due to the lateral redistribution of water, sediment and C. Decomposition of POC and DOC in stream and flooding waters is calculated at daily time step using a linear kinetic based on the prescribed turnover times of different POC (active, slow and passive) and DOC (labile and refractory) pools and a temperature factor[40]. CO_2 evasion from inland waters is simulated using a much finer integration time step of 6 min. The CO_2 partial pressures (pCO_2) in water column is first calculated based on the temperature-dependent solubility of CO_2 and the concentration of dissolved CO_2. Then the CO_2 evasion is calculated based on the gas exchange velocity, the water–air gradient in pCO_2, and the surface water area available for gas exchange[40]. More details of the ORCHIDEE-Clateral can be found in Zhang et al.[40].

ORCHIDEE-Clateral simulates not only the cycling of energy, water and C in terrestrial ecosystems, but also the lateral water, sediment and C transfers, thus it is an ideal tool to explore the interactions between terrestrial and aquatic biogeochemical cycles, and to investigate the respective contributions of different global change factors (i.e., atmospheric CO_2 increase, and climate and land use changes) to the interannual variations of sediment and C deliveries from land to river and sea.

Model evaluation. ORCHIDEE-Clateral has been calibrated and evaluated using observation data of runoff, bankfull flow, and riverine water discharge and concentrations of sediment, POC and DOC across Europe[40]. As no observational data on the sediment delivery rates from upland soils to the river network at large spatial scale (e.g., Europe) is currently available, the simulation results have been evaluated using the sediment yield database from European Soil Data Center (ESDAC). The ESDAC sediment yields were extracted from the WaTEM/SEDEM model outputs, which rely on high-resolution data of land cover, soil erodibility, topography and rainfall[41]. Overall, the evaluation reveals that the sediment deliveries from land to major European rivers simulated by the ORCHIDEE-$C_{lateral}$ are overall comparable to those derived from the ESDAC database[41]. Comparison between simulation results and measurements at hydrological gauging stations across Europe indicates that ORCHIDEE-$C_{lateral}$ can also satisfactorily reproduce the observed riverine discharges of water and sediment in major European rivers, as well as the concentrations of organic C (including DOC, POC, and total organic C) in rivers[72]. Furthermore, the simulated long-term (1961–2020) average amounts of flooding water in major European catchments are comparable to observations at gauging stations on major European rivers[40]. This is important to ensure that our model properly simulates sediment deposition in floodplains and DOC and CO_2 exports from flooded areas to the river network (sourced from the decomposition of submerged litter and soil organic matter).

In addition to site-level evaluations, we further compared the simulated lateral C delivery rate integrated over the European domain to previous data-driven estimates. We find that the simulated present-day (2005–2014) area-averaged soil POC loss rate is 0.54 ± 0.06 (mean ± standard deviation of annual values during 2005–2014) g C m^{-2} yr^{-1}, a value which is close to the estimate (0.56 g C m^{-2} yr^{-1}) of Borrelli et al.[41] based on high-resolution data of land cover, soil erodibility and topography, and of similar magnitude to the earlier estimate (0.49 g C m^{-2} yr^{-1}) by Ciais et al.[1] based on inventory data of major European rivers. Yet, our estimate of the area-averaged POC export rate from European rivers to the seas is lower than the estimates of Mayorga et al.[73] and Ludwig et al.[74]. Based on the same empirical relationship between riverine POC and sediment concentrations obtained from 19 large rivers worldwide, the area-averaged POC export rate to the seas estimated by Mayorga et al.[73] and Ludwig et al.[74] amount to 0.67 g C m^{-2} yr^{-1} and 1.10 g C m^{-2} yr^{-1}, respectively. These exports to the sea are in fact larger than our estimate of the POC loss rate from land to European rivers. The simulated area-averaged soil DOC loss rate in our simulation is 1.59 ± 0.08 g C m^{-2} yr^{-1}, which is close the estimate of Ludwig et al.[74] (1.54 g C m^{-2} yr^{-1}) using an empirical model, the estimate of Ciais et al.[1] (1.73 g C m^{-2} yr^{-1}) based on inventory data, and the estimate of Li et al.[57] (1.72 g C m^{-2} yr^{-1}) using a global-scale process-based model; but notably higher than the estimate of Mayorga et al.[73] (0.91 g C m^{-2} yr^{-1}). Overall, our estimates of both lateral POC and DOC delivery rates are within the range of previous estimates using empirical models or data-driven methods. In addition, the simulated area-averaged CO_2 emission from European rivers in this study (2.33 ± 0.27 g C m^{-2} yr^{-1}) is close to the latest data-driven estimate (1.98 g C m^{-2} yr^{-1}) by Lauerwald et al.[25].

Vegetation production directly affects plant input to soil and soil organic C dynamics[75,76], and is thus an important control factor of lateral C transfers (LCTs). Therefore, we extend our evaluation to key terrestrial ecosystem processes. Model results indicate that the simulated present-day (2005–2014) average vegetation gross primary production (GPP, 839 ± 24 g C m^{-2} yr^{-1}) and net primary production (NPP, 411 ± 15 g C m^{-2} yr^{-1}) over the whole of Europe and four broad subregions, are comparable to estimates based on remote sensing data, which give a range of estimates, illustrative of uncertainties (Supplementary Fig. S1). Moreover, our model basically captures the increasing trend of GPP and NPP observed over the past three decades (Supplementary Fig. S1). The simulated average annual net C uptake by European terrestrial ecosystems (i.e., the NBP in this study, see Eq. 8 below) for the period 2001–2010 (30 g C m^{-2} yr^{-1}) is also within the range of previous estimates based on inventory data of terrestrial ecosystems (23–41 g C m^{-2} yr^{-1})[77–81]. In addition, the simulated soil organic carbon stock in the top 0–30 cm soil horizon integrated over the whole of Europe is 107 Pg C, which is close to the value (106 Pg C) extracted from the observation-based Harmonized World Soil Database (HWSD)[82].

Simulations. In this study, the ORCHIDEE-Clateral with parameters calibrated by Zhang et al.[40] using observation data across Europe is applied to simulate the lateral transfers of water, sediment and C in Europe over the period 1901–2014. Before launching historical simulation, ORCHIDEE-Clateral is ran over a 'spin-up' period of 10,000 years until the plant biomass and SOC pools reached a steady state. During the 'spin-up' period, the meteorological data, land use maps and atmospheric CO_2 concentrations from 1901 to 1910 are looped over to drive the model. Forcing data of climate, land cover, soil properties, routing scheme and the topographic properties of European headwater basins used in this study are same to that used in Zhang et al.[40] (see Table S3 for details).

The state of climate, land use, and atmospheric CO_2 concentration during 1901–1910 is regarded as a reference in this study. To investigate the respective contributions of changes in climate, land use, and atmospheric CO_2 concentration to the variation of LCTs in Europe over 1911–2014, and explore the impacts of LCT to terrestrial C balance, we ran ORCHIDEE-Clateral for four different scenarios (Table S2). In scenario-1, the actual forcing data of climate, land use and atmospheric CO_2 concentration for each year from 1911 to 2014 are used to drive the model. In scenario-2, the model is driven by the actual climate and atmospheric CO_2 concentration, yet the land use map is fixed to the average land use condition during 1901–1910. In scenario-3, only climatic forcing data changed year by year, both land use and atmospheric CO_2 concentration are set to the average state during 1901–1910. The forcing data of ORCHIDEE-Clateral in scenario-4 are same to that in scenario-1. Yet the simulation of processes related to lateral water, sediment and C transfers was deactivated.

Statistical analysis. Using the simulation results for scenario-1, we analyzed the spatial and temporal (interannual and seasonal) variations of the total lateral sediment and C delivery from land to river and sea, the fractions of POC, DOC, and CO_2 in total C delivery, the CO_2 evasion from inland waters, and the deposition of sediment and POC in river channel and floodplains. By comparing the simulation results during 2005–2014 (i.e., the last 10 years of the study period) to that during 1901–2010, we quantified the changes in lateral sediment and C transfers caused by global and regional changes (i.e., atmospheric CO_2 increase, and climate and land use changes) during the past one century. The significance of the difference in each variable between periods 2005–2014 and 1901–2010 was determined by independent sample *t*-test.

The impact of each global change factor on LCT is quantified by comparing the simulation results between different scenarios. In specific, the impact of climate change on LCT was quantified by comparing the simulation results in scenario-3 for period 2005–2014 to simulation results during the reference period (Eqs. 1 and 4). The impacts of atmospheric CO_2 increase and land use change on LCT were quantified by comparing scenario-2 to scenario-3 (Eqs. 2 and 5), and scenario-1 to scenario-2 (Eqs. 3 and 6), respectively. The specific equations for quantifying the absolute and relative impacts of each global change factor on LCT can be expressed as:

$$d_{climate} = V_{s3} - V_{ref} \quad (1)$$

$$d_{co2} = V_{s2} - V_{s3} \quad (2)$$

$$d_{LUC} = V_{s1} - V_{s2} \quad (3)$$

$$r_{climate} = \frac{|d_{climate}|}{|d_{climate}| + |d_{co2}| + |d_{LUC}|} \times 100\% \quad (4)$$

$$r_{co2} = \frac{|d_{co2}|}{|d_{climate}| + |d_{co2}| + |d_{LUC}|} \times 100\% \quad (5)$$

$$r_{LUC} = \frac{|d_{LUC}|}{|d_{climate}| + |d_{co2}| + |d_{LUC}|} \times 100\% \quad (6)$$

where V_1, V_2, and V_3 are the simulation results of variable V during 2005–2014 in scenario-1, 2, and 3, respectively. V_{ref} is the simulation result of V during the reference period 1901–1910. $d_{climate}$, d_{co2} and d_{LUC} are the changes in variable V

caused by climate change, atmospheric CO_2 increase and land use change, respectively. $r_{climate}$, r_{co2} and r_{LUC} are the relative contribution of climate change, atmospheric CO_2 increase and land use change to the change in variable V, respectively.

By comparing the simulation results of scenario-1 and scenario-4, we further investigated the influence of LCT on the C budget of terrestrial ecosystems in Europe. More precisely, we investigated the changes in terrestrial C storage (e.g., SOC, DOC, litter stock and plant biomass), vegetation net primary production (NPP), net ecosystem production (NEP) and the net biome production (NBP) caused by LCT. The NEP in this study is defined as the balance between the C input from atmosphere to the terrestrial ecosystems over Europe (i.e., sum of vegetation gross primary production (GPP) and wet deposition of DOC (WD_{DOC})) and the emissions of CO_2 from aboveground vegetation (R_{ab}) and soil (R_s). In each grid cell, it is calculated as:

$$NEP = GPP + WD_{DOC} - R_{ab} - R_s \quad (7)$$

The NBP is defined as the net change in C storage of the terrestrial ecosystems. Based on NEP, the NBP in each grid cell can be further calculated as:

$$NBP = NEP + C_{transin} - C_{transout} - C_{harvest} - C_{LUC} \quad (8)$$

where C_{trans_in} is the C transfer from river to land through flooding waters. C_{trans_out} is the C loss from soil to river due to erosion and leaching. $C_{harvest}$ is the C loss due to harvest of crops and wood products. C_{LUC} is the C loss due to land use change.

We further investigated the spatiotemporal variation of LCT in different subregions of Europe because climate, land use and their trends over the last century vary drastically across Europe (Supplementary Figs. S6, S7 and Table S4). The whole Europe is divided into four subregions: (1) Northern Europe (EuN), which is dominated by the Nordic climate; (2) Middle-Western Europe (EuMW), which is dominated by the oceanic climate; (3) Middle-Eastern Europe (EuME) which is dominated by the continental climate; and (4) Southern Europe (EuS) which is dominated by the Mediterranean climate. Note that, to keep the mass balance of LCT in each catchment, we divided the four subregions based on not only the climate and land use in each grid cell, but also the river networks. All grid cells belonging to the same catchment were partitioned to one subregion (i.e., no catchment spans more than one subregions). Same to the analysis of LCT over whole Europe, we analyzed the spatiotemporal variation of the amount and composition of LCT in each of the four subregions, as well as the respective contributions of different global change factors to the changes in LCT, and the impact of LCT on the terrestrial C balance in the four subregions.

Data availability

All forcing and validation data used in this study are publicly available online. The specific sources for these data can be found in Table S3. The source data for graphs and charts in this study can be obtained from https://github.com/hchzhang/COMMSENV-22-0446A. All source data produced by our simulations are available from the corresponding author on reasonable request.

Code availability

The source code of ORCHIDEE-Clateral model used in this study is available online (https://doi.org/10.14768/f2f5df9f-26da-4618-b69c-911f17d7e2ed) from 22 July 2019.

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Author contributions

H.Z., P.R., and R.L. designed this study. H.Z. performed the specific simulation and data analysis, with help from P.R. and R.L. P.C., K.V.O., and B.G. contribute to the model evaluation and result visualization. H.Z., P.R. and R.L. wrote most of the manuscript, with substantial contributions from P.C., K.V.O., and B.G. All authors reviewed and revised the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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