- Estimating lateral nitrogen transfers over the last century
 through the global river network using a land surface model
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Abstract. Lateral nitrogen (N) transport from land to oceans through rivers is an 14 important component of the global N cycle. We developed a new model of this 15 aquatic system, called LSM Nlateral Off, which simulates the routing of water 16 in rivers, and the pertaining transport of dissolved inorganic N (DIN), dissolved 17 organic N (DON) and particulate organic N (PON) as well as the accompanying 18 biogeochemical processes of DON and PON decomposition, and denitrification 19 during transit from land to oceans through the global river network. Evaluation 20 against global observation-based datasets shows that the model effectively 21 captures both the magnitude and seasonal variations of riverine water discharges 22 and total nitrogen (TN) flows. Our model was then applied to reconstruct the 23 historical evolution of global N flows and transformations from land to rivers and, 24 ultimately, the oceans. Model simulation results indicate that, driven by 25 anthropogenic activities (e.g. application of mineral fertilisers and manure, 26 sewage water injection in rivers and land use change) and indirect effects of 27 climate change and rising atmosphere CO₂, TN exports increased from 27.5 Tg 28 N yr⁻¹ during the 1901-1920 period to 40.0 Tg N yr⁻¹ during the 1995-2014 period, 29 with DIN contributing most (80%) of this increase. Simulation results reveal 30 substantial spatial heterogeneities in annual mean TN flows and denitrification 31 rates while their seasonal amplitude is of similar magnitude as the large-scale 32 spatial variability. Compared to previously published regional or global aquatic 33 Ν transfer models (IMAGE-GNM, FrAMES-N, MBM, DLEM and 34 GlobalNEWS2), our model produces similar global and continental-scale TN 35 exports to the ocean, but shows distinct patterns at the finer scale of river basins. 36 LSM Nlateral Off is here coupled to the Land Surface Model (LSM) 37 ORCHIDEE, but the offline approach implemented in this work facilitates its 38 coupling with other land surface models in the future such as those synthesised 39 by the Nitrogen Model Intercomparison Project (NMIP). Our modelling approach 40 provides a comprehensive simulation of N transport and transformations from 41

terrestrial ecosystems to oceans at 0.5° spatial resolution and daily temporal
resolution, globally.

44 1. Introduction

Reactive nitrogen (N) is a vital element for all life on Earth, playing a 45 fundamental role in biological processes. The nitrogen cycle interacts with the 46 Earth's climate system and environment in multiple ways. One notable 47 interaction is through nitrous oxide (N₂O), a potent greenhouse gas that 48 influences the Earth's energy balance in a similar way as carbon dioxide (CO₂), 49 but with a global warming potential nearly 300 times greater on a per-molecule 50 basis (Sainju et al., 2014). N also plays a critical role in the C cycle, influencing 51 CO₂ and CH₄ fluxes by limiting primary production rates in many terrestrial, 52 freshwater, and marine ecosystems (Thornton et al., 2007; Morée et al., 2013; 53 Zaehle et al., 2014; Seiler et al., 2024). As a result, the N cycle is a key 54 regulator of the C cycle and climate change. This role underscores the need for 55 a comprehensive analysis of N dynamics in the context of a changing C cycle, 56 shifting climate conditions, and intensifying anthropogenic activities. 57

From an earth system perspective, the critical connection between 58 terrestrial and marine nitrogen (N) cycles via the Land-to-Ocean Aquatic 59 Continuum (LOAC) has been insufficiently addressed (Galloway et al., 2003; 60 Billen et al., 2013; Maranger et al., 2018; Battin et al., 2023). Existing studies 61 have largely treated the land and open ocean cycles separately, ignoring the N 62 processes occurring along the LOAC (Fowler et al., 2013; Zhang et al., 2021). 63 The representation of N processes within the LOAC is however required to 64 achieve a dynamic coupling between land surface and ocean biogeochemical 65 models, as this route plays a pivotal role in controlling the coupled terrestrial C-66 N cycles and their perturbations from anthropogenic activities (Gruber & 67 Galloway, 2008; Regnier et al., 2013; 2022). Over the past several decades, the 68

cumulative effects of climate change, population growth, industrialization and 69 increased use of agricultural fertilisers have accelerated the global N cycle, and 70 hence increased N leaching into the aquatic environment (Bouwman et al., 71 2005; Gruber & Galloway, 2008; Kim et al., 2011; Swaney et al., 2012; Beusen 72 et al., 2016a). This has resulted in negative human health and environmental 73 impacts, such as the degradation of drinking water quality and an increase in the 74 frequency and severity of eutrophication events (Dodds & Smith, 2016; Huang 75 et al., 2017; Costa et al., 2018; Lee et al., 2019; Dai et al., 2023). Most land 76 surface models (LSMs) include N leaching into aquatic systems; however, this 77 process is rarely evaluated in quantitative terms using observations collected 78 within the fluvial network. It has been shown that N leaching is inaccurate in 79 most LSMs (Feng et al., 2023), which in turn affects the simulation of the 80 response of terrestrial C and N cycles to anthropogenic activities and climate 81 change (Thomas et al., 2013). Furthermore, an explicit representation of the fate 82 of the land-derived N inputs into the LOAC is required to better constrain the 83 response of the ocean C cycle to increased nutrient inputs (Lacroix et al., 2021; 84 Resplandy et al., 2024) as well as to assess the extent to which N pollution 85 reduction scenarios can mitigate (Satter et al., 2014) eutrophication in riverine 86 and coastal aquatic ecosystems (Hashemi et al., 2016; Desmit et al., 2018, 87 Battin et al., 2023). 88

The representation of N lateral transfers through aquatic systems is 89 challenging as it requires to represent multiple N sources, transformation, 90 transport, and retention processes along the global fluvial network. A variety of 91 models with different structures and representations of the water and N cycles 92 have been developed to address this complexity (Luscz et al., 2015, 2017). 93 Models such as the Soil and Water Assessment Tool (SWAT) (Arnold et al., 94 1998; Liu et al., 2017), the Hydrologic Simulation Program-FORTRAN (HSPF) 95 (Bicknell et al., 2005; Wang et al., 2015) and the HYdrological Predictions for 96

the Environment (HYPE) (Lindström et al., 2010; Donnelly et al., 2014) were 97 designed to represent hydrological processes as well as N transport and 98 transformation in rivers, but mainly for catchment scale applications. Therefore, 99 their complexity and high requirements for hard-to-get forcing datasets 100 constrain their applicability, in particular for the long-term evolution of global 101 N fluxes and transformation processes. Simplified empirical approaches provide 102 an alternative for large-scale simulations. For instance, the Global Nutrient 103 Export from Watersheds 2 (GlobalNEWS2) model allows to estimate riverine N 104 exports to the ocean as a function of N deliveries from the surrounding 105 catchment with a highly simplified representation of N transport and in-stream 106 N processes (Seitzinger et al., 2005; Mayorga et al., 2010; Lee et al., 2016). The 107 Integrated Model to Assess the Global Environment-Global Nutrient Model 108 (IMAGE-GNM) provides a more process-based representation of the river 109 networks as it relies on a globally distributed, spatially explicit hydrological 110 model, PCR-GLOBWB (PCR aster Global Water Balance), to estimate N 111 delivery to surface waters and its subsequent transport (Beusen et al. 2015, 112 2016a & 2022; Vilmin et al., 2018). This model however still simulates N 113 retention using empirical formulas and is not dynamically coupled with 114 vegetation-soil N processes. Furthermore, it only provides annually averaged 115 fluxes, hence ignoring the seasonal fluctuations induced by the hydrology and N 116 cycling on land and in the river network. The Dynamic Land Ecosystem Model 117 (DLEM 2.0) provides a significant advancement as it simulates riverine N flow 118 from terrestrial ecosystems to rivers and coastal oceans using a unified process-119 based representation. So far, however, the model's simulation of N lateral 120 transfer has only been evaluated at the regional scale, specifically in eastern 121 North America (Yang et al., 2015), or for N₂O emissions on the global scale 122 (Tian et al. 2018; Yao et al., 2020). To complement these studies, we develop 123 here a new N lateral transfer model that can be linked to the outputs of different 124 LSMs. This model captures the hydrological dynamics and N transformation 125

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processes in the global river network at a temporal resolution from days to 126 months, that is, at a temporal resolution relevant for biogeochemical processes 127 in coastal and marine ecosystems. At the same time, this model has the capacity 128 to reconstruct and forecast the long-term (decadal to century-scale) evolution of 129 the aquatic N cycle as a result of a wide variety of anthropogenic factors, 130 including climate change. To achieve this aim, we apply an offline approach in 131 which lateral N transfers are constrained by outputs from an LSM. The resulting 132 model, called LSM Nlateral Off, is in the present study coupled to the 133 ORCHIDEE, a LSM developed by the Institute Pierre-Simon Laplace (IPSL, 134 France). 135

ORCHIDEE is a widely used land surface model (Krinner et al., 2005), 136 with many versions (or branches) focusing on different aspects of the terrestrial 137 C cycle and associated bio-elements. Here, we leverage ORCHIDEE-CNP, the 138 branch simulating the coupled cycles of carbon (C), N and phosphorus (P) in the 139 terrestrial biosphere (Sun et al., 2021), and ORCHIDEE-Clateral, the branch 140 simulating the leaching and erosion of C along the soil-inland water continuum 141 (Lauerwald et al., 2017, 2020; Zhang et al., 2022). Our study is structured as 142 follows: (1) we present the development of the offline N lateral transfer model 143 (LSM Nlateral Off) driven by outputs from ORCHIDEE-Clateral and 144 ORCHIDEE-CNP; (2) we evaluate our model using a collection of water 145 discharge and N concentration observations; (3) we investigate the spatio-146 temporal dynamics of N lateral transfers over the historical period (1900-2014); 147 and (4) we compare model results with those obtained from previously 148 published models. 149

150 **2. Methods and Data**

151 **2.1. Model development**

152 **2.1.1. The LSM_Nlateral_Off model**

The LSM, here ORCHIDEE, comprehensively simulates the cycling of 153 energy, water and C in terrestrial ecosystems (Krinner et al., 2005). As the 154 model evolved, many versions (or branches) emerged with various foci on 155 additional land surface processes impacting the climate system. In particular, 156 the ORCHIDEE-CNP branch features a detailed representation of the coupled 157 cycling of C, N, and P within vegetation and soil (e.g. root uptake of N, the 158 allocation of N in the tissue of different parts of vegetation biomass, N turnover 159 in litter and soil organic matter) and the leaching of NH_4^+ and NO_3^- from soils to 160 inland waters (Goll et al., 2017, 2018; Sun et al., 2021). The ORCHIDEE-161 Clateral branch simulates the large-scale lateral transfer and fate of water, 162 sediment, particulate organic carbon (POC) and dissolved organic C (DOC), 163 and CO₂ along the land-river-ocean continuum (Lauerwald et al., 2017; Hastie 164 et al., 2019; Bowring et al., 2020; Zhang et al., 2022). 165

Based on the land-to-river inputs of water, POC, DOC and inorganic N 166 simulated by ORCHIDEE-CNP and ORCHIDEE-Clateral, we developed 167 LSM Nlateral Off (Land Surface Model Nitrogen lateral Offline), simulating 168 the transfers and transformations of reactive N through the global river network. 169 The offline strategy provides a computationally efficient numerical model in 170 which the mathematical representation of aquatic biogeochemical processes can 171 easily be implemented, calibrated and evaluated. Furthermore, by construction, 172 it can also be used to route the N leaching fluxes produced by any other LSMs 173 in the future, allowing for applications at various scales and across different 174 regions. In this offline scheme, ORCHIDEE-CNP provides as input the leaching 175 rates of terrestrial dissolved inorganic N (DIN) with surface runoff and subsoil 176 drainage and dissolved organic N (DON) leaching from manure. Inputs of 177 terrestrial DON and particulate organic N (PON) are derived from the leaching 178 and erosional fluxes of DOC and POC simulated by ORCHIDEE-Clateral and 179

stoichiometric C:N ratios of dissolved organic matter (DOM) and particulate
organic matter (POM); please refer to section 2.1.2 for further details (Fig. 1).

N discharge from sewage is also included as an additional input to 182 LSM Nlateral Off, using the N sewage dataset (1900-2010, gridded maps 183 every five years) reported by Beusen et al. (2016b). Indeed, during the twentieth 184 century, global N (DIN and DON) discharge from sewage to surface waters has 185 increased about 3.5-fold to 7.7 Tg N yr⁻¹, and thus has a large impact on trends 186 in global N lateral transfers. Sewage-derived N comes from three main sources: 187 human waste from urban environments, animal waste, and industrial waste, 188 each of which follows distinct pathways. For further details, please refer to Van 189 Drecht (2009) and Morée et al. (2013). 190

Following delivery, PON, DON and DIN are then transported by water 191 flow advection from soils to rivers and through the river network all the way to 192 the coast. Within the river network, parts of the transported DON and PON are 193 decomposed into DIN, while part of the DIN is released back to the atmosphere 194 through denitrification. Following previous global modelling approaches 195 (Aitkenhead-Peterson et al., 2001; Bernot and Dodds, 2005; Wollheim et al., 196 2008), LSM Nlateral Off simulates the denitrification process without explicit 197 representation of the different DIN species (i.e. NO_3^- and NH_4^+) or their 198 interconversion via nitrification (Fig. 1). 199



200



203 2.1.2. Water and N delivery from soils to the river network

LSM Nlateral Off was developed to simulate N lateral transfer and 204 transformation during 1901-2014 in this study. The runoff and drainage simulated 205 by ORCHIDEE-Clateral were used to constrain water inputs from land to rivers. 206 This input dataset had a spatial resolution of 1° and a temporal resolution of daily 207 time steps (Table 1). The data were downscaled to the LSM Nlateral Off spatial 208 resolution of 0.5° using nearest-neighbour resampling (Patil, 2018). Runoff and 209 drainage are critical components that determine DIN, DON, and PON fluxes. 210 ORCHIDEE-CNP and ORCHIDEE-Clateral used the same scheme to simulate 211 soil hydrology (Sun et al., 2021; Zhang et al., 2022), and they have been run with 212 the same climate forcing data, land cover map and soil parameters maps (Table 213 1). The climate forcing data during 1901-2014 were obtained from Global Soil 214 Wetness Project Phase 3 (GSWP 3). Both ORCHIDEE-CNP and ORCHIDEE-215 Clateral used the ESA-CCI LUH2v2 plant functional type (PFT) distribution, 216 which combines the ESA-CCI land cover map for 2015 with the historical land 217 cover reconstruction from LUH2 (Lurton et al., 2020). Soil parameters in these 218 two models follow Reynolds et al. (1999) and the Harmonized World Soil 219 Database (FAO/IIASA/ISRIC/ISS-CAS/JRC, 2012). Therefore, the differences 220

in runoff (0.9%) and drainage (1.7%) simulated by the two ORCHIDEE branches 221 are relatively small (Fig. S1). 222

The lateral transfer of DOC and POC from land to rivers was used to 223 constrain inputs of DON and PON. PON erosion with runoff originates from 224 three soil organic matter (SOM) pools, each characterized by distinct C:N 225 ratios, set at 12, 25, and 8 for active, slow, and passive SOM pools, respectively 226 (Zhang et al., 2022). The PON erosion from each pool is calculated by dividing 227 the POC erosion flux from the same SOM pool by its corresponding C:N ratio. 228 For DON leaching with runoff and drainage, the calculation relies upon 229 measurements of the stoichiometry of dissolved organic matter, which report 230 C:N ratios in soil and rivers comprised between 8 and 25, with an average value 231 of around 12 (Kirkby et al., 2011; Lutz et al., 2011; Tipping et al., 2016; 232 Maranger et al., 2018; Rodríguez-Cardona et al., 2021). Therefore, the leaching 233 of DON with runoff and drainage was quantified using the DOC fluxes 234 simulated by ORCHIDEE-Clateral, and an average C:N ratio of 12. It is 235 important to note that this resulting flow excludes DON leaching sourced from 236 manure application, as this source is not included in the ORCHIDEE-Clateral 237 simulations. The spatial and temporal resolution of the resulting DON and PON 238 fluxes used to force LSM Nlateral Off was 1° with a daily time step (Table 1) 239 and these inputs were resampled to the nominal resolution of 240 LSM Nlateral Off (0.5°) using the nearest-neighbour resampling (Patil, 2018). 241 DIN (i.e. NH_4^+ and NO_3^-) inputs from soils to rivers were prescribed from 242 a simulation of ORCHIDEE-CNP (Goll et al., 2017a, 2018; Sun et al., 2021) 243 which include DIN leaching from both natural and cultivated (e.g. cropland and 244 pasture) ecosystems, and account for changes induced by atmospheric N 245

deposition, fertiliser use and manure application. DON inputs to rivers from manure application were also prescribed using ORCHIDEE-CNP. The approach 247 relies on a DON pool and a leaching factor, with a dedicated manure-derived 248

246

249	DON pool incorporated into ORCHIDEE-CNP to participate in subsequent N
250	cycling and leaching processes. The spatial and temporal resolution of this input
251	dataset was 2° with a daily time step and the data were downscaled to the
252	LSM_Nlateral_Off spatial resolution of 0.5° using the nearest-neighbour
253	resampling (Patil, 2018) (Table 1).
254	Finally, TN inputs from sewage (https://doi.org/10.17026/dans-zgs-
255	<u>9k9m</u>), provided at 0.5° globally with a yearly time step (Beusen et al, 2016b),
256	were evenly redistributed across each day of the year (Table 1). TN from
257	sewage was then partitioned into different N species following the approach of
258	Naden et al. (2016), which assumes that 10% of sewage TN is DON and the
259	remaining 90% is DIN.

Table 1. List of (1) forcing data used to run ORCHIDEE-Clateral, ORCHIDEE-CNP and LSM_Nlateral_Off, and (2) observational data used to evaluate the simulation results. S_{res} and T_{res} are the original spatial and temporal resolutions of the forcing data, respectively.

	Data	Sres	T _{res}	Data source
	Climatic forcing data (precipitation, temperature, incoming shortwave/longwave radiation, air pressure, wind speed, relative humidity)	1°	3 hours	Global Soil Wetness Project Phase 3 (GSWP 3) (Kim et al., 2017)
Forcing data of	Land cover	0.5°	1 year	ESA-CCI LUH2v2 (Lurton et al., 2020)
ORCHIDEE- Clateral and	Soil texture class	0.5°	/	Reynolds et al. (1999)
ORCHIDEE- CNP	Soil bulk density and pH	30"	/	HWSD v1.2 (FAO/IIASA/ISRIC/IS SCAS/JRC,2012)
	Fertiliser application	0.5°	1 year	(Lu et al., 2017)
	Manure application	5'	1 year	(Zhang et al., 2017)
	Nitrogen deposition	0.5	1 year	IGAC/SPARC CCMI

	Runoff			
	Drainage DOC and POC with runoff DOC and POC with	1°	1 day	ORCHIDEE-Clateral (Zhang et al., 2022; Zhang et al., under
	drainage			review)
	Soil temperature			
Forcing data of LSM-Nlateral -Off	DIN with runoff and drainage	1°	1 day	ORCHIDEE-CNP (Sun et al., 2021)
	DON leaching from manure application			
	DIN and DON with sewage	0.5°	5 years	(Beusen et al., 2016b)
	Flow direction Topographic index (<i>f</i> _{topo})	0.5°	/	(Vörösmarty et al., 2000)
	Riverine water discharge	/	1 day	GRDC ^a
Evaluation data	Riverine TN and NO ₃ ⁻ concentration	/	point measurement	GRQA ^b
	Riverine TN concentration	/	point measurement	Table S1

^a Global Runoff Data Centre (GRDC) (Federal Institute of Hydrology, 2018); ^b Global River
water Quality Archive (GRQA) (Virro et al., 2021).

266 2.1.3. N transport and transformation in the river network

LSM Nlateral Off simulates water discharge using a distributed routing 267 scheme (Vörösmarty et al., 2000). As shown in Fig. 2, surface runoff (F_{RO}) and 268 belowground drainage (F_{DR}), both derived from ORCHIDEE-Clateral, serve as 269 inputs to the LSM Nlateral Off. F_{RO} first feeds into the "fast" reservoir 270 (S_{fast H2O}), while F_{DR} feeds into the "slow" water reservoir (S_{slow_H2O}). The 271 delayed outflows from these reservoirs then feed into the "stream" water 272 reservoir ($S_{\text{stream H2O}}$). Water in the stream reservoir ($S_{\text{stream H2O}}$) in grid cell *i* 273 then flows downstream into the stream reservoir of grid cell i+1 (F_{streamout H2O}, 274 $m^3 d^{-1}$). The outflow rates from the fast (F_{fastout_H2O}), slow (F_{slowout_H2O}) and 275

stream ($F_{streamout_H2O}$) reservoirs are calculated at a daily time-step based on a grid-cell-specific topographic index f_{topo} (unitless, Vörösmarty et al., 2000) (Table 1) and a reservoir-specific water turnover factor τ , which translates f_{topo} into a water residence time for each reservoir attached to each river segment (Eq. 1).

$$281 \quad F_{out_H2O} = \frac{S_{H2O}}{\tau \times f_{topo}} \tag{1}$$

where F_{out_H2O} (m³ d⁻¹) represents water outflow rates from the fast ($F_{fastout_H2O}$) /slow ($F_{slowout_H2O}$) /stream ($F_{streamout_H2O}$) reservoir; S_{H2O} (m³) represents water stock in the fast (S_{fast_H2O}) /slow (S_{slow_H2O}) /stream reservoir (S_{stream_H2O}); τ represents water residence time for each reservoir, equal to 3.0 days, 25.0 days and 0.24 days for the fast, slow, and stream reservoirs, respectively (Ngo-Duc et al., 2006); f_{topo} represents the grid-cell-specific topographic index (unitless, Vörösmarty et al., 2000).





Figure 2. Schematic plot for the reservoirs and flows of water and N in

291 LSM_Nlateral_Off. S_{soil} is the soil pool. S_{fast}, S_{slow}, S_{stream} are the "fast", "slow"

and stream water reservoirs, respectively. F_{RO} and F_{DR} are surface runoff and

below-ground drainage (also called sub-surface runoff in other studies),

- respectively. $F_{fastout}$ is the flow from fast reservoir to stream reservoir. $F_{slowout}$ is
- the flow from slow reservoir to stream reservoir. $F_{upstream}$ and $F_{streamout}$ are the

upstream inputs from basin i-1 and downstream outputs to basin i+1, respectively. F_D is the wet and dry deposition of DIN from the atmosphere.

Following the routing scheme of water in LSM Nlateral Off, N 298 contained in surface runoff (F_{RO}) and belowground drainage (F_{DR}) flows into the 299 fast and slow reservoir, respectively. Subsequently, the N stocks in these 300 reservoirs are subject to decomposition and losses via denitrification, which are 301 governed by the water residence time. The remaining fractions further flow into 302 the stream reservoirs, which also receive direct N inputs from sewage (Fig. 2). 303 Within stream reservoirs, N is transformed by biogeochemical reactions and 304 flows from one grid cell to the next along the river routing scheme. The 305 timescale of these biogeochemical transformation processes scales to the water 306 residence time (and hence topography) within the river network, and the 307 fraction of N that is not lost to the atmosphere via denitrification is ultimately 308 exported to the coast. Biogeochemical reactions within each reservoir include 309 the decomposition of PON and DON to DIN, and the denitrification of DIN to 310 N gas which is assumed all released into the atmosphere (Fig. 2). The mass 311 balance equations for the N stocks in different reservoirs are calculated as 312 follows: 313

314
$$\frac{dS_{fast_PON}}{dt} = F_{RO_PON} - F_{fastout_PON} - R_{fast_PON}$$
(2)

315
$$\frac{dS_{fast_DON}}{dt} = F_{RO_DON} - F_{fastout_DON} - R_{fast_DON}$$
(3)

316
$$\frac{dS_{fast_DIN}}{dt} = F_{RO_DIN} - F_{fastout_DIN} - R_{fast_DIN}$$
(4)

317
$$\frac{dS_{slow_DON}}{dt} = F_{DR_DON} - F_{slowout_DON} - R_{slow_DON}$$
(5)

318
$$\frac{dS_{slow_DIN}}{dt} = F_{DR_DIN} - F_{slowout_DIN} - R_{slow_DIN}$$
(6)

319
$$\frac{dS_{stream_PON}}{dt} = F_{fastout_PON} + F_{upstream_PON} - R_{stream_PON} - R_{str$$

(7)

(8)

320 $F_{downstream_PON}$

321
$$\frac{dS_{stream_DON}}{dt} = F_{fastout_DON} + F_{slowout_DON} + F_{upstream_DON} + F_{sewage_DON} - F_{sewage_DON} - F_{stream_DON} + F_{st$$

322 $R_{stream_DON} - R_{downstream_DON}$

323
$$\frac{dS_{stream_DIN}}{dt} = F_{fastout_DIN} + F_{slowout_DIN} + F_{upstream_DIN} + F_{sewage_DIN} + F_{se$$

324
$$R_{stream_PON} + R_{stream_DON} - R_{stream_DIN} - F_{downstream_DIN}$$
(9)

- where $F_{upstream_PON}(g N d^{-1})$, $F_{upstream_DON}(g N d^{-1})$ and $F_{upstream_DIN}(g N d^{-1})$
- represent the inflow rates of PON, DON and DIN from upstream grids,
- respectively; $F_{streamout_PON}(g N d^{-1})$, $F_{streamout_DON}(g N d^{-1})$ and $F_{streamout_DIN}(g N d^{-1})$
- ¹) represent outflow rates of PON, DON and DIN from the given grid to
- downstream grid, respectively. For each N species, the N inputs to a stream
- reservoir in a given grid cell ($F_{upstream_PON}$, $F_{upstream_DON}$ and $F_{upstream_DIN}$) are equal
- to the sum of N outflow from the upstream stream reservoir in the adjacent grid
- cells ($F_{streamout_PON}$, $F_{streamout_PON}$ and $F_{streamout_PON}$), as calculated in Eq. 10.
- 333 R_{fast_PON} and R_{stream_PON} (g N d⁻¹) represent PON decomposition rates in the fast
- and stream reservoirs, respectively. R_{fast_DON} , R_{slow_DON} and R_{stream_DON} (g N d⁻¹)
- represent DON decomposition rates in the fast, slow and stream reservoirs,
- respectively. R_{fast_DIN} , R_{slow_DIN} and R_{stream_DIN} (g N d⁻¹) represent DIN

denitrification rates in the fast, slow and stream reservoirs, respectively.

We assume that N concentrations are homogeneously distributed within each reservoir of each grid and that N transfers between reservoirs simply follow that of water. N transfers are calculated as follows:

341
$$F_{out_N} = S_N \times \frac{F_{out_H_2O}}{S_{H_2O}}$$
 (10)

Where S_{H2O} represents water stocks (m³), and F_{H2O} represents flow rates of water (m³ d⁻¹). F_{out_N} represents PON flow rates from fast ($F_{fastout_PON}$) / stream 344 ($F_{streamout_PON}$) reservoirs, DON flow rates from fast ($F_{fastout_DON}$) / slow

345 $(F_{slowout DON})$ / stream $(F_{streamout DON})$ reservoirs, DIN flow rates from fast

346 (F_{fastout DIN}) / slow (F_{slowout DIN}) / stream (F_{streamout DIN}) reservoirs. The same

principle applies to the S_N (N stocks) terms.

Temperature controls the decomposition rates of organic N in rivers (Ferreira et al., 2020). Following the algorithm of Xia et al. (2013), the decomposition rates of PON and DON in each reservoir are calculated using first-order kinetics of the corresponding N stock and a Q10 temperature dependence based on water temperature.

353
$$R_{ON} = S_{ON} \times K_{ON} \times Q10^{\frac{TW - T_{ref1}}{10}}$$
 (11)

 R_{ON} (g N d⁻¹) represents decomposition rate of organic N (ON, i.e., PON and 354 DON); S_{ON} (g N) represents ON stocks in each reservoir. K_{ON} represents the 355 average PON decomposition rate ($K_{PON} = 0.028 \text{ d}^{-1}$) (Islam et al., 2012), and the 356 average DON decomposition rate ($K_{DON} = 0.07 \text{ d}^{-1}$) at the reference temperature 357 of 20°C in water (Xia et al., 2013). Q10 is the temperature sensitivity of PON 358 and DON decomposition rates set to 2.0 (Yang et al, 2015; Liu et al., 2021). TW 359 is the water temperature (°C). and T_{refl} is the reference temperature for PON and 360 DON decomposition, set to 20°C. R_{ON} (g N d⁻¹) represents PON decomposition 361 rates in fast (R_{fast_PON})/ stream ($R_{stream PON}$) reservoirs, and DON decomposition 362 rates in fast $(R_{fast DON})$ /slow $(R_{slow DON})$ / stream $(R_{stream DON})$ reservoirs. 363

The denitrification rates decrease with stream depth, because most denitrification happens in benthic sediments rather than in the water column, so high benthic area to water volume ratios result in high denitrification rates Aitkenhead-Peterson et al., 2005; Bernot and Dodds, 2005). In addition, denitrification rates are also controlled by temperature (Jung et al., 2014; Ma et al., 2022). The denitrification process is simulated by adapting equations from Pauer and Auer (2008):

371
$$R_{DIN} = \frac{S_{DIN}}{depth} \times K_{DIN} \times F_{T_DIN}$$
(12)

372
$$F_{T_DIN} = e^{\frac{-(TW - T_{ref2})^2}{(T_{ref2})^2}}$$
 (13)

373
$$depth = max (e^{2.56} \times Q^{0.423}, 1.0)$$
 (14)

where R_{DIN} (g N d⁻¹) represents denitrification rates in fast (R_{fast_DIN})/ slow 374 $(R_{slow DIN})/stream (R_{stream DIN})$ reservoirs; $K_{DIN}(0.15 d^{-1})$ represents the 375 denitrification rate in water at 25°C (Alexander et al., 2009); $F_{T DIN}$ (unitless) 376 represents the dependency of denitrification on temperature (Ma et al., 2022); 377 T_{ref2} is the reference temperature for denitrification (=25°C); $\frac{1}{denth}$ (unitless) 378 represents the factor that simulates the role of the benthic surface area to water 379 volume ratio, which serves as a key control factor of denitrification rates. The 380 stream depth is simulated according to the method in Raymond et al. (2012). 381 Therefore, aside from the availability of DIN stocks, denitrification rates are 382 spatially and temporally dependent through the effects of water residence time 383 (controlled by topography), temperature and water depths (controlled by 384 discharge). Tables A1 and A2 provide a summary of all variables, fluxes and 385 processes incorporated in LSM Nlateral Off. 386

387 2.2. Observational data

Riverine water discharge from the Global Runoff Data Centre (GRDC) 388 (Federal Institute of Hydrology, 2018) and riverine TN and NO₃⁻ concentrations 389 from the Global River water Quality Archive (GRQA) (Virro et al., 2021) were 390 used to evaluate LSM Nlateral Off (Fig. 3). We obtained observed water 391 discharge data from the GRDC website for 346 gauging stations with a 392 catchment area exceeding 50,000 km². Each station has over 12 months of 393 observational records and more than 25 observations per month (Fig. S4). For 394 GRQA data, only time-series with more than two observations per month in one 395 year were retained for model evaluation. For N concentrations, after removing 396

duplicates in the GRQA database, we obtained TN data for 3507 sites and NO₃⁻ 397 data for 1841 sites. Moreover, since observations of NO_3^- at a given site are 398 generally more frequent and cover a longer time span than those for TN, we 399 used the strong correlation between these two species to estimate TN 400 concentrations from NO_3^- when only NO_3^- data were available (represented by 401 yellow dots in Fig. 3). The prediction equation applied in this study (Eq. 15, 402 Fig. S2) was obtained based on GRQA data at 148 sites with simultaneous 403 concentrations of both TN and NO_3^- (R²=0.78): 404

405
$$C_{TN_obs} = 1.33 \times C_{NO3_obs} + 0.56$$
 (15)

406 where C_{TN_obs} (mg L⁻¹) and C_{NO3_obs} (mg L⁻¹) represent the observed 407 concentrations of TN and NO₃⁻, respectively.

The TN flow rates are equal to the water discharge rates multiplied by N concentrations. Therefore, for each GRDC site, the nearest GRQA site with reported N concentration (McDowell et al., 2021) was systematically selected to calculate the flux. We first calculated the monthly average N concentrations and monthly total water discharge, then determined the monthly N fluxes using Eq. 16. The total annual N flow is then obtained by summing the monthly N fluxes over the entire year.

415
$$F_{TN_obs} = F_{W_obs} \times C_{TN_obs} \tag{16}$$

416 where F_{TN_obs} (g N d⁻¹) and F_{W_obs} (m³ d⁻¹) represent observed rates of TN flow 417 and water discharge, respectively. This calculation was

Since TN concentrations for several large rivers (e.g., Amazon and
Chinese rivers) were missing in GRQA, we complemented this dataset by
collecting additional observational TN data from peer-reviewed literature
(represented by green dots in Fig. 3), resulting in the addition of 20 sites to our
database, see details of observed sites in Table S1.



Figure 3. Location of observational sites for N concentrations. Pink dots
represent sites with observations of total nitrogen (TN) concentrations,116 sites;
yellow dots represent sites with observations of NO₃⁻ concentrations, 53 sites;
green dots represent sites with observations of TN concentrations from
published literature, 20 sites (Table S1). Black stars represent sites with daily
time series of water discharge and TN flow.

+25 time series of water discharge and 110 now.

430 2.3. Simulation protocol and analysis of model results

431 **2.3.1. Simulation protocol**

423

LSM Nlateral Off was applied to simulate the lateral transfer of PON, 432 DON and DIN, as well as the decomposition of PON and DON, and the loss of 433 DIN by denitrification within the river network from 1901 to 2014. The model 434 was run at 0.5° spatial resolution and daily temporal resolution, using the 435 downscaled terrestrial forcings as inputs (see section 2.1.2). Running 436 LSM Nlateral Off on a daily step allows for the evaluation of the model's 437 performance in capturing not only long-term trends but also seasonality in 438 lateral N transfers and transformations within the global river network. The 439 model was evaluated on a daily time step by comparing the simulated and 440 observed TN lateral transfer at three sites with long time series of observed TN 441 flows. We also evaluated the performance of LSM Nlateral Off in simulating 442 annual lateral TN transfer using observational data from 189 sites worldwide, 443 each with records of both water discharge rates and N concentrations. The 444

simulated total amounts of PON, DON and DIN from land to river and from 445 river to ocean were further compared with previously published global N 446 models, namely IMAGE-GNM (Vilmin et al., 2018), the Frame-work for 447 Aquatic Modeling in the Earth System (FrAMES-N) (Wollheim et al., 2008), 448 the Mass Balance Model (MBM) (Green et al., 2004), and GlobalNEWS2 449 (Mayorga et al., 2010). 450

Table 1 summarises the forcing and evaluation data along with their 451 spatiotemporal resolution and references to the gridded products and point 452 datasets. 453

2.3.2. Model evaluation metrics 454

To evaluate the performance of LSM Nlateral Off in reproducing the 455 spatial variations of water and N flow, the mean bias error (MBE) and the 456 coefficient of determination (R^2) were determined. R^2 represents how much 457 variation in the observations can be explained by the model. For the definition 458 of R², please refer to Renaud et al. (2010). MBE quantifies the degree to which 459 LSM Nlateral Off overestimates or underestimates observations of water 460 discharge and TN flow at the grid level. 461

462
$$MBE = \frac{M-O}{O} \times 100\%$$
 (17)

where *M* is the mean of simulated values, *O* is the mean of observed values. 463

To assess the performance of LSM Nlateral Off in reproducing time 464 series of TN and water flows, the relative root mean square root (RRMSE) and 465 Nash-Sutcliffe coefficient (NSE) were calculated. 466

$$RRMSE = \frac{\sqrt{\frac{\sum_{j=1}^{n} (M_{j} - O_{j})^{2}}{n}}}{\bar{O}} \times 100\%$$
(18)

$$NSE = 1 - \frac{\sum_{j=1}^{n} (O_j - M_j)^2}{\sum_{j=1}^{n} (O_j - \bar{O})^2}$$
(19)

468

where *n* represents the total number of days/months with available observations 469 at a given site; O_i and M_i represent the observed and modelled values of 470 water/TN flow on day/month *j*. The NSE can take values between 1 and $-\infty$. An 471 NSE of 1 indicates a perfect fit between observed and simulated values, an NSE 472 of 0 means that using the mean observed value as a constant simulated value 473 would lead to as much deviation between observed and predicted values as 474 using the actual simulated values. If the NSE is negative, there is more 475 deviation between simulated and observed values than between the observed 476 values and their mean. 477

478 **2.3.3. Seasonality analysis**

To explore the seasonal variability of water discharge, TN flow, TN 479 concentration and denitrification rates during 1995-2014 at the global scale, we 480 constructed spatial maps of monthly anomalies following the method by 481 Roobaert et al. (2019). If FV denotes the rate of water flow (km³ yr⁻¹), 482 denitrification (Gg N yr⁻¹), TN flow (Gg N yr⁻¹) or TN concentration (mg L⁻¹) in 483 rivers, then for each grid cell, the monthly anomaly of FV can be calculated as 484 the difference between the FV value in a given month and the corresponding 485 annual mean value: 486

$$487 \quad FVA_t = FV_t - \overline{FV} \tag{20}$$

where FVA_t represent the anomaly of FV in month *t*, while FV_t and \overline{FV}_t represent the values of FV in month *t* and the annual mean, respectively.

The seasonality, defined as the amplitude of seasonal variations in water discharge, N flow rates, N concentrations and denitrification rates, is expressed as the root-mean-square (RMS) of the monthly *FVA*.

493
$$season_{FVA} = \sqrt{\frac{1}{12} \times \sum_{t=1}^{12} (FVA_t)^2}$$
 (21)

494 **3. Results and discussion**

495 **3.1. Model evaluation**

Evaluation of the simulated water discharge using GRDC data indicates 496 that for major rivers with drainage areas larger than 50 000 km² spread over the 497 globe, LSM Nlateral Off reproduces the magnitude and seasonal variations of 498 water discharge well. Overall, the model simulation explains 90% of the spatial 499 variations in the observed long-term average water discharges (Fig. 4a). The 500 absolute values of MBE for the simulated average water discharges are mostly 501 smaller than 50% (Fig. S3a). At 25 sites (13% of all sites), the absolute values 502 of MBE are larger than 100%, but the annual mean water discharge at each of 503 these sites is less than 100 km³ yr⁻¹ (about 3200 m³ s⁻¹), indicating that large 504 errors tend to occur at sites where water discharge is low (Fig. S3a). The 505 discrepancy between model simulations and observations at these sites may be 506 caused by three factors: (1) a potential discrepancy between the stream routing 507 scheme (delineation of catchment boundaries) defined by the 0.5° resolution 508 forcing data and the real river network; (2) the presence of stream channel 509 bifurcations that are poorly resolved by the model (Zhang et al., 2022); (3) 510 biases in runoff and drainage simulated by ORCHIDEE-Clateral, which may 511 stem from deviations in meteorological data and the parameterization of soil 512 hydraulic properties. At some sites, such as the Columbia, Rhine and 513 Mississippi Rivers for which continuous time series in TN flows are available, 514 LSM Nlateral Off also captures the seasonal variation in water discharges well, 515 with RRMSE ranging from 30% to 37% (Fig. 5 a1-a3). 516

517 Area-averaged TN flows simulated by LSM_Nlateral_Off are generally 518 consistent with observed TN flows at the 189 sites extracted from the GRQA

database and additional published literature. LSM Nlateral Off explains 77% 519 of the observed spatial variations of long-term TN flows across sites (Fig. 4b). 520 The absolute values of MBE for the simulated average TN flows are mostly 521 below 50% (Fig. S3b). LSM Nlateral Off significantly underestimated (MBE < 522 -100%) or overestimated (MBE > 100%) the observed TN flows at 32 sites 523 (17% of all sites), all located in regions with relatively low water discharge 524 (Fig. S3b). At 9 of these 32 sites (28%), the MBE of TN flow is very close to 525 that of water discharge, showing that discrepancies between observed and 526 modelled TN flows at these locations stem primarily from water discharge 527 rather than nitrogen concentrations. The results reveal that the MBE of TN flow 528 is relatively small in large rivers, such as at sites located in the lower reaches of 529 the Columbia, Rhine and Mississippi Rivers, where MBE values are -25%, -530 16% and 1%, respectively. LSM Nlateral Off also basically reproduces the 531 seasonal patterns of TN flow in these rivers, with RRMSE ranging from 30% to 532 62% (Fig. 5 ba-b3). At the Rhine River site, the NSE of TN flow is negative, 533 revealing that although the seasonal pattern of TN flow simulated by 534 LSM Nlateral Off is similar to that observed, the model does not capture 535 accurate trends on the daily scale (Fig. 5 b2). 536

The seasonality in water discharge is an important control factor for the 537 seasonality in TN fluxes. Therefore, the observational data derived from GRDC 538 was used to further assess the performance of LSM Nlateral Off in reproducing 539 the monthly seasonality of water discharge. At the 346 GRDC sites with 540 continuous measurements (Fig. S4), we computed the monthly average value, 541 taken as the observed water discharge of that month. For the world's 20 largest 542 rivers (Dai & Trenberth, 2002), which accounts for approximately 31% of the 543 total global river discharge (Table S2, Fig. S4), LSM Nlateral Off effectively 544 simulates both the magnitude and seasonality of water discharge (Fig. S5). The 545 Nash-Sutcliffe Efficiency (NSE) values range from 0.07 to 0.92, with 17 out of 546

the 20 rivers achieving an NSE greater than 0.5 (Fig. S5). However, the model
demonstrates a significantly weaker accuracy in capturing the seasonality of
water discharge in some low-flow rivers, with NSE values below zero at 84
(24% of the sites number contributing to 17% of the global river discharge) of
the 346 GRDC sites (Fig. S6). The model's limitations in capturing seasonality
are attributed to three main reasons, as discussed above.

As an additional evaluation, we compared our model results against 553 observed N concentrations and water discharges across the United States 554 provided by the U.S. Geological Survey (USGS). Based on these data, a 555 previous study (Scott et al., 2007) calculated the long-term (1975-2004) mean 556 annual loads of total organic N (TON) and TON fractions (TON yield / TN 557 yield) at 854 stations nationwide. LSM Nlateral Off simulates a spatial pattern 558 for the TON fraction which closely matches that reported by Scott et al. (2007), 559 with high values in western regions and low values in the east (Fig. S7). This 560 suggests that LSM Nlateral Off not only effectively simulates TN fluxes, but 561 also captures the organic and inorganic fractions across the United States 562 relatively well. Moreover, the simulated DIN concentrations display similar 563 spatial patterns as those obtained from a recent observation-based machine 564 learning (ML) assessment (Marzadri et al., 2021) in regions such as North 565 America, Western Europe, Eastern China, and India (Fig. S8). However, in 566 regions such as the Amazon, Africa, and Australia, LSM Nlateral Off 567 simulates lower DIN concentrations compared to the ML assessment (Fig. S8). 568 These lower DIN concentrations are attributed to different factors. In Australia, 569 low N inflow into rivers results in low DIN concentrations, whereas in the 570 Amazon and tropical rainforests of Africa, high denitrification rates are 571 primarily responsible for the low DIN concentrations in the model (Fig. 7). The 572 ML involves a significant degree of empirical modelling, and therefore does not 573 fully reflect real-world conditions. Therefore, this comparison cannot be 574

- regarded as a direct evaluation of the model based on observational data.
- 576 However, the consistency between the two models across most regions globally
- 577 (e.g., North America, Western Europe, Eastern China, and India) suggests that
- 578 LSM Nlateral Off overall performs reasonably well in simulating DIN lateral
- 579 transfer processes.



Figure 4. Evaluation of LSM_Nlateral_Off. Global-scale comparison between
observed and modelled annual-mean water discharge (a) and TN flow (b). Pink
symbols represent sites with observations of TN concentrations from GRQA,
yellow symbols represent GRQA sites for which TN concentrations were
estimated from observations of NO₃⁻ concentrations, and green symbols
represent sites with observations of TN from published literature.



Figure 5. Time series of water discharge (a) and TN flow (b). (a1) and (b1)
Columbia-river (46.18°N, 123.18°W); (a2) and (b2) Rhine-river (51.84°N,
6.11°E); (a3) and (b3) Mississippi river (32.25°N, -91.25°W).

591 **3.2.** Temporal and spatial patterns of N flows

Input data for LSM Nlateral Off are provided by ORCHIDEE-CNP and 592 ORCHIDEE-Clateral. Therefore, the magnitude and spatio-temporal patterns of 593 N inflows from land to rivers are exclusively derived from these two model 594 branches. In contrast, quantification of denitrification and N exports to oceans 595 result from the combined influence of the input data from ORCHIDEE and from 596 the process representation implemented in LSM Nlateral Off. In the following, 597 we investigate spatial, seasonal and decadal trends resulting from the offline 598 coupling of these three models. 599

600 3.2.1. Trends in global N flows

587

Averaged over the 1995-2014 period, the annual TN input from soils to 601 rivers, TN exports to oceans and denitrification in transit amount to 64.4 Tg N 602 yr⁻¹, 40.0 Tg N yr⁻¹, and 24.4 Tg N yr⁻¹, respectively. These three N fluxes show 603 increasing trends from 1901 to 2014. The global annual TN input to rivers 604 increased by 72.4 %, from 37.4 Tg N yr⁻¹ during 1901-1920 to 64.4 Tg N yr⁻¹ 605 during 1995-2014 (Fig. 6 a). The global annual TN export to oceans increased 606 by 45.6 % from 27.4 Tg N yr⁻¹ to 40.0 Tg N yr⁻¹. Most of this increase is 607 attributed to DIN, which doubled over the simulation period, rising from 10.0 608 Tg N yr⁻¹ to 19.9 Tg N yr⁻¹, while, in absolute terms, DON exports show a much 609 smaller increase but still substantial relative increase of 50.6 % (Fig. 6b). In 610 contrast, PON exports to oceans show a slightly decreasing trend. This decrease 611 is mainly attributed to global greening, which enhances vegetation cover 612 (Cortés et al., 2021; Wang et al., 2022) and reduces soil erosion, resulting in 613 lower PON inputs from the land and, thus, PON exports to oceans. The increase 614 in global denitrification mostly follows the rise in DIN inputs, with a relative 615 increase of 146.6 %, from 9.9 Tg N yr⁻¹ during 1901-1920 to 24.4 Tg N yr⁻¹ 616 during 1995-2014 (Fig. 6a). 617

The global TN input into rivers, TN export to oceans and denitrification in rivers all show a slight peak between 1926 and 1931 due to the relatively higher surface runoff during this period (Fig. S9). This higher runoff results mostly from meteorological forcings, as the global total amount of heavy rainfall (>25 mm d⁻¹) was higher during this period (Fig. S9). Note that Probst and Tardy (1989) provide empirical evidence for elevated global runoff during this period and we thus consider this peak as realistic.



Figure 6. Trends in global N flows from 1901 to 2014: (a) yearly mean TN
inputs into rivers, TN exports to oceans and denitrification rates; (b) yearly
mean DIN, DON and PON exports to oceans. TN: total nitrogen; DIN:
dissolved inorganic nitrogen; DON: dissolved organic nitrogen; PON:
particulate organic nitrogen.

631 **3.2.2.** Spatial patterns in N flows and concentrations

625

In this section, we examine the spatial distribution of contemporary N flows, and their changes compared to the early 20th century. Given the rapid increase in N flows since 1960 and the interannual variability induced by climate, we use the 1995-2014 average to represent contemporary N flows and the 1901-1920 average to represent early 20th-century conditions.

Annual mean TN input into rivers during 1995-2014 shows large spatial 637 heterogeneity, with higher values mainly located in eastern North America, 638 South America, Western Europe, tropical Africa, South Asia, Southeast Asia 639 and Southeast China (Fig. 7a). When compared with 1901-1920, TN inflow into 640 rivers increased in most areas (about 62%), with the highest increase (exceeding 641 300%) appeared in China, United States and Canada, Germany, France and 642 Spain (Fig. 8a). The annual mean contemporary denitrification rates (1995-643 2014) also exhibit large spatial heterogeneity (Fig. 7b) with high denitrification 644 rates in large tropical and subtropical rivers, such as the Amazon, Nile and 645 Congo rivers. Over the entire simulation period, the grid cells with the highest 646 relative denitrification increases are mostly located in the subtropical and north 647 temperate zone (Fig. 8b). 648

The TN export to oceans during 1995-2014 also varies substantially 649 across regions (Fig. 7c). The riverine TN exports are relatively low for the 650 Arctic Ocean, the western and southern coasts of Australia, and the coastal zone 651 adjacent to desert areas in South America (e.g., the Atacama Desert and the 652 Patagonian Desert), Africa (the Sahara Desert and the Namib Desert), and Asia 653 (e.g., the Arabian Desert, the Thar Desert in India, the deserts of eastern Iran, 654 and the Syrian Desert) (Fig. 7c). On the contrary, the Amazon region in South 655 America, the African rainforest region, Western Europe, South Asia, and 656 southeast China are prominent hotspots of riverine TN exports (Fig. 7c). 657 Unsurprisingly, TN exports to oceans have increased in approximately half of 658 the coastal areas since the early 20th century (Fig. 8c). In several regions, such 659 as the southeastern coastal areas of China and the eastern coast of the United 660 States, TN exports to oceans have even increased by more than 100% from 661 1901-1920 to 1995-2014 (Fig. 8c). 662

The annual mean contemporary concentration of TN at river mouths also 663 exhibits significant spatial heterogeneity (Fig. 7d), which differs from that of 664 TN export to oceans (Fig. 7c). For instance, the Amazon region is one of the 665 hotspots for TN exports, but its TN concentrations are low (<1 mg L⁻¹), because 666 the water discharge and denitrification rates are both high (Figs. 7b, S10 a). The 667 highest TN concentrations (>5 mg L^{-1}) are found in areas with intense human 668 activity, for example the San Francisco area, Chile, Spain, Egypt (Nile River 669 estuary) and the southeastern coastal areas of China (Bu et al., 2019; Hou et al., 670 2022; Yang et al., 2023). 671

The spatial distribution of changes in TN concentrations from 1901-1920 to 1995-2014 differs from that of TN exports (Fig. 8c, d). For example, along the western coast of Chile, and the western coast of Guinea, Sierra Leone, and Libya, TN exports to oceans decreased by more than 10%, while TN concentrations increased by more than 10% (Fig. 8c, d). This phenomenon is

- 677 due to negative trends in water discharge from the corresponding watersheds
- (Fig. S10 b). In most regions, the ratio of changes in TN fluxes to changes in
- TN concentrations ranges between 0 and 10, indicating that TN flux changes are
- 680 driven by the combined effects of changes in water discharge and TN
- 681 concentrations (TN inputs into rivers) (Fig. 9).



682

Figure 7. Spatial patterns of annual mean N fluxes and concentrations during

684 1995-2014: (a) TN inputs into rivers; (b) denitrification rates in rivers; (c) TN

- exports to oceans; (d) TN concentrations at rivers mouths. To display the spatial patterns of denitrification in rivers better, we excluded data with denitrification
- rates less than 0.001 GN yr^{-1} per grid.



688

Figure 8. Spatial patterns of changes from 1901-1920 to 1995-2014 of: (a) TN

- 690 inputs into rivers; (b) denitrification; (c) TN exports to oceans; (d) TN
- 691 concentrations.



692

Figure 9. Ratio of changes in TN exports to changes in TN concentrations from1901-1920 to 1995-2014.

695 **3.2.3. Seasonal variability in N flows and concentrations**

The seasonality of TN inputs into rivers during the period 1995-2014 is most pronounced in the central United States, Europe, South Asia, Southeast Asia and southeast China (Fig. 10a). The frequency distribution of the seasonal amplitude in inputs (Fig.10a) is broadly similar to that of the mean annual inputs (Fig 7a), suggesting a seasonal variability of similar magnitude than the broad, global scale spatial variability. A similar pattern is observed for
denitrification rates, with seasonal and spatial variations of comparable
magnitudes (Figs. 7b, 10b).

The seasonal amplitudes of TN exports to oceans during the period 1995-704 2014 shows highest values (> 10 Gg N yr⁻¹) along the coasts of South Asia, 705 southeast China and Mexico, and to a lesser extent (1-10 Gg N yr⁻¹) along the 706 coastline of the Amazon region, the rainforest regions of Africa, and Western 707 Europe (Fig. 10c). As expected, a significant portion of this seasonal variability 708 is due to river discharge (Fig. S11 a). Our results indicate that the spatial pattern 709 of seasonal amplitudes in TN concentrations at river mouths differs from that of 710 TN exports (Figs. 10, S12, S13). This result is important because the ocean 711 biogeochemical modelling community typically uses annual mean TN fluxes 712 derived from Global News to force their simulations (e.g., Lee et al., 2016; 713 Stock et al., 2020; Tjiputra et al., 2020; Lacroix et al., 2021), and downscales 714 these inputs to monthly values under the assumption that the seasonal variability 715 of the flux is entirely driven by river discharge. Our simulations thus stress the 716 need for models that explicitly resolve the seasonal variability of fluxes and 717 concentrations. 718

We also normalized the seasonality by the mean value of N flux or concentrations. For TN inputs into rivers, denitrification and TN exports, the normalized seasonal maps all show higher values in the middle and high latitudes of the Northern Hemisphere and lower values in the low latitudes and the Southern Hemisphere (Fig. S12). Moreover, the regional-scale heterogeneity in the normalized seasonality of TN concentration is little weaker than that of the TN flux (Figs. S12 c&d).



726

Figure 10. Spatial distribution of the seasonal amplitude (period 1995-2014) in:
(a) TN inputs into rivers; (b) rates of denitrification; (c) TN exports to oceans;
(d) TN concentrations at rivers mouths.

730 **3.3.** Comparison with other models

We compared the trends of global DIN input into rivers simulated by 731 ORCHIDEE-CNP with those generated by the recently published IMAGE-732 GNM model (Vilmin et al., 2018). Overall, both models capture a similar 733 increasing trend of global DIN delivery from land to rivers from 1901 to 2001 734 (Fig. 11a). During 1961-2000, the global-scale interannual variability of DIN 735 simulated by ORCHIDEE-CNP is comparatively stronger than that simulated 736 by IMAGE-GNM (Fig. 11a). This discrepancy may be partially explained by 737 differences in the temporal resolution of the two models (daily for ORCHIDEE-738 CNP, yearly for IMAGE-GNM) and the associated climate forcings. In other 739 words, ORCHIDEE-CNP calculates the annual means from daily fluxes, 740 whereas IMAGE-GNM does not resolve the intra-annual variability. In contrast, 741 the organic nitrogen (ON = PON + DON) fluxes simulated by ORCHIDEE-742 Clateral and derived from IMAGE-GNM differ significantly. The ON inflow 743 simulated by IMAGE-GNM shows a substantial increase from 24.9 Tg N yr⁻¹ 744 during 1901-1910 to 37.9 Tg N yr⁻¹ during 1991-2000, while ON simulated by 745

ORCHIDEE-Clateral exhibits a weaker increasing trend over the same period, 746 from 26.5 Tg N yr⁻¹ to 32.4 Tg N yr⁻¹. The weaker trend in ORCHIDEE-747 Clateral can primarily be explained by the increasing DON inflow being offset 748 by a decreasing PON inflow (Fig. 11c). The fundamental reason for the 749 discrepancy among the two models stems from their distinct structures and 750 algorithms. In ORCHIDEE-Clateral, the ON flows into rivers are calculated 751 separately for the dissolved and particulate compounds using a process-based 752 representation of the soil C stock dynamics and C:N ratios, as well as the rates 753 of runoff and drainage. The approach is different in IMAGE-GNM which 754 calculates the bulk ON flows (DON+PON) based on empirical formulas 755 (Vilmin et al., 2018). Specifically, IMAGE-GNM calculates ON delivery from 756 land to rivers with drainage based on the TN delivery rate with drainage, 757 assuming that 50% of TN flux is in the form of ON. For ON flows into rivers 758 with runoff, IMAGE-GNM distinguishes two runoff mobilisation pathways, i.e. 759 losses from recent nutrient applications in forms of fertiliser and manure, and a 760 761 memory effect related to long-term historical changes in soil nutrient inventories. These two pathways both are simulated based on empirical 762 formulas (Vilmin et al., 2018). In ORCHIDEE-Clateral, default C:N ratio in 763 different SOM pools were used to calculate the PON erosional fluxes from soils 764 using a process-based approach, and a constant C:N ratio (averaged values from 765 references) was applied to simulate DON flows out of soils. The assumption of 766 constant C:N ratio for dissolved matter in soil may contribute to the weaker 767 trend in ON delivery to rivers simulated by LSM Nlateral Off, since some 768 studies have revealed that DOC:DON ratios vary with time and land cover (Li 769 et al., 2019; Yates et al., 2019). 770





Figure 11. Global terrestrial N flows into rivers from 1901 to 2001 simulated by
ORCHIDEE model versions and IMAGE-GNM (Vilmin et al., 2018): (a) DIN;
(b) ON (DON+PON); (c) DON and PON derived from ORCHIDEE-Clateral.

The simulated lateral N flows from land to rivers and N exports to oceans 775 in this study are now compared with those simulated by other models across 776 different time horizons, noting that each model covers different time periods 777 (Fig. 12). Focusing first on the global N flows from land to rivers, we find that 778 for different time horizons, the N inputs used as forcings for LSM Nlateral Off 779 (i.e., simulated by ORCHIDEE-Clateral and ORCHIDEE-CNP) are very close 780 with those estimated by IMAGE-GNM (Vilmin et al., 2018) and FrAMES-N 781 (Wollheim et al., 2008), with differences between our simulations and other 782 models never exceeding 7% across different time horizons. Although the 783 fraction of DIN in TN over 1901-1910 simulated by LSM Nlateral Off (27%) 784 is slightly lower than that of IMAGE-GNM (29%), the DIN fractions simulated 785 by these two models both show obvious increasing trends with time, 786 LSM Nlateral Off and IMAGE-GNM reporting DIN fractions for the 1991-787

2000 period reaching 48% and 43%, respectively (Fig. 12a). These results are
consistent with a comprehensive cross-biome assessment of N composition in
rivers that also revealed a shift in the dissolved N from highly heterogeneous to
primarily inorganic N in response to human disturbances (Wymore et al., 2021).
This change in the composition of TN inputs from land to rivers is primarily
caused by the excess inorganic N released from agricultural (due to the
utilisation of fertilisers) and urban (due to the release of sewage) areas.

The global N export from rivers to oceans simulated by 795 LSM Nlateral Off is also comparable to estimates from other models. During 796 1901-1910, the global riverine N export to oceans is 29.0 Tg N yr⁻¹, a value that 797 falls within the range simulated by IMAGE-GNM (19.0 Tg N yr-1, Vilmin et 798 al., 2018) and DLEM (29.4 Tg N yr⁻¹, Tian, pers. com.) (Fig. 12b). For the most 799 recent period (2000s), the simulated riverine N export to oceans is converging, 800 with differences less than 10 % compared to other models such as 801 GlobaNEWS2 (Mayorga et al., 2010), IMAGE-GNM, and DLEM (Fig. 12b). 802 Although the global riverine TN export to oceans simulated by 803 LSM Nlateral Off is close to that simulated by GlobalNEWS2 (1970-2010), 804 the TN export reported here contains a slightly larger fraction of DIN and a 805 slightly lower fraction of PON compared to GlobalNEWS2 (Fig. 12b). 806

The TN export to oceans simulated by LSM Nlateral Off and 807 GlobalNEWS2 are also comparable at continental scale (Fig. 13a), with largest 808 TN exports from Asia, and lowest exports from Australia. However, the 809 simulated proportions of N species in the overall TN export show distinct 810 behaviours between these two models. For example, compared to 811 GlobalNEWS2, the DIN proportion in TN exports simulated by 812 LSM Nlateral Off is larger in Asia, Africa and South America but smaller in 813 Europe (Fig. 13a). 814

The magnitude of TN exports simulated by LSM Nlateral Off and 815 GlobalNEWS2 continues to diverge at basin scale (Fig. 13b). In 8 of the top 20 816 basins by area, the difference between the two models is less than 50%, such as 817 in the Congo, Mississippi, Ob, Parana, Yenisei, Changjiang, Mackenzie and 818 Nelson basins. Larger discrepancies can however be observed in several large 819 river systems. For instance, in the Amazon basin, the TN export simulated by 820 GlobaNEWS2 is about 2.5 times larger than that simulated by 821 LSM Nlateral Off. The evaluation of LSM Nlateral Off simulation results 822 against measurements of TN flow rates in the Amazon River indicates that 823 LSM Nlateral Off underestimates the TN flow in this basin (Fig. 4). At 824 Manacapuru and Óbidos, two observation sites on the main channel of the 825 Amazon River, the observed TN flow is 1.90 Tg N yr⁻¹ and 2.82 Tg N yr⁻¹, but 826 the simulated values are 0.92 Tg N yr⁻¹ and 1.57 Tg N yr⁻¹, respectively. To 827 evaluate whether this underestimation is caused by less TN inflow into rivers, 828 we set the N transformation processes (decomposition of DON and PON, and 829 denitrification) in rivers to zero, and found that the TN flows are 1.56 Tg N yr⁻¹ 830 at Manacapuru and 2.35 Tg N yr⁻¹ at Óbidos. Therefore, even with no N 831 removal, LSM Nlateral Off still underestimates the observed TN flows at these 832 two sites, suggesting that N delivery from terrestrial ecosystems to rivers (as 833 simulated by ORCHIDEE) is too low in the Amazon basin. In the Nile basin, 834 the TN export simulated by LSM Nlateral Off is thirty times larger than that 835 simulated by GlobalNEWS2. Observed annual exports of DIN and DON 836 amount to 0.079 Tg N yr⁻¹ and 0.038 Tg N yr⁻¹, respectively (Badr, 2016). These 837 observed values are of the same magnitude as those simulated by 838 LSM Nlateral Off, 0.113 Tg N yr⁻¹ for DIN and 0.048Tg N yr⁻¹ for DON. This 839 finding suggests that LSM Nlateral Off better captures the observed N export 840 for this specific basin than GlobalNEWS2. 841

It should be noted that the GlobalNEWS2 and IMAGE-GNM both have 842 an IMAGE part to simulate N inputs into inland rivers, but were developed 843 using different hydrological models and different methods to calculate N 844 transport and retention along the global river network. The hydrological model 845 embedded in GlobalNEWS2 is the Water Balance Model (WBM_{plus}) (Fekete et 846 al., 2010), and the NEWS models were then developed to calculate nutrient 847 retention in streams and reservoirs (Seitzinger et al., 2005, 2010; Mayorga et al., 848 2010). The hydrological model used in IMAGE-GNM is the PCRaster Global 849 Water Balance (PCR-GLOBWB) (Van Beek et al., 2011), and IMAGE-GNM 850 then applied the nutrient spiralling approach (Newbold et al., 1981) to describe 851 in-stream retention of both N and P with a yearly time step (following 852 Wollheim et al., 2008). 853

In summary, the global total N input to rivers and N export to oceans 854 simulated by the different models are comparable, but the spatial distribution of 855 N export to oceans at finer spatial scales shows increasing discrepancies, as 856 does the chemical speciation. This is mainly due to differences in model 857 structures, spatial and temporal resolutions and forcing data. Although our 858 model has been evaluated against the largest dataset of river discharge and N 859 concentrations from the recently assembled global GRDC and GRQA database, 860 significant cross-model discrepancies emerge as the analysis is refined to 861 regional patterns and individual river basins. This highlights the necessity for 862 improvements in model structure and quality of both forcing data and 863 evaluation data, as well as the implementation of ensemble-mean assessments, 864 akin to the recent approach applied to constrain carbon exports to the oceans 865 (Liu et al., 2024). 866



867

Figure 12. Comparison of global TN fluxes estimated by different models: (a)

869 global TN inputs to rivers; (b) global TN exports to oceans. IMAGE-GNM:

870 Integrated Model to Assess the Global Environment-Global Nutrient Model

871 (Vilmin et al., 2018); FrAMES-N: Framework for Aquatic Modeling in the

872 Earth System (Wollheim et al., 2008); MBM: Mass Balance Model (Green et

al., 2004); GlobalNEWS2: Global Nutrient Export from Watersheds 2

874 (Mayorga et al., 2010); DLEM, Dynamic Land Ecosystem Model, unpublished

875 (Tian, pers. com.).



876

Figure 13. Comparison of present-day (2001-2010) TN export to oceans
simulated by LSM_Nlateral_Off and GlobalNEWS2 (Mayorga et al., 2010) at:
(a) continental scale; (b) basin scale.

880 **3.4.** Model limitations and priorities for future research

LSM Nlateral Off currently relies on a simplified representation of the N 881 processes in benthic sediments and water, without explicit simulation of the 882 hyporheic exchange between sediments and water. The importance of these 883 processes is estimated using a scaling factor based on water depth, which itself 884 relies on a coarse approximation of the stream channel geometry based on 885 empirical formulas (Raymond et al., 2012). Global-scale databases on the 886 geomorphic properties of river channels, including river depth and width, are 887 available (Andreadis et al., 2013) and could be used in the future to further 888 refine the representation of N processes in river channels, including the 889

hyporheic exchange between sediments and water. The residence time method 890 was used to estimate water and N transport within river networks. This method 891 is simple and has been widely used in large scale simulations of fluvial water, 892 carbon and N transports (Beusen et al., 2015; Jepsen et al., 2019; Zhang et al., 893 2022). However, it may not fully capture the seasonality of water and N flows 894 accurately in some regions (Fig. 5 a2 & b2). To improve the accuracy of 895 simulating fluvial water and N transport, the residence time method currently 896 used in LSM Nlateral Off could be replaced with hydrological kinetic 897 equations in future versions of the model. 898

The current version of LSM_Nlateral_Off also has several limitations in terms of biogeochemistry. One limitation is the use of a constant C:N ratio to simulate DON fluxes from soils to rivers. Research has shown that the C:N ratio varies over time and across different land cover types (Li et al., 2019; Yates et al., 2019). The use of a constant C:N ratio may thus reduce the accuracy and informativeness of the estimated DON flux. Addressing this limitation is an urgent priority for future research.

At present, few studies have accounted for the effects of PON deposition 906 and resuspension on lateral N transfer in rivers because of the challenge of 907 representing these processes at the global scale. Moreover, PON deposition is 908 mainly controlled by the rate of sediment deposition, a process which is not 909 represented in the current model version. Therefore, PON deposition has not 910 been simulated either. Recent results from ORCHIDEE-Clateral suggest that 911 about 22% of POC entering the global river network is deposited with 912 sediments before reaching the coast (Zhang et al., under review). Assuming a 913 similar fraction of deposited PON, global PON export to oceans simulated by 914 LSM Nlateral Off could be approximately 20% lower (about 2 Tg N yr⁻¹) than 915 estimated here. 916

The role of autotrophic production is another process currently omitted. 917 Autotrophs (aquatic macrophytes, algae, cyanobacteria, bryophytes, some 918 protists, and bacteria) in freshwater systems take up DIN from the water column 919 (King et al., 2014) and may play a significant role in N cycling within rivers 920 (Wachholz et al., 2024). In future model developments, the role of autotrophic 921 production on N retention should thus be considered, although the large 922 dominance of the heterotrophic metabolism on a global scale suggests that in-923 situ aquatic production is a second-order control on N cycling (Battin et al., 924 2023). The transformation of PON to DON is also not included in the current 925 version of LSM Nlateral Off. A previous study suggests that the instream 926 transformation of POC to DOC is limited (about 0.3%) (Zhang et al., 2022). It 927 can thus be assumed that the fraction of PON transformed to DON is also rather 928 negligible. Nevertheless, we plan to incorporate this transformation process into 929 our model in the next phase of our research. 930

In the present version of LSM Nlateral Off, river-floodplain dynamics 931 and channel erosion are currently not represented, because of the incomplete 932 understanding of how these processes affect lateral N transfer and the lack of 933 reliable parameters from field studies to quantify their impacts at global scale. 934 Floodplain inundation not only facilitates N inputs into river, but also 935 significantly influences N retention efficiency in rivers (Martí et al., 1997; 936 Hanrahan et al., 2018), and N cycling (e.g., nitrification and denitrification) in 937 flooded soils (Sánchez-Rodríguez et al., 2019; Hu et al., 2020). For instance, in 938 the Jiulong River watershed in southeast China, flood events exported 47% and 939 42% of the annual land-derived ammonium (NH_4^+) and NO_3^- , respectively, 940 although they only occurred 24% of the time (Gao et al., 2018). This highlights 941 the critical role of flood events in N transport and cycling, emphasizing the need 942 to incorporate floodplain processes in future model development. 943

LSM Nlateral Off includes the major sources of riverine N with runoff 944 and drainage from natural, agricultural and urban ecosystems (Fig. 1). Yet, 945 several sources are still missing, for example atmospheric N deposition directly 946 onto rivers and N release from aquaculture (Filoso et al., 2003; Bouwman et al., 947 2013; Beusen et al., 2016a; Gao et al., 2020), suggesting that the N exports to 948 oceans simulated by LSM Nlateral Off might be conservative. On the other 949 hand, N retention and recycling in lakes and artificial reservoirs are currently 950 missing, which have the potential to decrease lateral N flows because they offer 951 ideal conditions for N burial in sediment or permanent loss via denitrification 952 (Saunders & Kalff, 2001; Harrison et al., 2009; Akbarzadeh et al., 2019). The 953 absence of these processes in the current model may lead to an overestimation 954 of N exports to oceans. 955

The forcing data used by the LSM Nlateral Off (Table 1) introduces 956 additional uncertainties into the simulation results. The routing scheme of water 957 and N is driven by a map of streamflow direction at 0.5° spatial resolution 958 (Vörösmarty et al., 2000, https://doi.org/10.1016/S0022-1694(00)00282-1). 959 There are obvious discrepancies between this routing scheme and the real river 960 network (Zhang et al., 2022). This deviation of flow direction induces 961 uncertainties in the simulated riverine water discharge and N flow because the 962 flow direction directly determines the area of each catchment and the routing of 963 the river. 964

Finally, although LSM_Nlateral_Off effectively reproduces the magnitude and seasonal variations of water and N transfer from land to rivers and oceans (Figs. 4 & 5), spatial and temporal biases in observational data also affect the evaluation of model performance. Most observations of riverine N are distributed in North America, South America and Europe, highlighting the crucial need to collect more measurements in other regions of the world, especially in Africa. In addition, despite the strong correlation between TN and

NO₃⁻ concentrations, the application of an empirical equation (Eq. 15) to estimate TN from NO₃⁻ introduces additional uncertainties in the observational dataset (Pisani et al., 2017; Niu et al., 2022).

975 4. Conclusions

We developed a global N lateral transfer model from land to oceans 976 through the river network, incorporating the decomposition of DON and PON 977 and denitrification of DIN during fluvial transport. Evaluations using 978 observational data from GRDC and GRQA indicate that LSM Nlateral Off 979 reproduces observed rates and seasonal variations of water discharge and N 980 flow well. The global simulation of LSM_Nlateral Off shows that global TN 981 inputs into rivers, TN exports to oceans and riverine denitrification rates 982 increased significantly over the last century. In particular, the TN exports to 983 oceans increased from 27.5 Tg N yr⁻¹ during 1901-1920 to 40.0 Tg N yr⁻¹ during 984 1995-2014, with DIN contributing 80% to the TN increase. Our results reveal 985 significant spatial heterogeneity in the global distribution of N inputs, 986 transformation and exports to oceans, with East Asia and Southeast Asia 987 identified as hotspots of N lateral transfers and their increase. The seasonal 988 amplitude of TN export is of similar magnitude to the large-scale spatial 989 heterogeneity in TN fluxes. Although the global and continental-scale TN 990 exports to oceans simulated by LSM Nlateral Off are similar to that of another 991 widely used model (GlobalNEWS2), their spatial distributions at the basin scale 992 reveal significant discrepancies. One key strength of LSM Nlateral Off is its 993 ability to resolve N processes at the daily timescale, using a framework fully 994 compatible with land surface model (LSM) outputs. This compatibility enables 995 the model to account for the effects of climate change, atmospheric composition 996 changes, land-use change, and agricultural practices (e.g., manure and fertiliser 997 use) in a fully consistent way. 998

LSM Nlateral Off has however its own limitations and we plan to further 999 enhance its capabilities with additional processes (e.g. autotrophy, variable C:N 1000 ratios, erosion-deposition on riverbed), additional sources (e.g. aquaculture, 1001 direct N deposition) and interconnections with other (semi)-aquatic and benthic 1002 systems (hyporheic zone, lakes, reservoirs, floodplains). Furthermore, additional 1003 observational data will be collected to further calibrate and evaluate 1004 LSM Nlateral Off. Last but not least, LSM Nlateral Off is currently being 1005 dynamically embedded into ORCHIDEE (Vuichard et al., 2019), the land 1006 surface scheme of the IPSL Earth System Model. This coupling opens new 1007 avenues towards fully coupled simulations of the land-ocean-atmosphere N 1008 cycle. Additionally, the current offline version of our model could also be easily 1009 coupled to other LSMs representing N cycling in terrestrial ecosystems, 1010 enabling broader applications and cross-model comparisons. 1011 1012

1013 Appendices

1014	Table A1. Abbreviation used in the text.	
1014	Table A1. Abbreviation used in the text.	

Abbreviation	Meaning	unit
F_{DR_DIN}	leaching rates of DIN with drainage	g N d ⁻¹
F_{DR_DON}	leaching rates of DON with drainage	g N d ⁻¹
F_{RO_DIN}	leaching rates of DIN with runoff	g N d ⁻¹
F_{RO_DON}	leaching rates of DON with runoff	g N d ⁻¹
F_{RO_PON}	erosion rates of PON with runoff	g N d ⁻¹
Fsewage_DIN	DIN inflow rates from sewage	g N d ⁻¹
F_{sewage_DON}	DON inflow rates from sewage	g N d ⁻¹
$F_{fastout_H2O}$	outflow rates of water from fast reservoirs to stream reservoirs	$m^3 d^{-1}$
$F_{fastout_DIN}$	outflow rates of DIN from fast reservoirs to stream reservoirs	g N d ⁻¹
$F_{fastout_DON}$	outflow rates of DON from fast reservoirs to stream reservoirs	g N d ⁻¹
$F_{fastout_PON}$	outflow rates of PON from fast reservoirs to stream reservoirs	g N d ⁻¹
$F_{slowout_H2O}$	outflow rates of water from slow reservoirs to stream reservoirs	$m^3 d^{-1}$
$F_{slowout_DIN}$	outflow rates of DIN from slow reservoirs to stream reservoirs	g N d ⁻¹
$F_{slowout_DON}$	outflow rates of DON from slow reservoirs to stream reservoirs	g N d ⁻¹
$F_{streamout_H2O}$	outflow rates of H ₂ O to downstream reservoirs	$m^3 d^{-1}$
$F_{streamout_DIN}$	outflow rates of DIN to downstream reservoirs	g N d ⁻¹
$F_{streamout_DON}$	outflow rates of DON to downstream reservoirs	g N d ⁻¹
$F_{streamout_PON}$	outflow rates of PON to downstream reservoirs	g N d ⁻¹
R_{fast_DIN}	denitrification rates in fast reservoirs	g N d ⁻¹
R_{fast_DON}	decomposition rates of DON in fast reservoirs	g N d ⁻¹
R_{fast_PON}	decomposition rates of PON in fast reservoirs	g N d ⁻¹
R_{slow_DIN}	denitrification rates in slow reservoirs	g N d ⁻¹
R_{slow_DON}	decomposition rates of DON in slow reservoirs	g N d ⁻¹
R_{stream_DIN}	denitrification rates in stream reservoirs	g N d ⁻¹
R_{stream_DON}	decomposition rates of DON in stream reservoirs	g N d ⁻¹
R_{stream_PON}	decomposition rates of PON in stream reservoirs	g N d ⁻¹
S_{fast_H2O}	water stock in fast reservoir	m ³
S_{fast_DIN}	DIN stock in fast reservoir	g N
S_{fast_DON}	DON stock in fast reservoir	g N
S_{fast_PON}	PON stock in fast reservoir	g N
S_{slow_H2O}	water stock in slow reservoir	m ³
S_{slow_DIN}	DIN stock in slow reservoir	g N
S_{slow_DON}	DON stock in slow reservoir	g N
S_{stream_H2O}	water stock in stream reservoir	m ³
S_{stream_DIN}	DIN stock in stream reservoir	g N
S_{stream_DON}	DON stock in stream reservoir	g N
S_{stream_PON}	PON stock in stream reservoir	g N
TW	water temperature	°C
F_{T_DIN}	dependency of denitrification on temperature	unitless

depth	depth of rivers	m
Q	water discharge	km ³ yr ⁻¹

015	Table A2. Values of the key parameters used in LSM_Nlateral_Off to simulate
016	the lateral transfer of N.

Parameter	Value	Description	Source
$ au_{\mathrm{fast}}$	3.0 days	A factor which translates the topographic index into the water residence time of the "fast" reservoir (Eq. 1)	Ngo-Duc et al., 2006
$ au_{slow}$	25.0 days	A factor which translates the topographic index into the water residence time of the "slow" reservoir (Eq. 1)	Ngo-Duc et al., 2006
$ au_{stream}$	0.24 days	A factor which translates the topographic index into the water residence time of the "stream" reservoir (Eq. 1)	Ngo-Duc et al., 2006
K _{PON}	0.028 d ⁻¹	the average PON decomposition rate at 20°C in water (Eq. 11)	Islam et al., 2012
K _{DON}	0.07 d ⁻¹	the average DON decomposition rate at 20°C in water (Eq. 11)	Xia et al., 2013
K _{DIN}	0.15 d ⁻¹	the average denitrification rate in water at 25°C (Eq. 12)	Alexander et al., 2000
Q ₁₀	2.0	the temperature sensitivity of PON and DON decomposition rates (Eqs. 11)	Liu et al., 2021
T _{refl}	20 °C	the reference temperature for PON and DON decomposition (Eqs. 11)	Zang et al., 2020
T _{ref2}	25 °C	the reference temperature for denitrification (Eq. 13)	Ma et al., 2022

Code and data availability. The source code of the LSM_Nlateral_Off model
is available online(<u>https://zenodo.org/records/13309551</u>). All forcing and
validation data used in this study are publicly available online. The specific
sources for these data can be found in Table 1.

1022

Author contributions. MM, HZ, RL, PR and PC designed the study. MM and HZ conducted the model development and simulation experiments. PR, RL and PC provided critical contributions to the model development and the design of simulation experiments. MM conducted the model calibration, validation, and data analysis. HZ, PR, RL and PC provided support on collecting forcing and validation data. MM wrote the paper. All authors contributed to interpretation and discussion of results and improved the paper.

1030 Competing interests. The contact author has declared that none of the authors1031 has any competing interests.

1032

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