Wu, S., Soulsby, C., Zheng, Y., Musolff, A., Tetzlaff, D. (2026):
Divergent evolution of nitrogen cycling along gradients of landscape water velocities
Science 392 (6803), 1188 - 1193
10.1126/science.aed0399

Divergent Evolution of Nitrogen Cycling Along Gradients of Landscape Water Velocities

Songjun Wu[1*], Chris Soulsby[2], Yi Zheng[3], Andreas Musolff[4], Doerthe Tetzlaff[1,5]

1 Department of Ecohydrology and Biogeochemistry, Leibniz Institute of Freshwater Ecology and Inland Fisheries, Berlin, Germany

2 Northern Rivers Institute, School of Geosciences, University of Aberdeen, UK

3 School of Environmental Science and Engineering, Southern University of Science and Technology, Shenzhen, China

4 Department of Hydrogeology, Helmholtz Centre for Environmental Research, Leipzig, 04318, Germany

5 Geography Institute and IRI THESys, Humboldt University of Berlin, Berlin, Germany

* Corresponding author: Songjun Wu (songjun.wu@igb-berlin.de)

Abstract

Increasing fertilization has pushed nitrogen cycle beyond planetary boundaries, yet its fate remains uncertain due to long-standing neglect of landscape water velocities in nitrogen models. Leveraging isotope-aided modelling across 3,821 European catchments, we demonstrate that evolution of nitrogen cycling is strongly linked to shifts in landscape water velocities since 1980. We propose the concept of "wetness boundaries", where hydrological transitions beyond boundaries amplify nitrogen accumulation and leaching, while conditions remaining within boundaries mitigate these processes. Applying this framework, we project reduced nitrogen leaching across 76% of Europe under mild hydrological shifts by 2100 but increasing nitrogen accumulation under pronounced deceleration of water cycling in Eastern and Southern Europe. These findings underscore emerging water-quality risks under climate change and the need to mitigate extreme hydrological shifts.

Planetary boundaries define a safe operating space for humanity, yet the boundary for nitrogen has been exceeded due to the doubling of anthropogenic inputs since pre-industrial era (*1*). This transgression poses substantial risks to freshwater eutrophication, food production, and ecosystem sustainability (*2*). Accordingly, nitrogen cycling has been extensively studied across terrestrial and aquatic ecosystems from catchment to global scales (*3*–*5*).

Despite decades of research, however, the fate of nitrogen at the landscape scale remains poorly understood, as it emerges from an intricate interplay among spatiotemporally variable fertilizer/manure application, fossil fuel combustion, hydrological transport, and a cascade of biogeochemical reactions from nitrification to denitrification (*4*, *5*). Among these dimensions, the influence of hydrology is, perhaps, one of the most pivotal yet comparatively under-explored aspects. As the primary medium transporting soluble nitrogen, water governs its movement and shapes the kinetics of biogeochemical transformations along flow paths (*3*, *6*). However, hydrological processes are often under-represented with simplified assumption/conceptualization in water quality models, with many applications relying solely on in-stream observations (typically discharge) for process inference (*3*). This, unfortunately, captures only half the story of how water and solutes move through landscapes (*7*), as a good model fit of discharge reflects only the celerity of the system's response to precipitation inputs (or the speed with which perturbations are transmitted), while the velocity of internal water fluxes remains unquantified. A key paradox is that stream discharge may respond within minutes or hours to rainfall or snowmelt, yet with flows comprised of water that can be years or decades old (*8*). While the lack of understanding of velocities may suffice for hydrological studies that focus more on the integrated response at a system's outlet (e.g., flood generation), this information is indispensable for understanding nitrogen cycling where fate of solutes emerges from pathways with vastly different velocities (*6*). With climate projections pointing to potential intensification of water cycling and increasing drought risks (*9*), a thorough knowledge of the role of landscape water velocities in nitrogen cycling, particularly how acceleration and deceleration of water cycling reshape nitrogen transport and transformation, remains elusive but needed.

Here, we employ stable water isotopes (^2H and ^{18}O) to fill this knowledge gap. As conservative tracers, these isotopes have been increasingly used to identify the water velocities and ages due to their independence of biogeochemical reactions (*10*, *11*). However, the power of isotopes has rarely been leveraged for understanding water quality, with existing pioneering studies limited to specific scales (plot to catchment scales), coarse resolution (lumped or semi-distributed) or simplified conceptualisations (*12–14*; details in text S5). Therefore, we present a continental-scale water velocity framework based on a fully distributed process-based ecohydrological model that simultaneously tracks water, isotopes, and nitrogen (EcoTWIN; text S1; (*15*)). EcoTWIN was applied in 3,821 catchments (50 to 802,032 km^2) across Europe at high spatial (5 km) and temporal resolution (daily timesteps since 1980) and validated against observed discharge, in-stream isotope compositions, and nitrate concentrations at 1,218 sites (text S2-4). Here, we used nitrate to reflect nitrogen cycling given its widespread dominance in surface waters and the availability of large-scale observations (uncertainties in text S10). With this fully distributed isotope-aided water quality modelling at the continental scale, we revisited water cycling across Europe through the lens of landscape water velocity and assessed how its acceleration and deceleration have reshaped nitrogen cycling since the 1980s and may evolve towards the end of the 21st century.

Slow Water Cycling Buffers Nitrogen Leaching

To uncouple the hydrological impacts from nitrogen cycling, we first quantified the water velocities using two indices - soil travel time (time for water traveling through ~2.5 m soils) and young water fraction (fraction of soil water younger than 90 days). Isotope-aided simulations revealed distinct regimes along geographic and climatic gradients in Europe, showing a gradual decline in young water along elevations (from high to low) and longitude (from west to east) (fig. 1A). The fastest water cycling was found at west-north coast of Europe (fig. 1C), where moist air masses are advected by the prevailing westerlies (*16*). Accordingly, the water cycling in these regions is generally more active with higher soil infiltration (1548 mm yr^{-1} averaged between 1982 to 2024) compared to the European average (685 mm yr^{-1}, fig. S5). The abundant precipitation led to rapid water velocities as evidenced by the significantly higher young water fraction (40.9%) and lower soil travel time (181 days) relative to the European median of 22.7% and 1063 days, respectively. Similar fast turnover rates are found in Alpine regions with abundant precipitation yet limited evapotranspiration (fig. 1C), where young water fraction reached up to 80.0% (though snow-dominated catchments are not included due to the dominant baseflow contribution during extended snow-dominated low-flow periods, e.g., in the Middle Alps) (*17*, *18*). In contrast, substantially slower water cycling was found in lowlands with soil travel times ranging from 81.1 to >3000 days with a median of 1182 days. This is attributed to the dual impacts of lower precipitation and higher evapotranspiration, returning a large proportion of precipitation to the atmosphere (accounting for 69% of infiltration; fig. S5). Such slow turnover rates are especially pronounced in regions dominated by continental or Mediterranean climate, where water might stay in the soil for years before leaching vertically to groundwater or laterally downslope (*7*, *19*).

The contrasts in hydrological regimes further propagate to nitrogen cycling, as rates of water fluxes not only govern nutrient transport, but also regulate biogeochemical reactions along flow paths through modulating moisture conditions (*20*). From a hydrological standpoint, nitrogen leaching can be viewed as the fraction of nitrogen leached from soil before being removed via biogeochemical processes (denitrification and plant uptake). Following the concept of soil Damköhler numbers (*14*), we expanded this dimensionless metric to reflect the balance between nitrogen retention and transport, defined as the ratio between transport timescale (average soil travel time) and reaction timescale (average time required to remove soil-water nitrogen via denitrification and plant uptake). Damköhler numbers <1 indicates the dominance of transport over removal processes and the potential of nitrogen leaching (*12*, *14*, *21*).

Generally, Damköhler numbers tend to be higher in regions with lower anthropogenic nitrogen inputs (fig. 1B). The median value in non-agricultural regions was 17.2, suggesting that most natural ecosystems effectively retain nitrogen at low levels. In contrast, croplands showed a median Damköhler numbers of 0.7, indicating insufficient nitrogen removal capacity relative to larger anthropogenic inputs and existing legacy. Meanwhile, the magnitudes of landscape water velocities played a vital role. This is evident from the comparison between the croplands in Eastern-Southern Europe and Central-Western Europe (fig. 1C): 59% and 40% of croplands exhibited Damköhler numbers > 1 in Eastern and Southern Europe, while the median Damköhler number dropped to 0.3 (minimal 0.05) in Central-Western Europe, implying reaction times are 3-20 times of travel times. This difference is mainly attributed to the slower turnover rates in the former regions. Longer soil travel times extended the timescales available for biogeochemical removal (denitrification and plant uptake), thus providing buffering against nitrogen leaching in Eastern and Southern Europe, while faster water velocities in Central-Western Europe increased the leaching risks. Such nitrogen stress has previously been attributed to a gradual build-up of legacy through historical fertilisation and the resulting intensification of nitrogen deposition (*22*). Here, using a water velocity framework, we demonstrate the role of water velocities in controlling the timescale for nitrogen removal and leaching potential (fig. 1D).

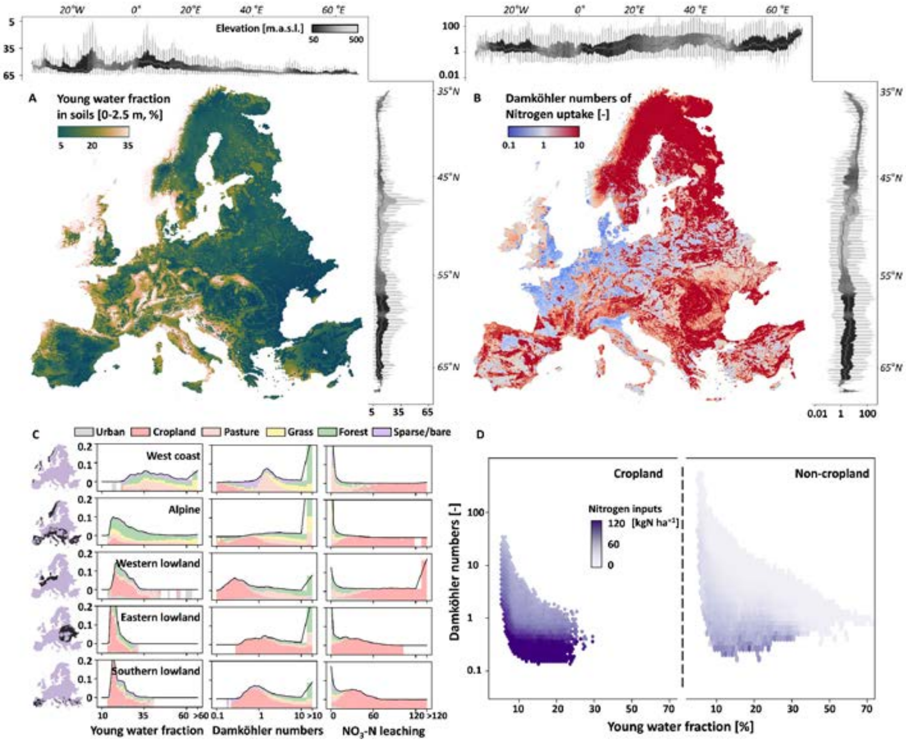

Fig. 1. Spatial patterns and statistical characteristics of young water fraction (YWF) and Damköhler number (Da) for nitrogen removal across Europe. Panels (A–B) show the mean values of YWF and Da during 1982–2024, with inset boxplots summarizing their minimum, maximum, 25th–75th percentiles, and median values along latitude, longitude, and elevation.

Panel (C) illustrates the probability density distributions of YWF (%), Da (-), and NO_3–N leaching (kg N ha^{-1}) across different land-use types. Panel (D) presents the correlations among YWF, Da, and nitrogen inputs.

Magnitudes of Hydrological Change Control the Evolution of Nitrogen Cycling

While spatial contrasts reveal the critical role of water velocity in buffering nitrogen leaching through a snapshot of the past 45 years, it remains unclear on how such buffering evolved under climate change. Therefore, we further investigated the temporal shifts in water velocities and the response of nitrogen cycling since the 1980s.

Hydrological shifts across Europe were primarily driven by precipitation (89.8%), evapotranspiration (6.1%) and land-use change (4.1%) (text S7; fig. S6). In regions with increasing precipitation inputs, water cycling accelerated with higher velocity of water fluxes. Typical examples include the Alpine regions in Southern Europe (e.g., Apennines, Pyrenees, and Iberian mountains; fig. 2A), where average travel time of soil water and young water fraction decreased by 219 days and 2.3% from 1980s to 2020s. In contrast, Central-Western Europe experienced a moderate precipitation decline (averaging 1 mm yr^{-1}). This, together with the ubiquitous increase in temperature and radiation fluxes, led to a higher fraction of water returning to atmosphere via evapotranspiration (0.1% increase per year) (*23*). The drying trend further decelerated water cycling, increasing water ages by 109 days and decreasing young water fractions by -1.3% in the past 45 years (fig. 2A, I). Similar deceleration of water cycling was also found in Northern Europe (sparsely distributed across Nordic mountains and adjacent highlands in mid Norway and Sweden with young water fractions varying by -1.0% from 1980s to 2020s), Eastern Europe (the Baltic Plain; -0.7%), Southern Europe (non-alpine regions in the Iberian Peninsula; -1.0%), and the South-West corner and eastern part of Turkish mountains (fig. 2A).

These hydrological shifts controlled the evolution of nitrogen cycling. From a hydrological standpoint, decelerated water cycling should, intuitively, mitigate nitrogen accumulation and leaching in soil water, as slower turnover rates tend to prolong the soil travel times, thus extending the window for nitrogen removal. However, our simulations revealed diverse responses, as the kinetics of biogeochemical processes (e.g., plant uptake and soil denitrification) were also modulated by moisture conditions, which concurrently affected reaction timescales (*3*). This resulted in bi-directional correlations between travel times and Damköhler numbers, formalised in four co-evolution schemes (fig. 3A-B):

Accelerating cycling: The acceleration of water cycling was mainly driven by enhanced hydrological fluxes, often accompanied by increased soil moisture. Higher saturation levels, coupled with warmer condition, favored nitrogen uptake by extending the duration of denitrification and plant uptake (*2, 5*). When biogeochemical acceleration outweighed the marginal decrease in soil travel times, an optimal Scheme A emerged, reflected by younger water, higher Damköhler numbers, and lower nitrogen leaching (fig. 3B-C). However, a sufficiently intense acceleration of water cycling could result in an opposing Scheme C with younger water and lower Damköhler numbers. This scheme occurs in relatively dry regions that undergo abrupt rewetting (with low soil moisture yet high infiltration increases; fig. 3C). The pronounced acceleration of water cycling enabled rapid flushing of nitrogen from soil instead of being removed by biogeochemical processes, elevating the risks of leaching.

Decelerating cycling: Decelerated water cycling could also nudge nitrogen cycling in both directions. Generally, deceleration enables longer residence times and reaction windows for biogeochemical processing, formalizing Scheme B with older water, higher Damköhler numbers, and lower potentials of nitrogen accumulation and leaching (fig. 3B-C). However, if climate change further amplifies drought conditions, continuous decrease in soil moisture may inhibit nitrogen uptake, especially denitrification that relies heavily on soil saturation (*24*). When such moisture-related biogeochemical depressions are no longer marginal but dominant compared to the extended travel time, the nitrogen surplus will increase considerably, leading to higher chances of both nitrogen accumulation and leaching (fig. 3C). This formalizes the last and worst Scheme D with older water and lower Damköhler numbers.

Overall, both accelerated and decelerated water cycling may drive nitrogen cycling along negative trajectories (with increased nitrogen accumulation or leaching). The outcome depends not only on the direction of hydrological shifts, but also on its magnitude (fig. 3B). Moderate hydrological shifts tend to guide nitrogen cycling towards positive trajectories with mitigated nitrogen accumulation and leaching, while increasing nitrogen risks might be triggered by intense hydrological shifts, including abrupt wetting (causing intense acceleration of water fluxes) or prolonged drying (causing potential violation of the criteria for biogeochemical processes). Here, to synthesize this behavior, we introduce the concept of "wetness boundaries" – shifting within these boundaries promotes positive nitrogen cycling with mitigated risks while vice versa (method and uncertainty in text S8, fig. S8). For general applicability, the boundaries are approximated by expressing precipitation change (%) as a function of variability in temperature ΔT (°C) and anthropogenic addition ΔA (kg $ha^{-1}yr^{-1}$):

$$\Delta P = [-6.4 + 0.8 * \Delta T - 0.04 * \Delta A, 10.9 + 0.5 * \Delta T - 0.1 * \Delta A]$$

Elevated anthropogenic inputs tend to narrow both upper and lower boundaries, increasing the likelihood of nitrogen accumulation and leaching through either acceleration or deceleration of water cycling. The latter scheme can also be triggered by warming, which reflects dual increase in vulnerability of water and nitrogen cycling.

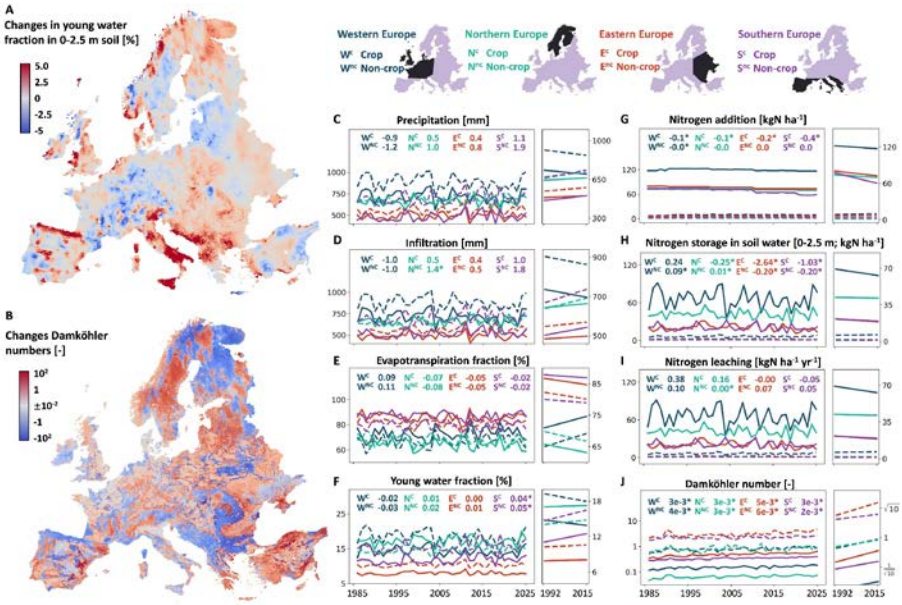

Fig. 2. Temporal dynamics of hydrological and nitrogen states/fluxes during 1982–2024. Panel (A) shows the differences of young water fraction and Damköhler numbers between 1982–1992 and 2014–2024 (A–B). Panels (C–J) show the annual dynamics of representative hydrological and nitrogen states, fluxes, and statistics across four different European regions (reflected with four colours). In each pair of panels, cropland (C) and non-crop (NC) regions are represented by solid and dotted lines, with linear regressions displayed in the right-hand. The numbers are the slopes of linear regression with * indicating the significance of monotonic trend ($P < 0.05$).

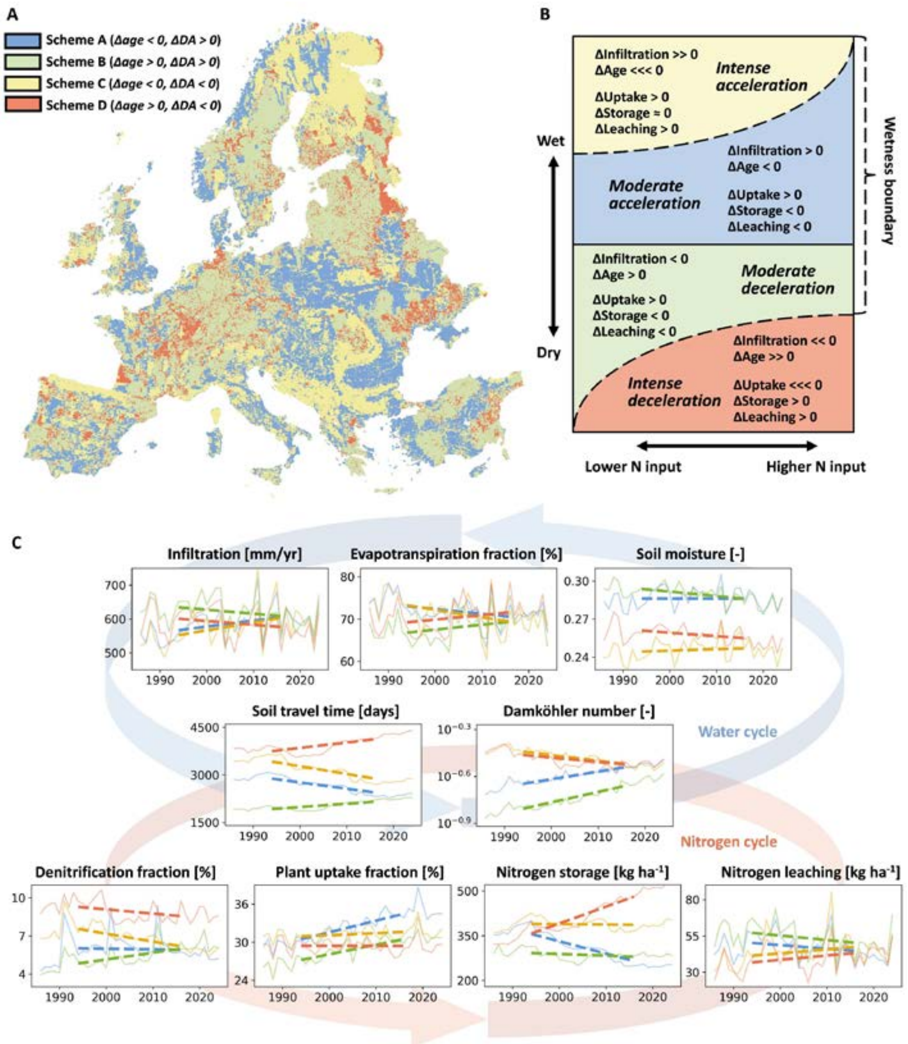

Fig. 3. Divergent evolution schemes of water and nitrogen cycling. Panel (A) and (B) show their spatial distribution and conceptual schematics, respectively. Panel (C) summarises temporal trends of key hydrological and nitrogen states, fluxes, and statistics under each of the four schemes. The full and dotted lines represent the annual means and linear regressions, respectively. Age, Uptake, Leaching, and Leaching are the abbreviation of soil travel time, nitrogen uptake (sum of soil denitrification and plant uptake), soil water nitrogen storage, and nitrogen leaching, respectively.

Divergent Trajectories of Nitrogen Evolution in an Aging Future

The co-evolution of water and nitrogen cycling over the past four decades raises a critical question: will these trajectories persist under future climate change? To explore this, we extended the simulation towards 2100 based on ensembles of CMIP6 projections (text S9). Two scenarios, SSP1-2.6 and SSP5-8.5, were investigated, representing low-emission sustainable and high-emission fossil-fuel development pathways, respectively.

Future projections reveal divergent hydrological responses along a latitudinal gradient, shifting from acceleration in high-latitude regions to deceleration in Central-West, Eastern, and Southern Europe (fig. 4D, S10). The trends are mainly driven by precipitation variability, exhibiting a "WWDD" trend where wet and dry regions get wetter and drier due to the warming-induced changes in atmospheric water-vapor concentration (fig. S9) (*25*). An amplified drying is expected under the high-emission scenario due to higher temperature, radiation and the resulting precipitation change following the Clausius-Clapeyron relationship (*26*). This led to stronger deceleration of water cycling, evidenced by a decrease in median young water fraction of 1.8% by 2100 under SSP5-8.5 while remaining relatively stable in most regions under SSP1-2.6 (fig. 4A). Notably, dry regions exhibited additional vulnerability to climatic change in both scenarios (*27*), as the median young water fractions declined to 4.8% (SSP1-2.6) and 3.5% (SSP5-8.5) across Eastern-Southern Europe, respectively (fig. S10).

This hydroclimatic discrepancy further propagated to nitrogen cycling, nudging it along diverse trajectories. Under the low-emission scenario (SSP1-2.6), the acceleration and deceleration of water cycling remained generally mild (fig. 4D), largely confined within the wetness boundaries under limited climatic variability (fig. S8). Combined with increasing temperatures that extended growing seasons, these conditions substantially enhanced nitrogen uptake, resulting in positive shifts in Damköhler numbers across 75.7% of Europe. Such enhanced uptake efficiency was even stronger under the high-emission scenario (SSP5-8.5) due to the higher temperature, accounting for 78.3% of Europe (fig. 4E). However, further drying in Eastern and Southern Europe projected potential hydrological perturbations beyond the lower wetness boundaries under SSP5-8.5, leading to systematic moisture-induced depression of nitrogen uptake that overrode the extended residence times (fig. S11C). In these regions, nitrogen storage is projected to double by 2100 (fig. S11F), posing severe challenges to water quality due to quick flushing of accumulated nitrogen during flood events (*28*), a risk exacerbated by projected shifts toward extreme precipitation events with higher frequency and intensity (*29*).

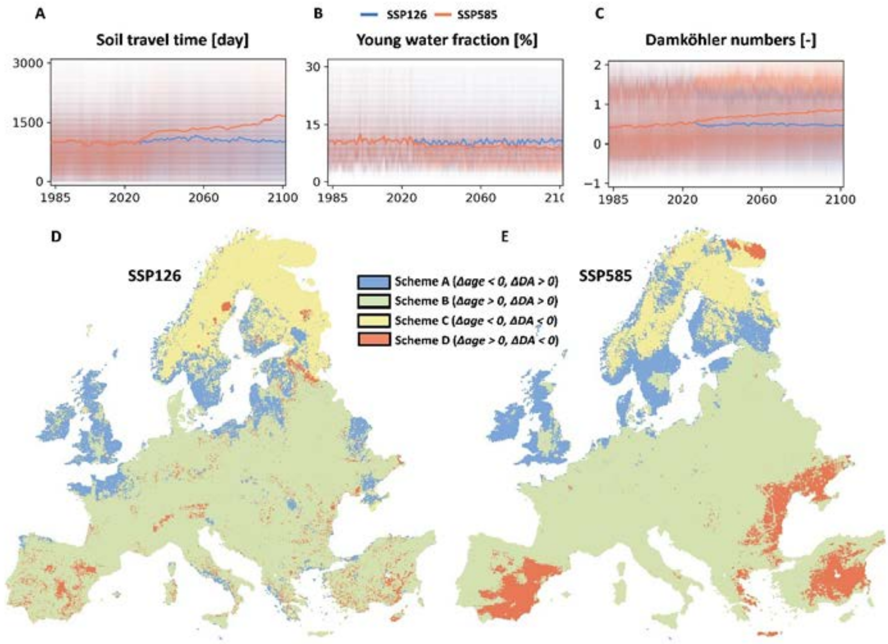

Fig. 4. The projected evolution of water and nitrogen cycling from 2025 to 2100. Panels (A-C) show the temporal dynamics of median soil travel time, young water fraction, and Damköhler numbers across Europe, respectively. Panels (D) and (E) show the spatial distribution of the four water–nitrogen co-evolution schemes under SSP1-2.6 and SSP5-8.5 scenarios. The schemes are classified based on the projected changes in soil travel time and Damköhler numbers between 2015-2025 and 2090-2100 (fig. S10).

Discussion

A persistent barrier of understanding nitrogen cycling arises from the long-standing neglect of water velocities (*7*), despite their fundamental control over solute transport and mixing in landscapes (*20*). Consequently, water quality modelling often remains a grey box — capable of reproducing riverine water-quality dynamics yet sometimes for "for the wrong reasons" (*8*, *12*, *30*). Here, our isotope-aided water velocity framework, though subject to some inherent uncertainties (text S10), represents a step towards unboxing the complex interactive water-nitrogen co-cycling through the lens of velocities. Such an approach complements previous studies focusing primarily on celerity and offers important perspectives on several enduring puzzles in nitrogen research. For example, the delayed ecohydrological response to nitrogen interventions, over years to decades (*31*), can not only be attributed to long groundwater residence times, but also the prolonged soil travel times in European lowlands. Likewise, the stronger leaching in Central-Western relative to Eastern-Southern Europe emerges not only from higher fertilizer inputs and deposition (*32*) but also from faster hydrological turnover.

Among these insights, one key finding is how water velocities have reshaped nitrogen cycling under climate change. We reveal that the evolution trajectories of nitrogen cycling depend primarily on the magnitude of velocity shifts: mild changes promote positive evolution (declining nitrogen storage and leaching), whereas strong acceleration or deceleration drives negative evolution (increasing storage or leaching). To conceptualise this behaviour, we introduce the concept of "wetness boundaries", where transitions within or across these boundaries determine whether evolution is positive or negative. These boundaries are shaped by the local wetness conditions and evolve along the changing climate and anthropogenic boundaries.

This theory reframes the long-standing question of whether global warming will exacerbate nitrogen pollution. Based on current simulations, the answer is unlikely for most of Europe. Warming generally decelerates water cycling, extending water travel times and promoting nitrogen uptake (*33*). Even where warming accelerates water cycling, as in Northern Europe, the shortened residence times are offset by temperature-stimulated uptake and extended growing seasons. However, the magnitude of hydrological shifts remains pivotal for nitrogen evolution, which may push ecosystems beyond the wetness boundaries under high-mission scenarios (SSP5-8.5). An example arises from Eastern and Southern Europe where moisture-induced suppression of nitrogen uptake was projected with subsequent nitrogen accumulation (*24*). Such drought-induced deterioration (Scheme D), reflecting dual vulnerability of water quantity and quality, is expected to emerge not only in Europe, but also in other regions like Central–East Asia where declining water storage has been observed (*34*) and is projected to exacerbate in the 21st century (*35*). Similarly, the opposite scheme (Scheme C) is likely to occur in tropical regions (e.g., South America and Southeast Asia) where rapid water cycling may further intensify under future warming (*27*). These extrapolations suggest a greater global risk of nitrogen deterioration than what European projections alone would imply (*36*).

How can such negative evolution trajectories be prevented? Within the water velocity framework, the central objective would be to keep ecosystems from crossing their wetness boundaries. The direct route is to limit the magnitude of climatic shifts by reducing fossil-fuels emissions (*37*). In parallel, hydrological shifts can be moderated through anthropogenic interventions, e.g., constructing wetlands as riverine buffers to decelerate flow velocities (*12*). Finally, we can expand the safe operating space by widening wetness boundaries through measures that reduce nitrogen inputs, including systematic crop rotation (*38*), optimizing fertilization (*39*), improved wastewater treatment (*40*), and developing varieties for improved nitrogen use efficiency with breeding or genetically engineering techniques (*36*).

References and Notes

1. W. Steffen, K. Richardson, J. Rockström, S. Cornell, I. Fetzer, E. Bennett, R. Biggs, S. Carpenter, W. Vries, C. de Wit, C. Folke, D. Gerten, J. Heinke, L. Persson, V. Ramanathan, B. Reyers, S. Sörlin, "Planetary Boundaries: Guiding Human Development on a Changing Planet." *Science*, doi: 10.1126/science.1259855 (2015).

2. M. T. H. van Vliet, J. Thorslund, M. Strokal, N. Hofstra, M. Flörke, H. Ehalt Macedo, A. Nkwasa, T. Tang, S. S. Kaushal, R. Kumar, A. van Griensven, L. Bouwman, L. M. Mosley, Global river water quality under climate change and hydroclimatic extremes. *Nat Rev Earth Environ* **4**, 687–702 (2023).

3. C. Wellen, A.-R. Kamran-Disfani, G. Arhonditsis, Evaluation of the Current State of Distributed Watershed Nutrient Water Quality Modeling. *Environmental Science & Technology* **49** (2015).

4. J. Galloway, A. Townsend, J. W. Erisman, M. Bekunda, Z. Cai, J. Freney, L. Martinelli, S. Seitzinger, M. Sutton, Transformation of the Nitrogen Cycle: Recent Trends, Questions, and Potential Solutions. *Science (New York, N.Y.)* **320**, 889–92 (2008).

5. D. E. Canfield, A. N. Glazer, P. G. Falkowski, The Evolution and Future of Earth's Nitrogen Cycle. *Science* **330**, 192–196 (2010).

6. J. Duncan, P. Groffman, L. Band, Towards closing the watershed nitrogen budget: Spatial and temporal scaling of denitrification. *Journal of Geophysical Research: Biogeosciences* **118** (2013).

7. J. McDonnell, K. Beven, Debates on Water Resources: The future of hydrological sciences: A (common) path forward? A call to action aimed at understanding velocities, celerities and residence time distributions of the headwater hydrograph. *Water Resources Research* **50** (2014).

8. J. W. Kirchner, Getting the right answers for the right reasons: Linking measurements, analyses, and models to advance the science of hydrology. *Water Resources Research* **42**, W03S04 (2006).

9. T. Huntington, Evidence for Intensification of the Global Water Cycle: Review and Synthesis. *Journal of Hydrology* **319**, 83–95 (2006).

10. D. Tetzlaff, J. Buttle, S. K. Carey, K. Mcguire, H. Laudon, C. Soulsby, Tracer-based assessment of flow paths, storage and runoff generation in northern catchments: A review. *Hydrological Processes* **29**, 3475–3490 (2015).

11. H. Jung, D. Tetzlaff, C. Birkel, C. Soulsby, Recent Developments and Emerging Challenges in Tracer-Aided Modeling. *Wiley Interdisciplinary Reviews Water* **12** (2025).

12. S. Wu, D. Tetzlaff, X. Yang, T. Sauter, C. Soulsby, Hydrological Connectivity Dominates NO3-N Cycling in Complex Landscapes – Insights From Integration of Isotopes and Water Quality Modeling. *Water Resources Research* **61**, e2025WR040525 (2025).

13. X. Yang, D. Tetzlaff, J. Jin, Q. Li, D. Borchardt, C. Soulsby, Linking terrestrial biogeochemical processes and water ages to catchment water quality: A new Damköhler analysis based on coupled modeling of isotope tracers and nitrate dynamics. *Water Research* **262**, 122118 (2024).

14. R. Kumar, F. Hesse, P. Rao, A. Musolff, J. Jawitz, F. Sarrazin, L. Samaniego, J. Fleckenstein, O. Rakovec, S. Thober, S. Attinger, Strong hydroclimatic controls on vulnerability to subsurface nitrate contamination across Europe. *Nature Communications* **11** (2020).

15. S. Wu, D. Tetzlaff, Y. Zheng, C. Soulsby, EcoTWIN 1.0: a fully distributed tracer-aided ecohydrological model tracking water, isotopes, and nutrients. *Geoscientific Model Development* **19**, 2257–2278 (2026).

16. R. Trigo, T. Osborn, J. Corte-Real, The North Atlantic Oscillation influence on Europe: Climate impacts and associated physical mechanisms. *Climate Research - CLIMATE RES* **20**, 9–17 (2002).

17. S. Jasechko, J. Kirchner, J. Welker, J. McDonnell, Substantial proportion of global streamflow less than three months old. *Nature Geoscience* **9** (2016).

18. A. Gentile, D. Canone, N. Ceperley, D. Gisolo, M. Previati, G. Zuecco, B. Schaefli, S. Ferraris, Towards a conceptualization of the hydrological processes behind changes of young water fraction with elevation: a focus on mountainous alpine catchments. *Hydrology and Earth System Sciences* **27**, 2301–2323 (2023).

19. Y. Vystavna, L. Chavanne, A. Harjung, D. Soto, A. Watson, J. Miller, J. Cullmann, Predicting river flow dynamics using stable isotopes for better adaptation to climate and land-use changes. *Nature Water* **2** (2024).

20. L. Li, J. Knapp, G.-H. Ng, J. Perdrial, P. Sullivan, W. Zhi, River water quality shaped by land–river connectivity in a changing climate. *Nature Climate Change* **14** (2024).

21. X. Yang, X. Zhang, D. Graeber, R. Hensley, H. Jarvie, A. Lorke, D. Borchardt, Q. Li, M. Rode, Large-stream nitrate retention patterns shift during droughts: Seasonal to sub-daily insights from high-frequency data-model fusion. *Water Research* **243**, 120347 (2023).

22. F. Mikayilov, E. Rouholahnejad Freund, S. Ashraf Vaghefi, R. Srinivasan, H. Yang, B. Klöve, A continental-scale hydrology and water quality model for Europe: Calibration and uncertainty of a high-resolution large-scale SWAT model. *Journal of Hydrology* **524** (2015).

23. L. Samaniego, Permanent shifts in the global water cycle. *Science* **387**, 1348–1350 (2025).

24. Z. Yan, B. Bond-Lamberty, K. Todd-Brown, V. Bailey, S.-L. Li, C. Liu, C. Liu, A moisture function of soil heterotrophic respiration that incorporates microscale processes. *Nature Communications* **9** (2018).

25. P. Greve, B. Orlowsky, B. Mueller, J. Sheffield, M. Reichstein, S. Seneviratne, Global assessment of trends in wetting and drying over land. *Nature Geoscience* **7** (2014).

26. R. B. Stull, C. D. Ahrens, *Meteorology for Scientists and Engineers* (Brooks/Cole, Pacific Grove, Calif, 2nd Ed.; https://www.eoas.ubc.ca/books/Practical_Meteorology/mse3.html).

27. S. Kumar, R. Allan, F. Zwiers, D. Lawrence, P. Dirmeyer, Revisiting Trends in Wetness and Dryness in the Presence of Internal Climate Variability and Water Limitations Over Land. *Geophysical Research Letters* **42**, 10,867-10,875 (2015).

28. T. Bowles, S. Atallah, E. Campbell, A. Gaudin, W. Wieder, S. Grandy, Addressing agricultural nitrogen losses in a changing climate. *Nature Sustainability* **1** (2018).

29. P. Yeh, C. Wu, Recent Acceleration of the Terrestrial Hydrologic Cycle in the U.S. Midwest. *Journal of Geophysical Research: Atmospheres* **123** (2018).

30. K. Beven, P. Smith, Concepts of Information Content and Likelihood in Parameter Calibration for Hydrological Simulation Models. *Journal of Hydrologic Engineering* **20**, 6916 (2015).

31. European Environment Agency, "European Waters-Assessment of Status and Pressures" (Publications Office of the European Union, Luxembourg, 2018); https://www.eea.europa.eu/publications/state-of-water.

32. M. A. Sutton, C. M. Howard, J. W. Erisman, G. Billen, A. Bleeker, P. Grennfelt, H. van Grinsven, B. Grizzetti, Eds., *The European Nitrogen Assessment: Sources, Effects and Policy Perspectives* (Cambridge University Press, Cambridge, 2011; https://www.cambridge.org/core/books/european-nitrogen-assessment/7156D2A2F03CD36FFA0B6EC60BF9A497).

33. T. Greaver, C. Clark, J. Compton, D. Vallano, A. Talhelm, C. Weaver, L. Band, J. Baron, E. Davidson, C. Tague, E. Felker-Quinn, J. Lynch, J. Herrick, L. Liu, C. Goodale, K. Novak, R. Haeuber, Key ecological responses to nitrogen are altered by climate change. *Nature Climate Change* **6**, 836–843 (2016).

34. J. Wang, C. Song, J. Reager, F. Yao, J. Famiglietti, Y. Sheng, G. MacDonald, F. Brun, H. Müller Schmied, R. Marston, Y. Wada, Recent global decline in endorheic basin water storages. *Nature Geoscience* **11**, 926–932 (2018).

35. C. Chou, J. Neelin, C.-A. Chen, J.-Y. Tu, Evaluating the "Rich-Get-Richer" Mechanism in Tropical Precipitation Change under Global Warming. *Journal of Climate - J CLIMATE* **22**, 1982–2005 (2009).

36. X. Zhang, E. A. Davidson, D. L. Mauzerall, T. D. Searchinger, P. Dumas, Y. Shen, Managing nitrogen for sustainable development. *Nature* **528**, 51–59 (2015).

37. D. Armstrong McKay, A. Staal, J. Abrams, R. Winkelmann, B. Sakschewski, S. Loriani, I. Fetzer, S. Cornell, J. Rockström, T. Lenton, Exceeding 1.5°C global warming could trigger multiple climate tipping points. *Science (New York, N.Y.)* **377**, eabn7950 (2022).

38. L. You, G. Ros, Y.-L. Chen, Q. Shao, M. Young, F. Zhang, W. Vries, Global mean nitrogen recovery efficiency in croplands can be enhanced by optimal nutrient, crop and soil management practices. *Nature Communications* **14** (2023).

39. C. Ren, L. He, L. Rosa, Integrated irrigation and nitrogen optimization is a resource-efficient adaptation strategy for US maize and soybean production. *Nature Food* **6**, 389–400 (2025).

40. G. Van Drecht, A. F. Bouwman, J. M. Knoop, A. H. W. Beusen, C. R. Meinardi, Global modeling of the fate of nitrogen from point and nonpoint sources in soils, groundwater, and surface water. *Global Biogeochemical Cycles* **17** (2003).

41. The model EcoTWIN v1.0 is archived in https://doi.org/10.5281/zenodo.16747633.

42. G. Lindström, C. Pers, J. Rosberg, J. Strömqvist, B. Arheimer, Development and test of the HYPE (Hydrological Predictions for the Environment) model – A water quality model for different spatial scales. *Hydrology Research* **41** (2010).

43. X. Yang, S. Jomaa, M. Zink, J. Fleckenstein, D. Borchardt, M. Rode, A New Fully Distributed Model of Nitrate Transport and Removal at Catchment Scale. *Water Resources Research* **54** (2018).

44. K. Winkler, R. Fuchs, M. Rounsevell, M. Herold, Global land use changes are four times greater than previously estimated. *Nature Communications* **12**, 2501 (2021).

45. Y. Liu, R. Liu, J. M. Chen, Retrospective retrieval of long-term consistent global leaf area index (1981–2011) from combined AVHRR and MODIS data. *Journal of Geophysical Research: Biogeosciences* **117** (2012).

46. J. Zhu, Y. Jia, G. Yu, Q. Wang, N. He, Z. Chen, H. He, X. Zhu, P. Li, F. Zhang, X. Liu, K. Goulding, D. Fowler, P. Vitousek, Changing patterns of global nitrogen deposition driven by socio-economic development. *Nat Commun* **16**, 46 (2025).

47. W. Adalibieke, X. Cui, H. Cai, L. You, F. Zhou, Global crop-specific nitrogen fertilization dataset in 1961–2020. *Sci Data* **10**, 617 (2023).

48. B. Grizzetti, O. Vigiak, A. U. Moinelo, A. Aloe, M. Zanni, F. Bouraoui, A. Pistocchi, C. Dorati, R. Friedland, A. D. Roo, C. B. Sanz, A. Leip, M. Bielza, Nitrogen and phosphorus diffuse sources in European freshwater (2005-2012) - (v. mar.2019), European Commission, Joint Research Centre (JRC) (2019); http://data.europa.eu/89h/e249aac0-5914-45f3-93ea-e3651a665fe1.

49. S. Wu, D. Tetzlaff, X. Yang, C. Soulsby, Disentangling the influence of landscape characteristics, hydroclimatic variability and land management on surface water NO_3 -N dynamics: spatially distributed modelling over 30 years in a lowland mixed land use catchment. *Water Resources Research*, e2021WR030566 (2022).

50. S. Wu, D. Tetzlaff, C. Soulsby, Revising Common Approaches for Calibration: Insights From a 1-D Tracer-Aided Hydrological Model With High-Dimensional Parameters and Objectives. *Water Resources Research* **61**, e2024WR037656 (2025).

51. B. S. Naz, W. Sharples, Y. Ma, K. Goergen, S. Kollet, Continental-scale evaluation of a fully distributed coupled land surface and groundwater model, ParFlow-CLM (v3.6.0), over Europe. *Geoscientific Model Development* **16**, 1617–1639 (2023).

52. C. Donnelly, J. C. M. Andersson, B. Arheimer, Using flow signatures and catchment similarities to evaluate the E-HYPE multi-basin model across Europe. *Hydrological Sciences Journal* **61**, 255–273 (2016).

53. S. Wang, M. Zhang, F. Chen, Y. Che, M. Du, Y. Liu, Comparison of GCM-simulated isotopic compositions of precipitation in arid central Asia. *Journal of Geographical Sciences* **25**, 771–783 (2015).

54. E. R. Jones, M. F. P. Bierkens, N. Wanders, E. H. Sutanudjaja, L. P. H. van Beek, M. T. H. van Vliet, DynQual v1.0: a high-resolution global surface water quality model. *Geoscientific Model Development* **16**, 4481–4500 (2023).

55. A. Smith, D. Tetzlaff, L. Kleine, M. Maneta, C. Soulsby, Quantifying the effects of land use and model scale on water partitioning and water ages using tracer-aided ecohydrological models. *Hydrology and Earth System Sciences* **25**, 2239–2259 (2021).

56. J. McDonnell, The two water worlds hypothesis: Ecohydrological separation of water between streams and trees? *Wiley Interdisciplinary Reviews: Water* **1** (2014).

57. M. Hrachowitz, H. Savenije, T. A. Bogaard, D. Tetzlaff, C. Soulsby, What can flux tracking teach us about water age distribution patterns and their temporal dynamics? *Hydrology and Earth System Sciences* **17**, 533–564 (2013).

58. M. Sprenger, C. Stumpp, M. Weiler, W. Aeschbach, S. Allen, P. Benettin, M. Dubbert, A. Hartmann, M. Hrachowitz, J. Kirchner, J. McDonnell, N. Orlowski, D. Penna, S. Pfahl, M.

Rinderer, N. Rodriguez, M. Schmidt, C. Werner, The Demographics of Water: A Review of Water Ages in the Critical Zone. *Reviews of Geophysics* **57** (2019).

59. G.-Y. Niu, Z.-L. Yang, K. Mitchell, M. Ek, M. Barlage, A. Kumar, K. Manning, D. Niyogi, E. Rosero, M. Tewari, Y. Xia, The community Noah land surface model with multiparameterization options (Noah-MP): 1. Model description and evaluation with local-scale measurements. *Journal of Geophysical Research* **116**, D12109 (2011).

60. K. Oleson, D. Lawrence, G. Bonan, B. Drewniak, M. Huang, C. Koven, S. Levis, F. Li, W. Riley, Z. Subin, S. Swenson, P. Thornton, A. Bozbiyik, F. Rosie, C. Heald, E. Kluzek, J.-F. Lamarque, P. Lawrence, L. Leung, Z.-L. Yang, *Technical Description of Version 4.5 of the Community Land Model (CLM)* (2013).

61. C. Yang, L. Condon, R. Maxwell, Unravelling groundwater–stream connections over the continental United States. *Nature Water* **3** (2025).

62. M. E. Baum, J. E. Sawyer, E. D. Nafziger, M. J. Castellano, M. D. McDaniel, M. A. Licht, D. J. Hayes, M. J. Helmers, S. V. Archontoulis, The optimum nitrogen fertilizer rate for maize in the US Midwest is increasing. *Nat Commun* **16**, 404 (2025).

63. R. Einarsson, A. Sanz-Cobena, E. Aguilera, G. Billen, J. Garnier, H. J. M. van Grinsven, L. Lassaletta, Crop production and nitrogen use in European cropland and grassland 1961–2019. *Sci Data* **8**, 288 (2021).

64. M. Batool, F. J. Sarrazin, X. Zhang, A. Musolff, T. V. Nguyen, S. Attinger, R. Kumar, Scenario analysis of nitrogen surplus typologies in Europe shows that a 20% fertilizer reduction may fall short of 2030 EU Green Deal goals. *Nat Food* **6**, 787–798 (2025).

65. A. Pistocchi, R. Friedland, M. Zanni, O. Vigiak, A. Aloe, B. Grizzetti, C. Dorati, F. Bouraoui, M. Bielza, A. Leip, C. Benitez Sanz, A. Udias Moinelo, A. De Roo, Scenarios of nitrogen and phosphorus point sources in European freshwater (v. mar.2019), version 49dfea8c3c3b4c96a0d363dbbc32ea2f, European Commission, Joint Research Centre (2020); https://doi.org/10.2905/JRC.J5YMB7R.

66. M. Meinshausen, Z. R. J. Nicholls, J. Lewis, M. J. Gidden, E. Vogel, M. Freund, U. Beyerle, C. Gessner, A. Nauels, N. Bauer, J. G. Canadell, J. S. Daniel, A. John, P. B. Krummel, G. Luderer, N. Meinshausen, S. A. Montzka, P. J. Rayner, S. Reimann, S. J. Smith, M. van den Berg, G. J. M. Velders, M. K. Vollmer, R. H. J. Wang, The shared socio-economic pathway (SSP) greenhouse gas concentrations and their extensions to 2500. *Geoscientific Model Development* **13**, 3571–3605 (2020).

67. R. A. Betts, O. Boucher, M. Collins, P. M. Cox, P. D. Falloon, N. Gedney, D. L. Hemming, C. Huntingford, C. D. Jones, D. M. H. Sexton, M. J. Webb, Projected increase in continental runoff due to plant responses to increasing carbon dioxide. *Nature* **448**, 1037–1041 (2007).

68. F. Li, J. Xiao, J. Chen, A. Ballantyne, K. Jin, B. Li, M. Abraha, R. John, Global water use efficiency saturation due to increased vapor pressure deficit. *Science* **381**, 672–677 (2023).

69. S. Fatichi, S. Leuzinger, A. Paschalis, J. Langley, A. Donnellan Barraclough, M. Hovenden, Partitioning direct and indirect effects reveals the response of water-limited ecosystems to elevated CO2. *Proceedings of the National Academy of Sciences of the United States of America* **113** (2016).

70. C. S. Lesk, J. M. Winter, J. S. Mankin, Projected runoff declines from plant physiological effects on precipitation. *Nat Water* **3**, 167–177 (2025).

71. D. Frank, B. Poulter, M. Saurer, J. Esper, C. Huntingford, G. Helle, K. Treydte, N. Zimmermann, G. Schleser, A. Ahlström, P. Ciais, P. Friedlingstein, S. Levis, M. Lomas, S. Sitch, N. Viovy, L. Andreu-Hayles, Z. Bednarz, F. Berninger, M. Weigl-Kuska, Water-use efficiency and transpiration across European forests during the Anthropocene. *Nature Climate Change*, doi: 10.1038/NCLIMATE2614 (2015).

72. B. Gu, X. Zhang, S. Lam, Y. Yu, H. Grinsven, S. Zhang, X. Wang, B. Bodirsky, S. Wang, J. Duan, C. Ren, A. Bouwman, W. Vries, J. xu, M. Sutton, D. Chen, Cost-effective mitigation of nitrogen pollution from global croplands. *Nature* **613**, 77–84 (2023).

73. A. Bouwman, A. Beusen, G. Billen, Human alteration of the global nitrogen and phosphorus soil balances for the period 1970–2050. *Global Biogeochemical Cycles - GLOBAL BIOGEOCHEM CYCLE* **23** (2009).

74. J. Galloway, F. Dentener, E. Boyer, R. Howarth, S. Seitzinger, G. Asner, C. Cleveland, P. Green, E. Holland, D. Karl, A. Michaels, J. Porter, A. Townsend, C. Vöosmarty, Nitrogen Cycles: Past, Present, and Future. *Biogeochemistry* **70**, 153–226 (2004).

75. N. Hofstra, A. Bouwman, Denitrification in Agricultural Soils: Summarizing Published Data and Estimating Global Annual Rates. *Nutrient Cycling in Agroecosystems* **72**, 267–278 (2005).

76. S. Seitzinger, J. Harrison, J. Bohlke, A. Bouwman, R. Lowrance, B. Peterson, C. Tobias, G. Drecht, Denitrification Across Landscapes and Waterscapes: A Synthesis. *Ecological applications : a publication of the Ecological Society of America* **16**, 2064–90 (2007).

77. B. Pan, L. Xia, S. K. Lam, E. Wang, Y. Zhang, A. Mosier, D. Chen, A global synthesis of soil denitrification: Driving factors and mitigation strategies. *Agriculture, Ecosystems & Environment* **327**, 107850 (2022).

78. A. F. Bouwman, A. H. W. Beusen, J. Griffioen, J. W. Van Groenigen, M. M. Hefting, O. Oenema, P. J. T. M. Van Puijenbroek, S. Seitzinger, C. P. Slomp, E. Stehfest, Global trends and uncertainties in terrestrial denitrification and N2O emissions. *Philos Trans R Soc Lond B Biol Sci* **368**, 20130112 (2013).

79. Y. Xu, Z. Xu, Z. Cai, F. Reverchon, Review of denitrification in tropical and subtropical soils of terrestrial ecosystems. *J Soils Sediments* **13**, 699–710 (2013).

80. D. Fowler, M. Coyle, U. Skiba, M. Sutton, J. Cape, S. Reis, L. Sheppard, A. Jenkins, B. Grizzetti, J. Galloway, P. Vitousek, A. Leach, A. Bouwman, K. Butterbach-Bahl, F. Dentener, M. Amann, M. Voss, The Global Nitrogen Cycle in the Twenty-First Century. *Philosophical transactions of the Royal Society of London. Series B, Biological sciences* **368**, 20130164 (2013).

81. A. Bouwman, K. Klein Goldewijk, K. Van der Hoek, A. Beusen, D. Vuuren, J. Willems, M. Rufino, E. Stehfest, Exploring global changes in nitrogen and phosphorus cycles in agriculture induced by livestock production over the 1900-2050 period, Livestock and Global Change Special Feature. *Proceedings of the National Academy of Sciences of the United States of America* **110** (2011).

82. Y. Peng, I. C. Prentice, K. J. Bloomfield, M. Campioli, Z. Guo, Y. Sun, D. Tian, X. Wang, S. Vicca, B. D. Stocker, Global terrestrial nitrogen uptake and nitrogen use efficiency. *Journal of Ecology* **111**, 2676–2693 (2023).

83. J. B. Fisher, S. Sitch, Y. Malhi, R. A. Fisher, C. Huntingford, S.-Y. Tan, Carbon cost of plant nitrogen acquisition: A mechanistic, globally applicable model of plant nitrogen uptake, retranslocation, and fixation. *Global Biogeochemical Cycles* **24** (2010).

84. H. Vallicrosa, K. Fleischer, M. Delgado-Baquerizo, M. Fernández-Martínez, J. Černý, D. Tian, A. Kourmouli, C. Mayoral, D. Grados, M. Lu, C. Terrer, Nitrogen deposition and climate drive plant nitrogen uptake while soil factors drive nitrogen use efficiency in terrestrial ecosystems. *Earth System Dynamics* **16**, 1183–1196 (2025).

85. J. Liu, L. You, M. Amini, M. Obersteiner, M. Herrero, A. Zehnder, H. Yang, A high-resolution assessment on global nitrogen flow in cropland. *Proceedings of the National Academy of Sciences of the United States of America* **107**, 8035–40 (2010).

86. J. Cui, X. Zhang, S. Reis, C. Wang, S. Wang, P. He, H. Chen, H. Grinsven, B. Gu, Nitrogen cycles in global croplands altered by elevated CO2. *Nature Sustainability* **6**, 1–11 (2023).

87. M. C. Braakhekke, K. T. Rebel, S. C. Dekker, B. Smith, A. H. W. Beusen, M. J. Wassen, Nitrogen leaching from natural ecosystems under global change: a modelling study. *Earth System Dynamics* **8**, 1121–1139 (2017).

Acknowledgments

Funding: Tetzlaff's and Wu's contributions were partly funded through the WETSCAPES2.0 project (DFG TRR410/1 2025). Tetzlaff was also partly funded through Leibniz Excellence project ISOSCALE and received funding from the "Wasserressourcenpreis 2024" of the Rüdiger Kurt Bode-Foundation. Contributions from Soulsby were supported as an International KSLA Guest Professor at SLU by the Wallenberg Foundation (WP2023-0001) and by Leibnitz Association Germany in the project Wetland Restoration in Peatlands.

Author contributions:

Conceptualization: SW, CS, YZ, AM, DT

Data curation: SW

Methodology: SW

Software: SW

Visualization: SW

Supervision: CS, DT

Writing (original draft preparation): SW

Writing (review and editing): SW, CS, YZ, AM, DT

Competing interests: Authors declare that they have no competing interests.

Data and materials availability: The model EcoTWIN v1.0 is archived in https://doi.org/10.5281/zenodo.16747633 (*41*). For further development please refer to GitHub repository: https://github.com/songjun-wu/EcoTWIN. The sources of geographic and climatic data and observations of discharge, in-stream isotopes, and instream nitrate are introduced in Supplementary material (text S2 and S3). No new materials were generated for this study.

Kommentiert [JS1]: This dataset must be assigned a reference number and included in the list of References and Notes because it has a DOI. Please note that this means references listed in both the main manuscript and the Supplementary Materials must be renumbered accordingly.

Supplementary Materials

Materials and Methods

Supplementary Text

Figs. S1 to S11

Tables S1 to S9

References (42-87)