Evaluation of global change impacts on diffuse pollution
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Abstract
In a global change context, several recent advances in the field of hydrology and biogeochemistry suggest that a move from a riparian to a river drainage basin perspective is necessary to reframe research and thus provide a more integrated scientific understanding to inform water- and land-use management and policy. We explore this assertion using the control of diffuse pollution as an exemplar.

Introduction and context
Climate warming and concomitant socioeconomic change over the next few decades threaten to alter hydrological and physicochemical characteristics of water bodies (including rivers, lakes and wetlands) and so impact on water-dependent ecosystems [1,2]. Shifts in aquatic and riparian ecosystem structure and function are expected due to so-called ‘unavoidable climate change’, with potential for impacts to be either heightened by ‘additive pressures’ or mitigated by management at the local scale. Climate change may increase the frequency and magnitude of floods while drought and water scarcity may also become more severe [3]. Such shifts in extreme climate-driven events (similar to those experienced across Europe in the summer of 2003) have major consequences for water quantity and quality. Even the type and status of water bodies (wetland, ephemeral stream and so on) are intrinsic climatic functions, hence subject to variation under climate change. In the near future, modifications to global agricultural policy may be expected to influence decision making and land-use management practices within watersheds. These policy changes are likely to occur irrespective of climate change, which itself will create new challenges and opportunities for land and water resource managers. Indeed, the marked acceleration of the global nitrogen cycle measured today is the direct consequence of significant anthropogenic inputs (mostly chemical fertilizer application) over the last 60 years, which have led to a doubling of reactive nitrogen in terrestrial ecosystems [4]. Moreover, the acceleration of the hydrological cycle due to climate change has resulted in a more rapid transfer of nitrogen through river drainage basins [5].

Since Hynes’s seminal paper [6] on the importance of considering the river and its valley in context, there has been a growing understanding of the role of the drainage basin in river water quality. Yet, for a long time, dominant paradigms (notably, the river continuum concept [7] and the spiralling concept [8]) have stressed instream microbial processes as the main factors in recycling and removing energy and matter originating from the drainage basin. Meanwhile, Peterjohn and Correll [9] provided evidence for the role of riparian zones along streams in buffering nutrient input from upslope. In the past three decades, hundreds of studies have evaluated the capacity of these channel-marginal wetlands to retain and/or remove different pollutants [10]. A concerted research effort identified riparian zones as biogeochemical ‘hot spots’ [11], which can efficiently remove nitrogen by a microbial denitrification process in temperate, continental, or Mediterranean climates so long as the local hydrogeomorphology conditions facilitate transfer of upslope nitrate to anaerobic riparian sites [12]. However, several recent advances in this field stress that emphasis on the role of riparian and instream processes in the regulation of upslope diffuse pollution input has its limitations, especially in the context of climate change. Of particular note, we submit that a move from a fluvial hydrosystem to a river drainage
basin perspective is necessary to reframe research and thus provide a more integrated scientific understanding to inform water- and land-use management and policy.

**Major recent advances**

Several recent findings (highlighted below) suggest that, when addressing the impact of global change on the controls on diffuse pollution, it would be most pertinent to move away from a classical impact assessment of climatic change on riparian zones and adopt a broader spatio-temporal perspective. Adopting a drainage basin approach to understanding the consequences of climate change on water quality would allow the research community to address the problem of intrinsic limitation of nutrient removal in landscape structures; to tackle the impact of land-use change on river flow; and to grasp the consequences of the interdependency of element cycles and the cumulative effect of the long-term human impact on river systems.

**Intrinsic limitations of nitrogen removal in riparian zones and instream**

In a recent field study in riparian zones of small Dutch streams, Hefting et al. [13] found that nitrogen buffering capacity decreased with nitrate load but that the rate of emission of nitrous oxide, a potent greenhouse gas, increased dramatically with nitrogen load.

At the drainage basin level, Montreuil and Merot [14] quantified the nitrogen removal capacity of valley bottom wetlands in 18 agricultural drainage basins ranging from 3 to 30 km² in a temperate-oceanic climate. They determined that, on average, 11% of valley bottom wetlands within a drainage basin can reduce stream nitrate concentration by up to 30% at the outlet. They also estimated that a 5% increase in the area of the existing valley bottom wetlands can decrease nitrate concentration by another 30%. Their results emphasise the importance of riparian wetlands as a powerful tool to reduce diffuse nitrogen pollution but also show that riparian zones cannot buffer the entire diffuse pollution load. In this context, a recent report on 15N addition to 72 streams across multiple biomes and land uses in the US showed that both total uptake velocity and denitrification uptake velocity (which account for the nitrogen removal in rivers) decreased with nitrate concentration [15]. This suggests that instream processes cannot cope with excess nutrient input. In another independent study, Brookshire et al. [16] developed a mechanistic model of instream cycling and transport of nitrogen and phosphorus and tested it on more than 140 stream reaches. They found that small streams tend toward no net increase or retention of nutrients, with removal and storage balancing input. Therefore, they suggest that the chemistry of small streams represented an integrated measure of terrestrial nutrient losses.

**Impact of land use on river flow**

Alterations of the spatial and temporal distributions of river flows and groundwater recharge are determined by changes in temperature (evapotranspiration) and precipitation (water balance) but modified by river basin properties (including land use). Climate is the first-order driver of river flow regimes, with the basin being a second-order control [17]. Climate change impacts on hydrological processes may be identifiable already and further shifts in hydrological regime are predicted or anticipated [5]. Disaggregating climate from land-use controls on river flow is challenging, and although the matter is open to debate, the role of basin properties is considered insufficiently in many studies assessing river flow sensitivity to climate change. In an international water balance modelling study of 1,508 basins, long-term river flow was shown to be explained by land cover attributes [18]. Results indicate that land cover information makes a small but nonetheless significant contribution to model efficiency. However, research into 459 Austrian basins suggests that land use, soil type, and geology do not exert a major influence on runoff coefficients [19]. These different results can be reconciled if we consider that, with increasing spatial (basin area) and temporal (seasonal, annual, and beyond) scale, climate drivers override land-use influences on river flow [20].

**Impact of global change on carbon and nitrogen fluxes**

The recent widespread increase in concentration of dissolved organic carbon (DOC) in surface water in the Northern Hemisphere has been attributed to a consequence of a decline in the sulphate content of atmospheric deposition [21]. Using δ13C and δ14C analysis, Guo and Macdonald [22] demonstrate that, in large Arctic rivers, particulate organic carbon (POC) is derived from permafrost thawing and river bank erosion whereas DOC originates from modern terrestrial biomass. They infer that, during Arctic warming, an increase in DOC fluxes (caused by an increase in plant productivity following a shift from tundra to broadleaf plants) as well as an increase of POC fluxes (through melting of previously frozen soil horizons) can be expected. These results highlight the high interdependency between the carbon cycle and climate change, as well as the contribution of land-use change, as confirmed by Evans et al. [23] who demonstrated that relatively carbon-poor mineral soils under moderate to intensive grazing export DOC from older soil carbon. Similarly, there is clear evidence that vegetation and soils (and therefore land use) control the nutrient export from watersheds [24]. Landscape simplification and land-use intensification lead to diffuse
pollution, which is often more focused than generally realised [25] because certain parts of basins generate disproportionate pollution risks and because hydrological disconnection along flowpaths means that, even where risks are generated, they do not necessarily connect with the drainage network. The consequence of this is that riparian zones are often bypassed by, or disconnected from, diffuse nutrient input from upslope [26]. In turn, this necessitates considering the possible evolution of the landscape as a whole under global change, the riparian zone being one landscape element among others.

Interdependency of element cycles
In a recent study of dissolved inorganic nitrogen retention transport through a headwater basin, Triska et al. [27] demonstrated that biologically active carbon controlled nitrate removal during both hillslope and riparian zone transport. This carbon control on nitrate removal operated via its role as an electron donor for microbial denitrification. A recent synthesis indicated that, of the 1.9 Pg C per year delivered from land to river; half was consumed within the river systems before reaching the oceans [28]. This new evaluation stresses the importance of land-derived, biologically available carbon for heterotrophic microbial processes such as denitrification in river systems and the tight coupling of the nitrogen and carbon cycles [29].

Long-term legacy of anthropogenic impact
Walter and Merrits [30] demonstrated that recent historical human settlement has completely changed the geomorphology of a large part of the mid-Atlantic streams of the northeast coast of the USA but also the perception of what is, and was, a ‘natural’ stream. This result questions strongly the resilience capacity of streams and their riparian zones under climate change and the confidence to be given to restoration goals and the concept of reference sites in human-impacted areas.

Future directions
Assessing ecosystem sensitivity to hydrological change
Although aquatic ecosystem vulnerability to global change is recognised, knowledge of the most ecologically sensitive periods to hydrological events/extremes (for example, floods, droughts/low flows, and soil moisture deficit) and associated water stress and habitat disturbance [1] is limited. In addition to a focus on extremes, there is an evolving awareness of the importance of considering the spectrum of hydrological conditions experienced by habitats and their linkages to ecosystem structure and functioning. Long-term data sets have a crucial role to play in elucidating climate-hydrology-ecology links and setting short-term variability in a wider context (for example, [31]).

Uncertainty: reduction and management
Numerous authors have called for research to reduce uncertainty over (a) how climate change may affect freshwaters and (b) how water- and land-use managers should mitigate and adapt to climate change [32]. Uncertainties in predicting impacts may be attributed to limitation of historical data (in terms of duration, spatial coverage, homogeneity and so on) for model parameterisation, calibration, and validation; incomplete knowledge of complex process nonlinearity and feedbacks; general circulation model (GCM) scenarios; downscaling of GCM data to basin scale; and hydrological models [33,34]. Improved characterisation of model uncertainty is necessary to better inform risk management approaches and water- and land-use managers’ decision making [35].

New ways
Sutka et al. [36], using intramolecular distribution of nitrogen isotopes (isotopomer) in N2O, measured significant differences in 15N site preference of N2O between nitrification and denitrification. This approach has the potential to decipher the respective role of these two processes in emitting this greenhouse gas in different ecosystems and their in situ driving factors. Clément et al. [37] were able to measure iron-driven denitrification in riparian wetland, allowing oxidation of ammonia under anaerobic conditions and further denitrification of the nitrate produced by denitrification. If this new pathway is confirmed to be widely occurring, it challenges the currently accepted belief that denitrification in riparian zones is limited by nitrate production under anaerobic conditions or allochthonous input to anoxic hot spots. This would require reconsidering the current conceptual functioning of riparian buffer zones.

Abbreviations
DOC, dissolved organic carbon; GCM, general circulation model; POC, particulate organic carbon.

Competing interests
The authors declare that they have no competing interests.

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