1. Introduction

Nutrient and contaminant losses in agricultural landscapes are directly controlled by hydrological (flow pathways), chemical (sorption, speciation and transformations), biological processes (fixation, uptake) and indirectly by demographic (growing population), economic (food production) and societal drivers (individual attitudes, farming tradition) that control how agricultural landscapes are managed. Global change will likely increase nutrient and contaminant losses due to intensified hydro-meteorological drivers, higher nutrient inputs to meet growing food demand and continuous losses of nutrients from legacy stores in soils and aquifers. To offset the impact of global change and manage nutrients and contaminants in an efficient way, there is a growing need to mitigate their losses along the transfer continuum from agricultural land to aquatic ecosystems. To meet this challenge, we need a comprehensive understanding of how hydrological and chemical controls influence contaminants over a range of spatial and temporal scales, for different catchment typologies, and residence times, while also considering the role of changing climatic drivers, as well as economic and societal factors.

Our current understanding of nutrient and contaminant losses is framed by the pollutant transfer continuum [1]. This framework identifies critical pollutant loss pathways in space and time including sources, mobilisation, delivery and impact. Existing research rarely covers the full continuum, due to large distances and time lags between sources and delivery points in the stream network where we observe the impact. Current research mostly focuses on individual steps along the pollutant transfer, analysing their complexity and links with the immediate next steps in the continuum. At any step of the transfer continuum, an attenuation or transformation of pollutant delivery can occur, e.g., at the interface of land and water in the soil, groundwater, hyporheic and riparian zones [2]. This nutrient attenuation capacity in agricultural catchments can be further enhanced by implementing mitigation measures by farmers and land owners. The effect of mitigation measures on water quality is controlled by a number of environmental and societal factors, e.g., how awareness of pollution problems affects behavioural change and uptake of measures by stakeholders.

This special issue provides an overview of novel approaches to identify and understand the complexity of pollutant transport processes and suggest solutions how this evidence can inform
management decisions. We summarise these findings and provide a synthesis of future needs and directions in monitoring and managing of nutrient and contaminant losses in agricultural landscapes.

2. Overview of the Articles

High spatial and temporal resolution hydrochemical data are now more available thanks to advances in monitoring techniques [3], and used to elucidate controls on nutrient mobilisation and delivery in agricultural streams. Although hydrochemical data can show similar patterns, the underlying mechanisms can be different depending on catchment typology and specific land use and management. In their study, Vero et al. [4] showed that elevated stream phosphorus concentrations, typically observed in Irish streams during summer months, can result from a number of controls: transport through groundwater and lack of dilution in a well-drained grassland, in-stream mobilization in a well-drained arable catchment, and a combination of point sources and cumulative loading in a poorly drained grassland catchment. The authors point out that although phosphorus fluxes are generally low during summer baseflows, high concentrations pose a great risk to stream ecology. These pressures are likely to increase under future climate resulting in a worse ecological status in summer and a worse chemical status in winter.

High spatial and temporal monitoring is also needed to evaluate the effectiveness of mitigation measures. Johnes et al. [5] showed large complexity of multiple pathways and water sources feeding into a riparian wetland, generating large spatial and temporal variations in nutrient cycling and retention. They suggest a multiproxy approach based on the identification of the spatial distribution of functional units, using vegetation mapping, geophysical surveys, and isotopic ratios in soil water, sediments and vegetation, to estimate potential effectiveness of mitigation measures before they are implemented. This type of comprehensive monitoring can provide evidence for building and testing conceptual models of biogeochemical functioning of riparian wetlands and an important diagnostic tool for catchment managers.

Although critically needed to inform management decisions in undrained agricultural catchments, the identification of key delivery pathways for nutrients and other contaminants can be extremely complex. Adams et al. [6] demonstrated an approach in which high-frequency water quality data were used to calibrate an event-scale model of flow and P transport, the Catchment Runoff Attenuation Flux Tool (CRAFT). This approach, referred to by the authors as event forensics, provides detailed analysis of multiple flow and water quality parameters, along with their key pathways.

Artificial subsurface drainage in agricultural landscapes provides fast delivery pathways that can contribute large amounts of nutrients and sediments to streams. However, as shown by Klaiber et al. [7], despite higher total runoff, an artificially drained field contributed lower soluble reactive phosphorus and suspended sediment exports in a two-year study that compared two arable plots. Both drained and undrained plots contributed similar exports of total phosphorus and further studies from different soil, climatic and land use conditions are needed to corroborate these results. However, the study did not consider the effect of drainage on other pollutants, e.g., nitrate, which can be particularly important for achieving overall improvements in ecological status and avoiding pollution swapping effects [8].

Transport of contaminants, especially nutrients, along flow pathways is typically not conservative due to biogeochemical transformations that can change nutrient concentrations and fluxes. This makes establishing the causal links between flow pathways, source and delivery points and impact difficult and requires application of modelling studies or smart tracers. Severini et al. [9] tested a conservative tracer that helps to monitor vertical transport of nitrate from agricultural soils to groundwaters. The authors found that silica SiO$_2$ and nitrate NO$_3^-$ showed a similar spatial distribution in the subsurface suggesting their common origin from manure spreading. They also pointed out large spatial heterogeneity in redox conditions and distribution of microbial communities that can have critical impact on nitrate behaviour and concentrations. As many aquifers exceed nitrate concentration limits, we need better tracing techniques to resolve discrete flow pathways and the significance of biogeochemical heterogeneity at multiple scales.
Hydrological controls in agricultural catchments are also changing as shown by Uusheimo et al. [10]. The authors showed a significant increase in winter nutrient and sediment loads driven by increased flashiness. Small agricultural catchments are particularly sensitive to changes in hydrological forcing, with large consequences for water quality impairment. To offset these changes in hydrology, more intensive management efforts are needed, including implementation of mitigation measures that increase water residence time in the catchment e.g., retention ponds and constructed wetlands. Although the efficiency of these mitigation measures changes on a seasonal basis, their overall effect on water quality is positive and the authors encourage harnessing of all available retention potential in agricultural catchments.

Hydrological pathways and climatic variability are intrinsically linked. Uusheimo et al. [10], as well as Vero et al. [4], point to the sensitivity of nutrient outcomes to weather variability at the scale of individual rainfall events, up to seasonal variation in flow. These contributions add to our understanding of the role that climate variability has on the multitude of processes controlling nutrient export from agricultural catchments. Such knowledge lets us appreciate variability in outcomes despite mitigation efforts [8], as well as identify apparent trends that may be the result of longer-term weather variations (e.g., Mellander et al. [11]).

An important aspect in addressing diffuse pollution challenges in agricultural landscapes is our ability to understand and influence the attitudes of land owners and farmers, who may have the best understanding of pollution pressures in agricultural catchments. These stakeholders are key for the implementation of best management practices and mitigation measures to reduce nutrient and contaminant losses from land to water. Okumah et al. [12] systematically reviewed the evidence base of awareness–behavioural change–water quality improvement pathway, with a focus on Europe and North America. They found that although there is a generally a positive effect between awareness and behavioural change, meaning that well-informed farmers are more likely to introduce land management changes for water quality improvement, the link between awareness and water quality is typically more complex and less understood. This contrasting evidence for a relationship between uptake of mitigation measures and improvements in water quality results from a number of confounding factors, e.g., time lags, variation in climatic drivers and the level of uptake by farmers, making the effectiveness of mitigation efforts both pollutant, and measure dependent. This inherent complexity within the pollutant transfer continuum and the uptake of mitigation measures challenge our ability to design universally effective mitigation measures. A growing body of evidence shows that, while management practices help to decrease nutrient loading in agricultural catchments, their positive effects can be counterbalanced by increasing runoff attributed to changes in climatic drivers [8,11,13].

3. Novel Science Drives New Solutions to Future Catchment Monitoring and Management

Meeting future environmental, economic and societal needs required from agricultural landscapes, calls for new approaches in catchment monitoring and management as well as better communication among researchers, stakeholders and policy makers. The scientific papers in this special issue provide innovative solutions towards such new approaches.

To improve future monitoring, high spatial and temporal resolution measurements of stream water and sediments should be targeted to different catchment typologies, as similar hydrochemical patterns do not necessarily indicate the same underlying controls on nutrient and sediment losses [4]. Understanding of these controls is critical in designing appropriate mitigation measures and management practices that are likely to reduce sources and intercept pollution pathways. Johnes et al. [5] provided a tool for evaluation of potential impact of riparian wetlands that can be validated for other types of mitigation measures and catchment typologies. Minimum information requirement models, as in the study of Adams et al. [6], can be used to interpret water quality data and provide evidence needed for smart management of flow pathways. Decision makers can use this information to design appropriate mitigation measures along critical pollution pathways. However, as Severini et al. [9] point out, contaminant transport along flow pathways is rarely conservative. The challenge here is to be able
to identify critical sinks along the flow pathways and further enhance their function through mitigation options. For this, a combination of modelling studies and smart tracers is needed. This intensified management effort is critically needed with a changing climate, according to Uusheimo et al. [10]. Okumah et al. [12] stress that land owners and farmers are more likely to adopt mitigation practices if they receive a well-placed, consistent, and well-framed message on pollutant pressures, mitigation options and their expected outcome. This provides a challenge for researchers and policy makers to make this information transparent and evidence-based.

A common theme of this collection of papers is that of complexity, and how we unravel the important processes, mediating and moderating factors in both the social and biophysical spheres of complex catchment systems. While, over the past decade, high-resolution observational data have allowed us better insights into the ‘black boxes’ that may appear to represent complex catchment systems, such data is expensive to obtain at large scale [5] and may not even ‘readily indicate the type or location of sources or quantification of the pathways’ [4]. Hence, several papers in this special issue are proposing lower-cost solutions that could be used to answer outstanding knowledge gaps at larger geographical scales. Application of conservative tracers [6,9], and stable isotopes [5], to understand pollutant sources and transformations is proving particularly promising in this respect. In combination with additional techniques, including geophysical and vegetation surveys [5] and synoptic geochemical sampling at lower temporal resolution [4,5], these approaches may offer practical tools to inform the spatial targeting and understand the likely effectiveness of mitigation measures. However, major knowledge gaps still remain both in the physical domain along the source–mobilisation–delivery–impact continuum, and in the social domain along the awareness–behavioural change–water quality improvement continuum. Anticipating catchment responses to common drivers remains difficult, as, whilst the symptoms may appear similar, the underlying causes often differ and are context specific [4,6]. Whilst this complexity may appear frustrating to both scientists and catchment managers, it also presents opportunities for the development of novel research tools and transdisciplinary frameworks to gain fresh insights into catchment controls of nutrient and contaminant losses.

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References
1. Haygarth, P.M.; Condron, L.M.; Heathwaite, A.L.; Turner, B.L.; Harris, G.P. The phosphorus transfer continuum: Linking source to impact with an interdisciplinary and multi-scaled approach. Sci. Total Environ. 2005, 344, 5–14. [CrossRef]
2. Wollheim, W.M.; Bernal, S.; Burns, D.A.; Czuba, J.A.; Driscoll, C.T.; Hansen, A.T.; Hensley, R.T.; Hosen, J.D.; Inamdar, S.; Kaushal, S.S.; et al. River network saturation concept: Factors influencing the balance of biogeochemical supply and demand of river networks. Biogeochemistry 2018, 141, 503–521. [CrossRef]
3. Rode, M.; Wade, A.J.; Cohen, M.J.; Hensley, R.T.; Bowes, M.J.; Kirchner, J.W.; Arhonditsis, G.B.; Jordan, P.; Kronvang, B.; Halliday, S.J.; et al. Sensors in the Stream: The High-Frequency Wave of the Present. Environ. Sci. Technol. 2016, 50, 10297–10307. [CrossRef]
4. Vero, S.E.; Daly, K.; McDonald, N.T.; Leach, S.; Sherriff, S.C.; Mellander, P.-E. Sources and Mechanisms of Low-Flow River Phosphorus Elevations: A Repeated Synoptic Survey Approach. Water 2019, 11, 1497. [CrossRef]
5. Johnes, P.; Gooddy, D.; Heaton, T.; Binley, A.; Kennedy, M.; Shand, P.; Prior, H. Determining the Impact of Riparian Wetlands on Nutrient Cycling, Storage and Export in Permeable Agricultural Catchments. Water 2020, 12, 167. [CrossRef]
6. Adams, R.; Quinn, P.; Barber, N.; Burke, S. Identifying Flow Pathways for Phosphorus Transport Using Observed Event Forensics and the CRAFT (Catchment Runoff Attenuation Flux Tool). Water 2020, 12, 1081. [CrossRef]
7. Klaiber, L.B.; Kramer, S.R.; Young, E.O. Impacts of Tile Drainage on Phosphorus Losses from Edge-of-Field Plots in the Lake Champlain Basin of New York. *Water* 2020, 12, 328. [CrossRef]

8. Bieroza, M.; Bergström, L.; Ulén, B.; Djodjic, F.; Tonderski, K.; Heeb, A.; Svensson, J.; Malgeryd, J. Hydrologic Extremes and Legacy Sources Can Override Efforts to Mitigate Nutrient and Sediment Losses at the Catchment Scale. *J. Environ. Qual.* 2019, 48, 1314. [CrossRef]

9. Severini, E.; Bartoli, M.; Pinardi, M.; Celico, F. Reactive Silica Traces Manure Spreading in Alluvial Aquifers Affected by Nitrate Contamination: A Case Study in a High Plain of Northern Italy. *Water* 2020, 12, 2511. [CrossRef]

10. Uusheimo, S.; Tulonen, T.; Huotari, J.; Arvola, L. Long-Term (2001–2020) Nutrient Transport from a Small Boreal Agricultural Watershed: Hydrological Control and Potential of Retention Ponds. *Water* 2020, 12, 2731. [CrossRef]

11. Mellander, P.E.; Jordan, P.; Bechmann, M.; Fovet, O.; Shore, M.M.; McDonald, N.T.; Gascuel-Odoux, C. Integrated climate-chemical indicators of diffuse pollution from land to water. *Sci. Rep.* 2018, 8, 944. [CrossRef]

12. Okumah, M.; Chapman, P.; Martin-Ortega, J.; Novo, P. Mitigating Agricultural Diffuse Pollution: Uncovering the Evidence Base of the Awareness–Behaviour–Water Quality Pathway. *Water* 2018, 11, 29. [CrossRef]

13. Ockenden, M.C.; Hollaway, M.J.; Beven, K.J.; Collins, A.L.; Evans, R.; Falloon, P.D.; Forber, K.J.; Hiscock, K.M.; Kahana, R.; Maceold, C.J.A.; et al. Major agricultural changes required to mitigate phosphorus losses under climate change. *Nat. Commun.* 2017, 8, 161. [CrossRef]

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