



Challenges of Reducing Phosphorus Based Water Eutrophication in the Agricultural Landscapes of Northwest Europe

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In this paper, we outline several recent insights for the priorities and challenges for future research for reducing phosphorus (P) based water eutrophication in the agricultural landscapes of Northwest Europe. We highlight that new research efforts best be focused on headwater catchments as they are a key influence on the initial chemistry of the larger river catchments, and here many management interventions are most effectively made. We emphasize the lack of understanding on how climate change will impact on P losses from agricultural landscapes. Particularly, the capability to disentangle current and future trends in P fluxes, due to climate change itself, from climate driven changes in agricultural management practices and P inputs. Knowing that, future climatic change trajectories for Western Europe will accelerate the release of the most bioavailable soil P. We stress the ambiguities created by the large varieties of sources and storage/transfer processes involved in P emissions in landscapes and the need to develop specific data treatment methods or tracers able to circumvent them, thereby helping catchment managers to identify the ultimate P sources that most contribute to diffuse P emissions. We point out that soil and aqueous P exist not only in various chemical forms, but also in range of less considered physical forms e.g., dissolved, nanoparticulate, colloidal and other particulates, all affected differently by climate as well as other environmental factors, and require bespoke mitigation measures. We support increased high resolution monitoring of headwater catchments, to not only help verify the effectiveness of catchments mitigation strategies, but also add data to further develop new water quality models (e.g., those include Fe-P interactions) which can deal with climate and land use change effects within an uncertainty framework. We finally conclude that there is a crucial need for more integrative research efforts to deal with our incomplete understanding

of the mechanisms and processes associated with the identification of critical source areas, P mobilization, delivery and biogeochemical processing, as otherwise even high-intensity and high-resolution research efforts will only reveal an incomplete picture of the full global impact of the terrestrial derived P on downstream aquatic and marine ecosystems.

Keywords: phosphorus, cycling, soil, eutrophication, climate change, colloidal and particulate, water quality

INTRODUCTION

The increasing world-wide issue of the eutrophication of our lakes, reservoirs, rivers and coastal waters has highlighted an urgent need for interdisciplinary action across research fields (e.g., Moss, 2012; Withers et al., 2014; Elmgren et al., 2015; Pinay et al., 2017; Charlton et al., 2018). It is now well documented that anthropogenically derived phosphorus (P) and nitrogen (N) pollution are currently the main drivers of eutrophication, with the excessive inputs of these two nutrients into freshwater and estuarine water bodies being considered as one of the most urgent environmental issues that human societies face (Rockström et al., 2009; Steffen et al., 2015; George et al., 2017). Agricultural activities and urbanization can both deliver excess P and N to aquatic ecosystems, which may cause eutrophication of water courses and which in turn may alter the native ecological communities, degrade ecosystem services and directly or indirectly impact water supply, recreational uses, and human health (e.g., cyanobacteria blooms). Already, in the late 1960s a fundamental change started to occur in most developed countries regarding the type of P input (see **Table S1** for P terminology and additionally Haygarth and Sharpley, 2000), namely a very marked decrease of point-source P due to an increased waste water treatment (Persson, 2001; Billen et al., 2007; Grizzetti et al., 2012; Scavia et al., 2014; Minaudo et al., 2015). This decrease has had rapid and marked effects on the P loading and trophic status of downstream water bodies (e.g., a 75% reduction in 25 years of the P flux in the Seine River and Lake Geneva; disappearance of cyanobacteria blooms in some lakes, such as Lake Erie in the USA, Lake du Bourget in France, and Lake Mjøsa in Norway; Nesheim et al., 2010; Jacquet et al., 2014; Scavia et al., 2014; Romero et al., 2016). However, this reduction in point sources has been counterbalanced by the stagnation or even the increase of diffuse P emissions from agricultural soils (Scavia et al., 2014; Dupas et al., 2015d; Stoddard et al., 2016). This has happened even despite the fact that overall fertilizer usage in NW Europe since the 1980's has decreased significantly (Schoumans et al., 2015). For example in Sweden, lower P fertilizer use is now accompanied by more or less balanced P inputs and outputs in agriculture (Bergström et al., 2015). Both dissolved/colloidal P, i.e., the most bio-available forms of P for algae (Dupas et al., 2015a; Mellander et al., 2016; Gu et al., 2017), as well as particulate P (Bechmann and Deelstra, 2013) are involved in this increase of P emissions from agricultural soils. For Norway increased delivery of particulate P to surface waters can partly be assigned to higher soil erosion due to climate change effects (i.e., increased runoff) (Deelstra et al., 2011). Whereas,

for Sweden a modeling study (Arheimer and Donnelly, 2013) suggested that the total mean load to the Baltic Sea will increase for P, but may decrease for N by 2100 due to climate-induced changes. Enhanced particulate P can be formed in the surface waters due to the discharge of iron (Fe) and calcium (Ca)-bearing groundwater and subsequent precipitation of P-rich minerals (Baken et al., 2013; Van der Grift et al., 2014, 2018). Depending on the receiving water body, most of this particulate P could in the long run become available to algae (e.g., Yang et al., 2016; Yao et al., 2017).

Maintaining high levels or even increasing diffuse P losses from agricultural landscapes hampers any expected improvements in water quality, thereby challenging our society's ability to combat eutrophication. The issue here concerns not only lakes, rivers and estuaries, but also the multitude of small water bodies of natural and artificial origin, some which may serve as reservoirs for drinking water production.

For 30 years or more, much knowledge has been gained on the basic physico-chemical processes by which P is mobilized in soils and transported to rivers, as well as on the chemical forms of the mobilized and transported P. For example, the role of wetting-drying cycles as catalysts for the production and transfer of dissolved and colloidal phosphorus is now well known (e.g., Turner et al., 2003; Butterly et al., 2011; Blackwell et al., 2013; Chen et al., 2016; Gu et al., 2018). In particular, the important role of colloids to serve as carrier of P in soils and waters has been highlighted (e.g., Henderson et al., 2012; Gottselig et al., 2014; Liu et al., 2014; Baken et al., 2016; Jiang et al., 2017; Gu et al., 2018; Missong et al., 2018). Also well-constrained is the capacity of P to bind with manganese (Mn) and iron oxides and the influence the redox state of soils and waters has on P mobility in the environment (Scalenghe et al., 2012; Van der Grift et al., 2014; Jiang et al., 2015a,b; Smolders et al., 2017). Along with these understandings, novel high resolution P water quality monitoring, such as sensor technology, can be used to understand the processes of dissolved and particle-bound substances in waters. For example by using turbidity as a proxy for particulate P we can now reliably achieve high-frequency observations of P losses from agricultural landscapes over multi-annual periods (Jordan et al., 2007; Skarbøvik and Roseth, 2014; Rode et al., 2016; Shore et al., 2017). This information, combined with ever advancing analytical techniques (Kruse et al., 2015) as well as improvements in landscape visualization tool kits, is helping to facilitate efforts to better quantify P dynamics in time and space, from plot to catchment and landscape.

Transforming this knowledge into appropriate cost effective mitigation strategies remains a major challenge (Schoumans

et al., 2014, 2015; Dodds and Sharpley, 2015; Kleinman et al., 2015; Sharpley et al., 2015; Withers et al., 2015a,b; Rowe et al., 2016). The degree of integration and knowledge is such today that some of the most recent studies can go as far as integrating the costs of management in the reduction of diffuse phosphorus emissions, in connection with national or local legislation or land property issues (e.g., McDowell et al., 2016; Vinten et al., 2017; Zhang et al., 2017).

The EU Water Framework Directive (WFD) has since its emergence in the beginning of this millennium set the scene for water management in Europe. Its implementation has resulted in massive efforts to classify all water bodies according to their ecological status, within this context monitoring of European waters has, at least tentatively, become more harmonized through the Common Implementation Strategy (CIS) guidelines. Achieving Good Ecological Status of waterbodies across Europe under the Water Framework Directive will require an improved understanding of the link between P concentrations, loads and sources in the soil and recipient freshwater ecosystems within the context of multiple stressors (Whitehead and Crossman, 2012; Crossman et al., 2013). However, in this paper, we will not focus on all the aspects of the WFD or other EU legislation, but only few aspects that are linked to the main foci of our review. These include the types of monitoring that can give new insight into catchment processes, including investigative monitoring and source apportionment.

Furthermore, we are taking into account the considerable amount of papers already published on the topic of P transfer and P management in agricultural landscapes, including several recent review papers (e.g., Kleinman et al., 2011; Chowdhury et al., 2014; Schoumans et al., 2014; Sharpley et al., 2015; Kadlec, 2016; McDowell et al., 2016). As well as related contemporary topical reviews on aspects like scenario analysis and mitigation (Roberts et al., 2012; Schoumans et al., 2014; Dodd and Sharpley, 2016; Ahmad et al., 2017; Liu et al., 2017; Wu et al., 2017), policy and implementation (Christen and Dalgaard, 2013; McDowell et al., 2016), novel modeling approaches and management insights (Shepherd et al., 2011; Radcliffe et al., 2015; Xie et al., 2015; Ouyang et al., 2017, water quality modeling (Rode et al., 2010; Wellen et al., 2015; Hashemi et al., 2016), erosion (Panagos et al., 2017) and other more general appraisal of future needs and directions (Fernandez-Mena et al., 2016; Garnache et al., 2016). Hence our intention in this article is not to produce an exhaustive review which also covers the state of knowledge in these related research fields. Rather, our paper is aimed at highlighting certain important knowledge gaps and research challenges on P cycling and transfer processes in agricultural landscapes in Northwest Europe in relation to the continued issue of eutrophication of inland and coastal surface waters. Thereby, implicitly also identifying research needs for these 4 key challenges. These main challenges for us concern: (i) a better understanding of the processes and variables controlling P mobilization and P transfer in headwater catchments, (ii) a better consideration of how the basic mechanisms involved in P mobilization in soils at the small scale aggregate themselves at larger scales, and how this aggregation ultimately control P diffuse emissions in agricultural landscapes, (iii) the resolution of ambiguities in the

interpretation of river P dynamics for identification of sources in catchments, and (iv) a better understanding and prediction of the effects of climate change on P fluxes and P physical-chemical forms.

HEADWATER CATCHMENTS: THE RIGHT PLACE TO BE TO MANAGE AND MONITOR DIFFUSE PHOSPHORUS EMISSIONS

Combating eutrophication relies on effective reduction in P losses from land to water through appropriate management of P sources and pathways in the landscape. The key scientific and management questions are: where is the excess P coming from? How are these P sources mobilized and delivered to surface waters? What is their impact on water bodies? For still unknown reasons, some headwater catchments (surface area <50 km²) in agricultural landscapes appear more resilient (have more buffering capacity) than others with regards to P loading, resulting in huge differences in average annual P concentrations and annual P fluxes, as exemplified by the case of Brittany, one of the most intensively farmed regions of France (Legeay et al., 2015; Abbott et al., 2017). This increased variability of P concentration and P fluxes with decreasing catchment size is as a result of headwater catchments having lower intense cultivation rates. In fact the majority of these Brittany headwater catchments shown in **Figure 1** are intensively cultivated (>80% of arable land) throughout (Legeay et al., 2015; Abbott et al., 2017). They were also found to be insensitive to the general decreased particulate P emissions that has been recorded in Brittany since the beginning of the 1990's, as this decrease left the amplitude of particulate P flux variability in headwater catchments unchanged (see **Figure 1**).

The increased variability of P emissions when moving upstream in headwater catchments is found both for dissolved and particulate P fluxes (**Figure 1**). Land use, and especially the share of arable land within a catchment, is known to have a strong positive relationship on nutrient losses (Evans et al., 2014). Indeed, plotting median TP (total P) losses against the share of arable land in Swedish catchments included in the monitoring program River outlets and Trend Watercourses results in strong ($R^2 = 0.79$, $p < 0.005$) and positive relationship (**Figure 2**). However, long term data from another Swedish water quality program, monitoring program arable catchments, also highlighted an inherent great variability in nutrient delivery among small catchments (<50 km²) with high portion of the arable land (**Figure 2**). For these small catchments with high share of arable land, the relationship was not statistically significant at all ($R^2 = 0.02$, $p = 0.41$). Consequently, measured P concentrations from some small catchments were remarkably low in spite of intensive agricultural production, whereas other catchments were found to be less resilient and much more vulnerable to P losses.

The high spatial variability observed among headwater catchments appears persistent over time suggesting that occasional synoptic sampling of headwater catchments can provide valuable information for catchment characterization and

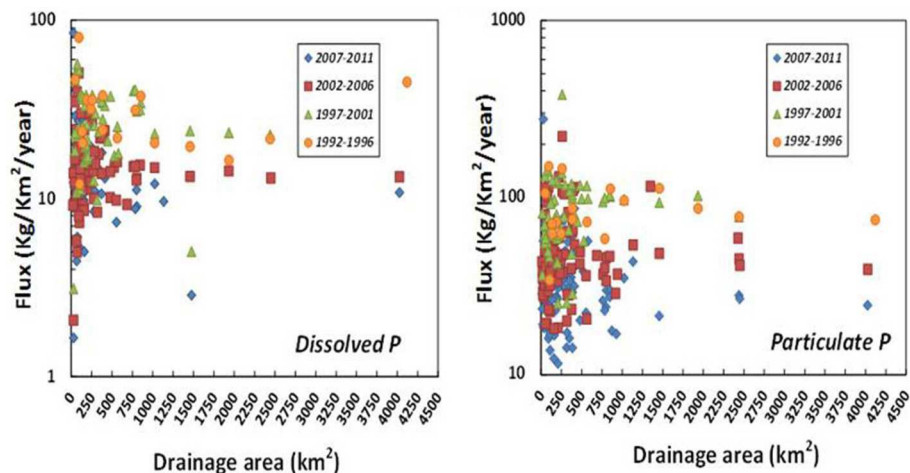


FIGURE 1 | The relationship between dissolved and particulate P fluxes and drainage areas as observed in Brittany rivers, Western France. Fluxes have been calculated as average values for periods of four consecutive years, in order to minimize flux uncertainties. The two plots data from 117 monitoring stations. Drainage areas range from 11 to 4,000 km². For both dissolved and particulate P fluxes, an increase in variability is observed when moving from large rivers toward headwater catchments, the maximum of variability occurring for headwater catchments of drainage area <50 km² (Legeay et al., 2015).

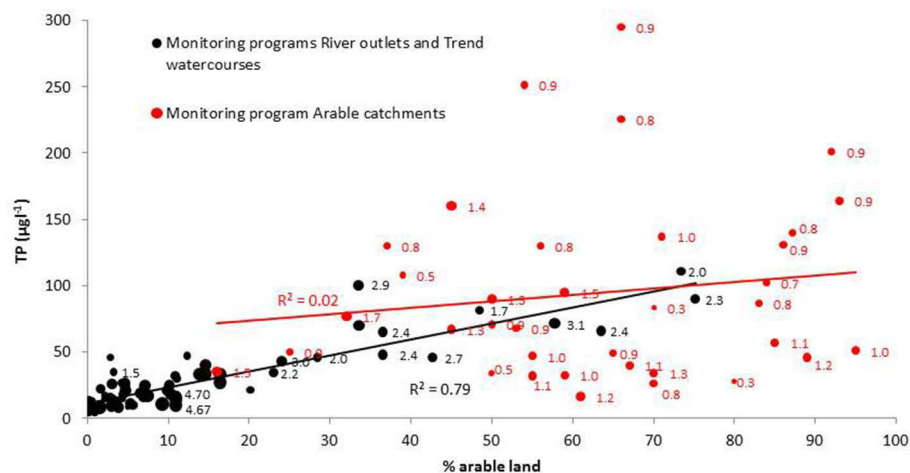


FIGURE 2 | The relationship between the portion of a arable land and median total phosphorus (TP) concentration for the period 2000–2016 included in two Swedish water equality monitoring programs: (a) 77 catchments (area 8–50,110 km²). River outlets and trend watercourses (black filled circles) and (b) 36 smaller (area 1.8–54 km²) catchments dominated by agriculture (red filled circles). The size of the filled circles is proportional to the catchment area, and the label value stands for the log₁₀ area of the catchment (km²).

Original source data:

Monitoring programs River outlets and Trend Watercourses: <http://miljodata.slu.se/>

Monitoring programs Arable catchments: <http://jordbruksvatten.slu.se/>.

management with regards to P emissions. This finding also raises the question of the spatial threshold at which the landscape splits into poorly and highly contributing headwater catchments with regards to P emissions, which has been recently referred to as the “landscape grain size” concept (Abbott et al., 2017). There are many potential parameters that could modify the capacity of headwater catchments to release P to river networks, and thus create variations in P emission properties at a specific spatial threshold, or given landscape grain size (see also Dodds and

Oakes, 2008; Haygarth et al., 2012). Included are the extent of preferential flow paths in soils and aquifers, which determine residence times in different catchment components, as well as the connectivity between land and water which may strongly vary among headwater catchments (Dahlke et al., 2012; Dupas et al., 2015d, 2016; Mellander et al., 2015). Differences in P applied to or present in soils, the biogeochemical cycling in soils, linked with variations in groundwater dynamics, themselves influenced topography, could be also involved (e.g., Gu et al.,

2017). Differences in past land-use history and soil properties will likewise play a role (Stutter et al., 2015; Gu et al., 2017).

Irrespective of the processes and factors that control the ability of headwater catchments to act as sinks or sources for P, the understanding where P is coming from is important both to target and reduce these sources and also to establish who is responsible for their management according to the polluter pays principle (as defined in the WFD). There is however less clarity in WFD who will pay for the monitoring to obtain this information, in fact, the direct monitoring of point sources is often left to the owner of the waste water treatment plants (Skarbovik et al., 2014).

Knowing the spatial structure and the typical grain size of source and sink headwater catchments with regards to diffuse P emissions in the landscape could improve site selection for targeted management efforts. Should the difference in resilience properties be of high spatial and temporal stability as found by Legeay et al. (2015) and Abbott et al. (2017), then the intervention in those headwater catchments showing the lowest resilience or highest source properties would potentially yield the largest catchment-level improvements at the lowest cost.

Knowing the landscape typical threshold or grain size could be also helpful for rationalizing water quality monitoring networks. Owing to the temporal stationarity of the spatial distribution of poorly and highly contributing headwater catchments, occasional synoptic sampling of headwater catchments would provide valuable information for identifying high and low contributing headwater catchments in agricultural landscapes. The redistribution of agricultural activity based on the difference in P emission properties of headwater catchments could be a cost effective management strategy for decreasing P loads of larger rivers. This could be implemented even in the absence of a clear understanding of the underlying mechanisms and factors that cause differences in headwater catchment properties. The implementation over the long-term of high frequency monitoring devices at the outlet of larger river basins where the river channel integrates multiple headwater catchments would make it possible to assess the effectiveness of the mitigation strategies thus deployed in the most contributive headwater catchment areas.

DIFFICULTIES IN DISENTANGLING POINT-SOURCE, DIFFUSE PHOSPHORUS EMISSIONS AND PRE-AGRICULTURAL PHOSPHORUS CONCENTRATION BASELINES

Load apportionment models are often used to distinguish between diffuse (mainly from agriculture) and point P sources (sewage treatments, industry sources, fish farming etc.) (Bowes et al., 2008). In many European countries, monitoring networks (statutory monitoring) were established well before 2000 to monitor pollution from point sources and to assess long-term water quality status (Bieroza et al., 2014; Kyllmar et al., 2014), but recently more new emphasis has been put on such networks as part of the obligations and the verification of compliance

with EU legislative directives (e.g., WFD). Furthermore, both the location of the sampling points and the monitoring interval are not always suitable to capture the spatial and temporal dynamics of diffuse sources (Jordan and Cassidy, 2011; Bieroza et al., 2014). Thus, targeting diffuse P is difficult, since the sources operate sparsely in both space and time, except for specific locations in the catchment, so-called Critical Source Areas (CSA), and during specific times e.g., rainfall and snowmelt events (Djodjic and Villa, 2015).

Haygarth and Jarvis (1999) stated that having good quantitative knowledge of the baseline P concentrations is necessary when assessing spatiotemporal trends in water quality in agricultural areas. This is also a requirement in the WFD, which states that the environmental goals should deviate only slightly from the background or reference conditions. However, the baseline P concentrations can be expected to vary widely among regions. For example, Phillips and Pitt (2015) demonstrated the differences between European countries of nutrient boundaries used for the WFD. This may be due to natural variations in soil types and topography, but may also be a result of limitations in catchment understanding.

At any rate, such baseline levels should be based on scientific methods, and not on political concerns. Estimations of baseline P is especially difficult in the lower lying areas where the land has been used for agricultural production for centuries, and where such baseline P concentrations must either be modeled or estimated based on limited data. Additionally, those P sources originating from forests and highlands areas ("background runoff"), which are generally poorly quantified themselves, do also contribute to overall baseline P concentrations in the waters of agricultural and urban catchments. Furthermore, the baseline P values may also change, due to large-scale changes such as climate change or the reductions in acid rain. The latter has led to brownification (increased darkening of the water color, due to the increase of dissolved organic carbon content) of Scandinavian waters, which again may have impacted on P losses (Fölster et al., 2014).

CHALLENGES TO MONITOR AND MODEL PHOSPHORUS TRANSFER AT THE LANDSCAPE SCALE

Monitoring and modeling of P transport pathways in the landscape remains a challenge. This is particularly true for the transfer of particulate P, which is a highly dynamic process, and targeting particulate P sources depends on the exact timing of mitigation measures in relation to plant growth, soil management and soil moisture. Modeling approaches based on detailed measurements of topography (e.g., Lidar) are normally used to target both sources and pathways (e.g., erosion gullies) of sediment-bound P (Thomas et al., 2016). On tile drained soils, significant amounts of particulate P can also be transferred through macropores, which are difficult to monitor and manage (Bechmann et al., 2017a). In addition, elucidating the sources and pathways of dissolved P is also a challenge since it involves understanding of sub-surface pathways (e.g., shallow

groundwater) and their connectivity with water bodies as well as its interactions with soil chemistry governing P sorption and release. Shallow sub-surface pathways and hyporheic flow have been found to be important for dissolved P delivery to streams (Bieroza and Heathwaite, 2015; Mellander et al., 2015). Recent advances in high-temporal resolution sampling with *in situ* analyzers and sensors have yielded information to help delineate potential pathways through hysteresis pattern analysis (Bieroza and Heathwaite, 2015; Dupas et al., 2015b). Furthermore, establishing robust correlations between routinely measured parameters (e.g., turbidity) or smart environmental tracers and P fractions can further help to gain improved understanding of processes and assist to reduce the monitoring cost (Bieroza and Heathwaite, 2016; Minaudo et al., 2017; Stutter et al., 2017).

Statutory monitoring data are used to assess chemical and ecological status of water bodies and to evaluate the impacts of diffuse pollution on stream nutrient concentrations. Despite intensive efforts to reduce these negative impacts, many rural catchments still show increasing long-term P concentration trends (Bechmann et al., 2017b), possibly due to the influence of weather events on local landscape P dynamics, highlighting the continued need for a deeper understanding of these weather-related processes (Bieroza et al., 2014). Seemingly similar catchments (e.g., percentage arable land in a catchment) are known to show opposing nutrient trends, which in turn may indicate a differing resilience to environmental change (Legeay et al., 2015). The observed disconnection between management practices and water quality in many catchments necessitates further efforts to better target P sources and pathways in space and time. The persistence of P pollution also requires more catchment-tailored approaches to mitigate diffuse pollution. Use of high-resolution mapping of CSAs, including soil chemistry, topography and soil type, can help to identify locations in the landscape that will benefit most from the implementation of mitigation measures (Thomas et al., 2016; Djodjic et al., 2017). Using statutory and high-frequency P monitoring in parallel could further help in identifying the most critical time periods and pathways of diffuse pollution (Dupas et al., 2015c). Additionally, new upscaling modeling approaches are needed to link single location patterns to landscape-wide process understanding.

AMBIGUITIES IN THE INTERPRETATION OF RIVER P DYNAMICS FOR IDENTIFICATION OF SOURCES IN CATCHMENTS

In order to limit P transfer and reduce eutrophication, it is important to identify the sources and the mobilization/delivery mechanisms of P into rivers. One commonly used method for source identification is statistical analysis of water quality time series. In particular, load apportionment models (LAMs) based on concentration - discharge (C-Q) relationships (Bowes et al., 2008, 2014, 2015; Greene et al., 2011; Lamba et al., 2015; Crockford et al., 2017; Glendell et al., 2018) or the identification

of periods of the year when one source is believed to dominate over other sources, a method is used to disentangle point sources from diffuse sources (Legeay et al., 2015). Equations in LAMs all rely on the assumption that point source emissions are constant in time (leading to negative C-Q relationships due to dilution when discharge increases) while diffuse source emissions increase with discharge (leading to positive C-Q relationships due to mobilization and delivery of P during storm events). Furthermore, analysis of C-Q hysteresis loops during storm events has been used to distinguish proximal (remobilization of stream bed sediments, bank erosion, erosion of riparian area) and distal sources (hillslope erosion, subsurface transfer) (Stutter et al., 2008; Outram et al., 2014, 2016; Bieroza and Heathwaite, 2015; Bowes et al., 2015; Dupas et al., 2015a,b; Perks et al., 2015; Sherriff et al., 2016). Clockwise loops are often interpreted as originating from proximal sources whereas anticlockwise loops are interpreted as originating from distal sources. However the interpretation of P dynamics observed in rivers, and both LAMs and analysis of C-Q hysteresis loops are subject to “ambiguity” problems. Here, the term ambiguity describes situations when several processes (or sources) can lead to the same observed P dynamics in rivers. This can lead to difficulties in inferring the dominant controlling process (or source) from statistical analysis of water quality time series. Jarvie et al. (2012) have shown that remobilization of streambed sediment during winter high flow is often (wrongly) attributed to diffuse source, whereas these sediments may have been enriched by point sources during the summer low flow period, and thus the primary source is predominantly a point source. This ambiguity may lead to overestimation of diffuse sources in LAMs.

The reductive dissolution of Fe oxyhydroxides in sediments of lowland rivers (Smolders et al., 2017) or riparian wetlands in upland systems (Dupas et al., 2017) can lead to soluble reactive phosphorus (SRP) release during summer low flows, increasing summer SRP concentrations in rivers in the same manner as does an undiluted point source. This ambiguity may lead to overestimation of point sources in LAMs. Likewise, ambiguities in river P dynamics have been highlighted in the identification of proximal versus distal sources within hillslopes. Bieroza and Heathwaite (2015) have shown that in the case of two successive storm events, the first event could transport a distal P source to the near-stream zone, which can be remobilized during a second event (exhibiting a clockwise hysteresis loop). Therefore, if only the second event was monitored, there is an ambiguity in the location (proximal/distal) of the primary source, because the primary mobilization of a proximal source can lead to the same observed P dynamics pattern as the secondary mobilization of a distal source (both would exhibit clockwise hysteresis loops). Furthermore, *in-situ* monitoring of uncultivated soils in the near stream zone have highlighted that these zones represented a significant contribution to annual SRP exports in a temperate agricultural catchment (Gu et al., 2017). However, it is not clear to which extent these zones are primary sources (due to legacy P inputs before their conversion into uncultivated buffers) or secondary sources (as they accumulate and re mobilize P rich eroded soils from cultivated upslope areas). At the scale of the hyporheic zone, Van der Grift et al. (2014, 2018) have shown that

SRP via Fe(II) oxidation at the groundwater-surface water interface can be followed by remobilization of newly formed particulate P. Thus, there can be an ambiguity in the location (proximal/distal) of the source of the particulate P, which can be attributed to a proximal source (the river bank) whereas it may stem from remobilization of a more distal primary source. Another example of ambiguity is the P release pulses observed in summer from river sediments whose ultimate source can be a mixture of several point and diffuse sources, located further upstream (Cooper et al., 2015a; Dupas et al., 2017; Smolders et al., 2017).

Given the important implications of these ambiguities in the identification of P sources and pathways in rural landscapes, several improved methods can be proposed. Firstly, it is possible to use tracers (e.g., stable isotopes and plant-specific biomarkers) and/or complementary water quality parameters in addition to P (Jarvie et al., 2012; Cooper et al., 2015b; Alewell et al., 2016; Glendell et al., 2018) to trace the primary sediment source. For example, Ahlgren et al., 2012 found Barium (Ba) being a promising tracer element, being present in significantly higher amounts in waters affected by agricultural runoff. Whereas Paruch and co-authors developed microbial source-tracking techniques based on advanced DNA methods to distinguish between different sources of microbial contamination (Paruch et al., 2015; Paruch and Paruch, 2017). Furthermore, recent work do suggest that the composition of P carrier colloids could also be a 'tracer/fingerprint' the source of P (Missong et al., 2018). Secondly, more observational data is needed, combining monitoring at the catchment outlet with *in situ* observation within functional zones such as riparian wetland or groundwater (Mellander et al., 2016; Gu et al., 2017) to disentangle primary mobilization from secondary delivery/remobilization. Thirdly, interpretation of long term change in water quality dynamics ("trajectory") in catchments where management changes, such as removal of point sources, decrease in soil P status or installation of buffer zones, have been recorded over several decades (Bieroza et al., 2014) may help to resolve ambiguities in river P dynamics. Therefore, meta-analysis of large catchment datasets may help to identify typical situations where one mechanism (e.g., summer sediment P release versus point source P input) dominates over others. In doing so, "typical" situations can be identified without ambiguity and the P dynamics pattern can be transferred to other, less studied areas as a guide to infer what are the key mechanisms controlling P loads. Finally, consideration of time lags and legacy effects is crucial when communicating results of source identification studies to catchment managers, so that they do not only target secondary proximal sources (short term vision) but also primary more distal sources (long term vision).

DIFFICULTIES IN PREDICTIONS OF DISSOLVED/COLLOIDAL/PARTICULATE CATCHMENT PHOSPHORUS "HOT SPOTS" AND "HOT/CRITICAL" MOMENTS

The difficulty in attributing spatio-temporal variations in P fluxes and P concentrations in streams and rivers draining agricultural landscapes to specific sources or specific mobilization/transfer

processes is well known (e.g., Haygarth et al., 2005; Edwards and Withers, 2007; Granger et al., 2010; Trevisan et al., 2012; Hahn et al., 2013; Sharpley et al., 2015). This difficulty mainly arises from the fact that the transfer of P in agricultural landscapes is not often caused by a single process or a single source, rather being the consequence of a series of processes, mobilizing P from several permanent or temporary sources. A good illustration of this difficulty and of the ambiguities it may create in terms of process and source identification was provided by the dissolved/colloidal P release pulses observed in riparian wetland zones. Riparian zones in agricultural catchments are active in transforming poorly mobile particulate P into highly mobile dissolved and colloidal P, which is directly or indirectly bioavailable (Stutter et al., 2009; Dupas et al., 2015a; Gu et al., 2017). Indeed, these P release pulses come from the solubilisation of particulate P eroded from the upland cultivated fields which are temporarily accumulated in wetland zones. This implies that the primary sources of the released P are the upland cultivated fields from which P originates. Quite clearly, such entanglement of processes and existence of temporary sources may lead to ambiguities in the clear identification of the zones and processes contributing most to the risk of P mobilization and P transfer at the catchment scale. Therefore, moving toward full understanding the overall sequence of transfer and retainment of P process chain and how P release capacities vary in space and time is challenging, but it is a knowledge prerequisite to better control and reduce P loss in agricultural landscapes.

In order to better evaluate and quantify how the above mentioned processes control P emissions in catchments, we need to understand how their spatio-temporal occurrence and intensity is controlled by landscape properties, and how the zones where these processes take place are connected with each other and to the river network. Though relevant for particulate P, the issue is particularly relevant for dissolved and colloidal P, which both appear much more significant contributors to P losses from agricultural lands than previously assumed (Kleinman et al., 2011, 2015; Dupas et al., 2015c; Jiang et al., 2015a; Mellander et al., 2015, 2016; Gu et al., 2017), and are also potentially bioavailable P forms. From the perspective of locating dissolved and colloidal P sources in the landscape, the way landscape structures, in interaction with climatic variables, control ground- and surface water dynamics is of fundamental importance. Groundwater level changes in the overall landscape, in response to precipitation and evapotranspiration, influence the location, timing and duration of soil water saturation and wetting-drying episodes that in turn cause reduction of soil Fe-oxyhydroxides. Ground- and surface water dynamics are thus anticipated to be the important determinants of hot spots and hot moment release of dissolved/colloidal P in catchments. An example of such complex control of landscape structure on P release dynamics in catchments has been provided in a recent study conducted on riparian wetlands in a small French catchment (Gu et al., 2017). Large differences have been observed with regards to P release dynamics in the riparian zone, depending on the topography and the control it exerts on the location and timing of biogeochemical processes such as soil Fe-oxyhydroxide reduction or P leaching following soil rewetting. This study

has led to the development of a coherent, spatially integrated concept of “landscape biogeochemistry” (see **Figure 3**) in which the topography, as a first-order control on spatial variation of hydrological conditions (Sørensen et al., 2006), appears to control spatial and temporal distribution of P release processes, the inputs of P from cultivated soils situated upslope (control of topography on soil erosion) and the organic and inorganic nature of released P through indirect control of topography on the rate of soil organic matter mineralization.

The chemistry of P in the soil-water continuum is often controlled through interactions with iron (Fe). Iron is a redox sensitive element and is commonly present in the soil-groundwater-surface water system. Mechanisms such as reduction of soil Fe-oxyhydroxides or rewetting of dried soils are known to trigger the release of P in soils, notably that of dissolved and colloidal P fractions (Turner and Haygarth, 2001; Stutter et al., 2009; Blackwell et al., 2010, 2013; Obour et al., 2011; Scalenghe et al., 2012; Gu et al., 2017; Missong et al., 2018). Redox gradients also result in the precipitation or dissolution of Fe-rich particles, which have a major impact on the fate and bioavailability of P (Jiang et al., 2017). Particulate or colloidal P is formed at the groundwater-surface water interface in groundwater-fed lowland catchments (Van der Grift et al., 2014, 2018; Baken et al., 2015). The formation of Fe hydroxyphosphate precipitates, with molar P/Fe ratio of 0.5 is the main immobilization process of dissolved P during Fe(II) oxidation (Voegelin et al., 2013; Van der Grift et al., 2016) and therefore a major control on the P retention in natural waters that drain anaerobic aquifers. As a consequence, Fe-bound P can be the dominant P fraction in suspended particulate matter under such conditions (Van der Grift, 2017). The Fe(II) oxidation may also lead to formation of Fe(III) bearing colloids. Furthermore, redox mediated P release from river sediments in lowland rivers has been identified as dissolved reactive P (DRP) release mechanism during summer anoxia (Smolders et al., 2017). Reductive dissolution of ferric Fe oxides was found to be associated with mobilization of P to the water column from sediments with a molar P/Fe ratio >0.4. In contrast, no sediment DRP release was found for a lower P/Fe ratio irrespective of temperature and dissolved oxygen treatments. Hence, the P/Fe molar ratio in sediments is an indicator for P mobilization. Clearly, Fe redox dynamics should be taken into account when describing and predicting P transfer from both soils and sediments to surface water.

Particulate P, the dominant P form in many agricultural areas in Northwest European countries like Norway (Bechmann et al., 2008), may be transferred within the headwater catchments adsorbed onto soil particles, but it may also be released from the soil particles when it reaches the recipient waterbody. In addition, some algae can also use particulate P directly. When soil particles do contain high levels of P (high soil P status), these particles may release high amounts of P when diluted in water. Therefore, the importance of particulate (including colloidal) associated phosphorus for algal growth, both on a short and long-term time-scale requires further investigation.

Locating and apportionment of the sources and fluxes of the dissolved vs. colloidal P conveyed by rivers in landscapes remains

a longstanding and fundamental challenge for catchment scientists (see McClain et al., 2003; Bol et al., 2016; Gottselig et al., 2017b; Missong et al., 2018), with major implications for land management and nutrient pollution mitigation strategies. Colloidal P is characterized as dissolved P in most routine water quality monitoring programs (Gottselig et al., 2017a), however, it is believed to have reduced bioavailability compared to the truly dissolved P (Baken et al., 2014). Furthermore, colloids which consist of organic matter, Fe/Al oxyhydroxides and clay minerals (Jiang et al., 2015a; Missong et al., 2017) are important P carriers in and to surface waters (Gottselig et al., 2014, 2017a,b) and thus should be more explicitly accounted for in P budgets. These interactions between P and colloids need to be understood to better assess and predict P mobility, availability and eutrophication in surface waters, such as homo- and hetero-aggregation behavior of colloidal particles as a controlling factor for P retention. While the mobilization and retention of P cannot be fully understood without understanding its interaction with Fe it is currently omitted from current water quality models and one of their important limitations.

UNCERTAIN IMPACTS OF WEATHER AND CLIMATE CHANGE ON P STATUS

Human influence on the global climate and a range of future climate projections are now well established (Jenkins et al., 2010; IPCC, 2014). However, less information is currently available in the literature on how climate change will impact on current and future P losses from agricultural landscapes (Ockenden et al., 2017). Similarly, how to disentangle current and future trends in P fluxes due to climate change itself from those related to climate driven changes in agricultural management practices and P inputs (Dupas et al., 2016; Bussi et al., 2017). Furthermore, the responses to climate change could vary widely across geographical scales due to localized differences in the sensitivity of catchment P losses to climatic drivers (Mellander et al., 2018), thus highlighting the need to establish climate vulnerability maps for better assessments of the sensitivity of P water quality trends (such as eutrophication potential) to climatic factors. For example, in Western Europe the weather patterns are influenced by both anthropogenic warming and by decadal trends in the North Atlantic Oscillation (NAO) (Hurrell, 1995). Thus long term changes in weather are not spatially uniform and the effects on water quality are expected to vary for different physical and chemical settings, making an area more or less susceptible to P loss (Mellander et al., 2018). We therefore need to better understand both the heterogeneity in large scale weather changes and the influence on small scale processes of P release, retention and transfer pathways within the landscape, as each pathway has a different impact on water quality at the catchment scale. This knowledge is needed both in order to assess current water quality trends (concentrations and loads) and to model future scenarios for specific regions.

As said, while the impacts of projected climate on river hydrological regimes are widely studied, fewer projections of the likely impact on future P pollution in running waters exist

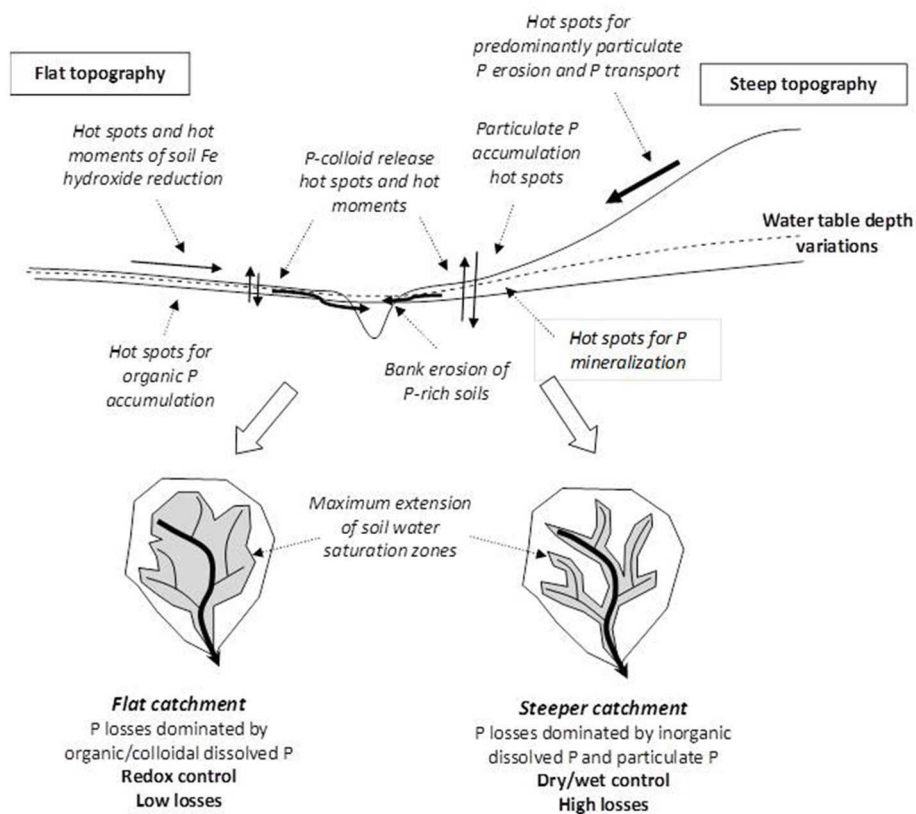


FIGURE 3 | Sketch illustrating the concept of landscape biogeochemistry applied to dissolved, colloidal and particulate P emissions in catchments (adapted from Gu et al., 2017).



FIGURE 4 | Overland flow on an arable field with well drained soils (country Wexford, Ireland), following heavy rainfall in November 2014 [photo: Michael Fleming].

(Whitehead et al., 2009; Dunn et al., 2015; Mehdi et al., 2015; Hesse and Krysanova, 2016; Ockenden et al., 2016, 2017). This may be related to the continuing complexity of human activities and its impact on water quality across a range of different spatial scales, including the small headwater catchment scale, while climate scientists traditionally focus their work mostly only at the larger scales (Michalak, 2016). In addition, while it is possible to model the link between precipitation and discharge with reasonable certainty, the relationship between climate change and water quality is subject to uncertainties in our understanding of the potential effects of climate change on the physical and biogeochemical processes along the nutrient source-mobilization-delivery-impact continuum. Furthermore, it is difficult to disentangle direct climate change impacts from the indirect consequences of adaptive land use mediated by climate change (Bussi et al., 2016, 2017). Thus, water quality modeling with respect to climate change impacts is subject to more weaknesses and uncertainties compared to rainfall-runoff modeling (Hesse and Krysanova, 2016). Understanding the impact of climate change on water quality will require an understanding of the link between P concentrations, loads and recipient freshwater ecosystems within the context of multiple stressors (Whitehead and Crossman, 2012; Crossman et al., 2013), including point source and diffuse pollution sources and water abstraction.

Across NW Europe, national scale climate projections vary from relatively straightforward patterns of drier summers and wetter winters in the Atlantic climate across the British Isles (Jenkins et al., 2010), to more complicated regional patterns and higher uncertainty related to both the direction and magnitude of change in precipitation in the more continental climatic regime of Germany (e.g., Van der Linden and Mitchell, 2009; Gädeke et al., 2017). Meanwhile, in Scandinavian countries (Norway and Sweden), climate projections have indicated greater warming in the northern latitudes than in southern latitudes, particularly during winter. The precipitation in Sweden is expected to increase throughout the country but more moderately in summer (Lind and Kjellström, 2008), while in Norway precipitation is expected to increase across the country with the highest increase expected during spring in Mid-Norway (Hanssen-Bauer et al., 2009). Air temperatures affect soil P mineralization, therefore increased temperatures in summer will likely lead to a buildup of labile P pool in the soil (Turner and Haygarth, 2001), ready for transfer to waterbodies by the autumn/winter rains. Thus, areas where both summer temperatures and winter rainfall are projected to increase are likely to become more prone to P mobilization and thus vulnerable to P loss to water (Jordan et al., 2012) (**Figure 4**). Soil rewetting after a dry summer will mobilize P of mostly microbial origin, largely controlled by the soil P status (Dupas et al., 2015a), while prolonged wet periods due to more rain or more frequent rain may further enhance P release due to reductive dissolution of Fe (hydr)oxides (Van der Grift et al., 2016; Gu et al., 2017). In both cases, the mobilized P consists mainly of dissolved (both organic and inorganic) P forms, i.e., of P forms which are potentially highly bioavailable to algae.

Drying-rewetting and freezing-thawing episodes of soils are two of the most common forms of abiotic soil perturbation,

resulting in solubilisation and release of P (Blackwell et al., 2009). In response to climate change it is likely that the frequency of such episodes and/or the duration of the dry and frozen periods will change differently in different areas. Furthermore, soil freezing-thawing events can increase the rates of P losses due to freezing of plant material (e.g., Bechmann et al., 2005). In Northern European countries, such as Sweden, projected warmer temperatures will likely shorten the period of persistent snowpack and cause more soil freezing-thawing episodes (Mellander et al., 2007) with increased erosion and loss of particulate P. In German lower mountain ranges snowmelt events are often an important source of sediment and P loss (Ollesch et al., 2005, 2006). Increasing temperatures may therefore shorten periods with snow cover, but could in contrast to Northern Europe also lower surface runoff due to smaller number of snowmelt events (Anis and Rode, 2015). These may mobilize both the dissolved and particulate P. In Norway, the trend in the number of freeze-thaw events depends on altitude, with an increasing number observed in the mountains (Hanssen-Bauer et al., 2009) and a decrease observed in the lower lying agricultural areas along the coast (Bechmann and Eggstad, 2016).

Catchments at high risk of P transfer are particularly responsive to changes in climate. For example, in a hydrologically flashy agricultural catchment in Ireland the total reactive phosphorus (TRP) loss was threefold that of a more hydrologically buffered catchment, despite similar soil P status (Mellander et al., 2015). The inter-annual variability in P loss in the flashy catchment was larger than the difference between the two catchments, highlighting the catchment dependence of P transfer risk and sensitivity to more rain, or more frequent rain, to catchment different hydrological flow paths. In another example from Norway, soil erosion was found to be the main process of P transfer in agricultural catchments with arable land and a flashy hydrology. In these catchments the soil P status appeared to have an influence on the catchment P loss (Bechmann et al., 2008). Thus changing rainfall patterns and rainfall intensity under climate change may affect rainfall erosivity and therefore particulate P mobilization and transfer to watercourses (Panagos et al., 2017; Poggio et al., 2018).

An understanding of the magnitude of different hydrological flow paths, their response to climate change and their potential effects on nutrient and soil loss processes will be necessary for choosing the right mitigation measures under future climate scenarios. While the amount of P lost from catchments increases with runoff, several mitigation measures, such as sedimentation ponds, have been implemented to reduce P loading from agricultural catchments, with an effective annual retention of 8–35% of TP (average 18%) (Blankenberg et al., 2013). However, a recent study found that current mitigation efforts will not be sufficient to combat the effects of climate change on P losses, particularly in areas where increased winter runoff will lead to an overriding increase in P loads (Ockenden et al., 2017).

The importance of concentrations vs. loads and seasonality of climate change impacts (i.e., summer vs. winter) will vary with the type of receptor waterbodies (e.g., headwater catchments vs. downstream river reaches and lakes) and will have different

impacts on freshwater ecology. For example, increased winter river flows may lead to increased total annual P loads (Ockenden et al., 2017), leading to negative impacts on standing waters such as lakes and reservoirs during the ecologically active period of spring and summer (Stamm et al., 2014). Conversely, increased P concentration during reduced summer flows (Bussi et al., 2017), could lead to negative impacts on river ecosystems (Stamm et al., 2014). These likely differential seasonal and water-body effects will be important to consider when modeling the impact of climate change scenarios on aquatic ecology. Thus improvement of our mechanistic understanding at multiple scales along with development of novel methods for accommodating rigorous error analysis are the imperative challenges for the future of integrated water quality modeling (Rode et al., 2010). Meanwhile, modeling the complex interactions between climate change impacts, social and economic adaptation and land use change is not a trivial task and whilst it is critical for future adaptation policies, to our knowledge, only limited examples of such integrated analysis are available to date (Dunn et al., 2012; Mehdi et al., 2015; Sample et al., 2016).

CONCLUSION FOR FUTURE RESEARCH

Independent of the variety of issues highlighted in the present paper which do highlight the fact that the reduction of phosphorus (P) based water eutrophication in the agricultural landscapes of Northwest Europe is neither simple nor straightforward. We like to conclude that there is a crucial need for more integrative research efforts to deal with our

incomplete understanding of the mechanisms and processes associated with the identification of critical source areas, P mobilization, delivery and biogeochemical processing, as otherwise even high-intensity and high-resolution research efforts will likely only reveal an incomplete picture of the full global impact of the role of terrestrial derived P on downstream aquatic and marine ecosystems.

AUTHOR CONTRIBUTIONS

All authors listed have made a substantial, direct and intellectual contribution to the work, and approved it for publication.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: <https://www.frontiersin.org/articles/10.3389/fmars.2018.00276/full#supplementary-material>

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