



## Wetland buffer zones for nitrogen and phosphorus retention: Impacts of soil type, hydrology and vegetation



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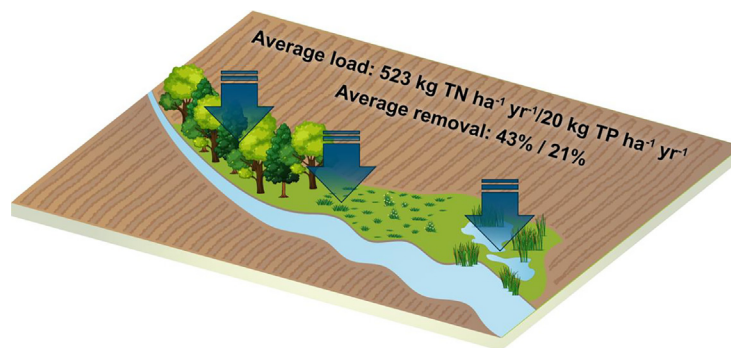
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### HIGHLIGHTS

- The efficiency of wetland buffer zones for nutrient retention was reviewed.
- Organic and mineral soils as nutrient filters or sources were compared.
- Processes driving phosphorus and nitrogen fluxes were described.
- The indirect and direct impact of vegetation were unraveled.
- Implications for wetland restoration and open research questions were specified.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Wetland buffer zones (WBZs) are riparian areas that form a transition between terrestrial and aquatic environments and are well-known to remove agricultural water pollutants such as nitrogen (N) and phosphorus (P). This review attempts to merge and compare data on the nutrient load, nutrient loss and nutrient removal and/or retention from multiple studies of various WBZs termed as riparian mineral soil wetlands, groundwater-charged peatlands (i.e. fens) and floodplains. Two different soil types ('organic' and 'mineral'), four different main water sources ('groundwater', 'precipitation', 'surface runoff/drain discharge', and 'river inundation') and three different vegetation classes ('arboraceous', 'herbaceous' and 'aerenchymous') were considered separately for data analysis. The studied WBZs are situated within the temperate and continental climatic regions that are commonly found in northern-central Europe, northern USA and Canada. Surprisingly, only weak differences for the nutrient removal/retention capability were found if the three WBZ types were directly compared. The results of our study reveal that for example the nitrate retention efficiency of organic soils ( $53 \pm 28\%$ ; mean  $\pm$  sd) is only slightly higher than that of mineral soils ( $50 \pm 32\%$ ). Variance in load had a stronger influence than soil type on the N retention in WBZs. However, organic soils in fens tend to be sources of dissolved organic N and soluble

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reactive P, particularly when the fens have become degraded due to drainage and past agricultural usage. The detailed consideration of water sources indicated that average nitrate removal efficiencies were highest for ground water ( $76 \pm 25\%$ ) and lowest for river water ( $35 \pm 24\%$ ). No significant pattern for P retention emerged; however, the highest absolute removal appeared if the P source was river water. The harvesting of vegetation will minimise potential P loss from rewetted WBZs and plant biomass yield may promote circular economy value chains and provide compensation to land owners for restored land now unsuitable for conventional farming.

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**Summary of the main finding from this work (Capsule):** The efficient removal of nutrients found in almost all the studies investigated in this review suggests that the role of wetland buffer zones should be recognised in large scale, long-term pollution management. Differences in retention between organic and mineral soils were negligible compared with the effect of variable loading rates and, in addition to this, forested WBZs proved to be the most effective in nutrient retention. Harvesting is highly recommended to improve the nutrient removal capabilities of rewetted buffer zones.

## 1. Introduction

Three billion people are expected to join the global population over the next two decades, thereby accelerating the degradation of natural resources and escalating the competition for them (Scanlon et al., 2017). Along with this increase, the threat to water quality is alarming, eutrophication induced by agricultural diffuse sources being one of the main causes of aquatic ecosystem deterioration (Kronvang et al., 2007; Jabłońska et al., 2020). In a business-as-usual scenario, the global demand for clean water will exceed viable resources by 40% by 2030 (WWAP, 2015). In Europe, an increasing demand for water and a major pressure on aquatic ecosystems come from the still-growing agricultural sector. Despite substantial efforts to reduce fertiliser application and adopt best land use and management practices (Neal and Jarvie, 2005; Lam et al., 2011), nutrient pollution problems persist. This is in part due to the ongoing loss of natural riparian wetlands that act as important water and nutrient regulators in the landscape (Verhoeven et al., 2006; Wharton and Gilvear, 2007; Zak et al., 2018a). The majority of European wetlands are currently used for agriculture and forestry purposes, which exacerbates the problem of nutrient pollution of aquatic systems (van Diggelen et al., 2006).

Floods or water deficiency, nutrient pollution, biodiversity loss and increasing greenhouse gas emissions have led to the implementation of large wetland restoration projects in not only many European countries, but also in other parts of the world (Zak et al., 2018a). In this context, a major emphasis is put on the rehabilitation of river systems and adjacent wetland buffer zones (WBZ) (Gericke et al., 2020). Wetland buffer zones capture and recycle part of nutrient-rich runoff water before it reaches rivers and lakes, hereby reducing nutrient loads in surface waters at the land-water interface (Dørgé, 1994). Restoration of WBZ systems is considered to be an effective, broad-scale approach to mitigating nutrient pollution and degradation of freshwater and marine ecosystems (Klimkowska et al., 2007), thereby meeting the goals of the EU Water Framework Directive (2000/60/EC). In Denmark and other European countries, WBZ restoration has been incorporated into policy actions to improve the quality of inland surface waters and neighbouring seas (Iversen et al., 1993; Hoffmann and Baattrup-Pedersen, 2007). For example, the 2nd and 3rd Danish Action Plans on the Aquatic Environment (implemented in 1998 and 2004) successfully used restoration of riverine environments and creation of 50,000 ha of WBZ to reduce nitrogen (N) and phosphorus (P) loads into adjacent water bodies (Action Plan for the Aquatic Environment III, 2005–2009). However, due to extensive modern agricultural drainage,

riparian wetlands commonly exhibit significantly altered physical and chemical soil properties and hydrological regimes, so rewetting cannot restore the original hydrological and ecological functioning within a human life-time (Tiemeyer et al., 2007; Zak et al., 2018a). Long-term dehydration of the topsoil results in soil degradation and subsidence, upon which accumulation of rainwater and flooding from the adjacent water bodies may follow. This, in particular, holds true for long-term drained groundwater-charged peatlands (i.e. fens) (Zak et al., 2010). It is unlikely that rewetting of these degraded wetland systems will restore the hydrology to predominantly groundwater-percolated peatlands; instead it will create new systems fed by a mixture of groundwater, rainwater and surface water (Wheeler and Proctor, 2000). High nutrient availability in the altered wetland soils poses a risk of nutrient flux to surface waters post-rewetting (Tiemeyer et al., 2006; Zak et al., 2018a). Therefore, planting and harvesting of wetland plants (e.g. cat-tail, reed) are considered valuable options to remove additional nutrients from the system (Zak et al., 2014).

Over the last three decades, a wealth of studies describing the importance of different riparian wetland types for nutrient retention, including systematic or critical reviews and meta-analyses, has been published (see Gericke et al., 2020 and references herein). Accordingly, extensive knowledge is already available, not least of N removal in the riparian zones of agricultural watersheds (Martin and Reddy, 1997; Sabater et al., 2003; Hill, 2019; Valkama et al., 2019). Also, it is well documented that wetlands may act as significant P sinks (Mander et al., 1997; Abu-Zweig et al., 2003; Fisher and Acreman, 2004; Lowrance and Sheridan, 2005), even though rewetted wetlands may act as P sources several years after restoration (Audet et al., 2020). Furthermore, previous reviews indicate that the rates and processes of nutrient removal differ markedly depending on nutrient load and nutrient form, soil type/texture, vegetation and flow pathways (Syversen and Borch, 2005; Lind et al., 2019; Gericke et al., 2020). However, most of the existing knowledge of the biogeochemical processes and factors involved in nutrient removal is gained from constructed wetlands, as in a recent systematic review of N and P retention in restored freshwater wetlands (Land et al., 2016). Likewise, the biogeochemical processes occurring after rewetting of wetlands are generally well documented (e.g. Zak et al., 2018a). Yet, the relative importance of the individual processes in different WBZ types in relation to hydrological pathways is still poorly investigated (Hill, 2019). As a step forward, the objective of this paper was to compile the current knowledge of nutrient retention in various WBZ types with special emphasis on soil quality, vegetation forms and different hydrogeological settings. This will help to improve our understanding of the risks and benefits of rewetting wetlands and to scale up the scope of restoration from single sites to entire river systems. Specifically, the following three questions are addressed:

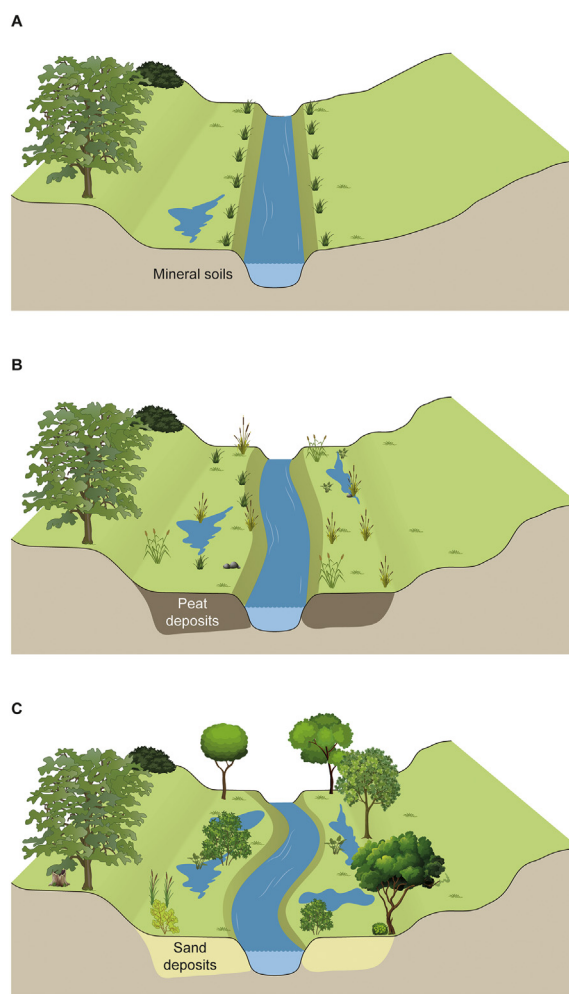
1. Are WBZs on organic soils more effective N and P filters than those on mineral soils?
2. How do hydrology and biogeochemical processes influence the retention of nutrients?
3. How plants and harvesting of plants affect the nutrient budget of a WBZ?

## 2. Materials and methods

### 2.1. Characterisation of wetland buffer zones and their filter function

In this study, the definition of a WBZ is a zone of transition, or an ecotone, between terrestrial and riverine ecosystems (Naiman and Decamps, 1997). WBZs can either have a terrestrial character in the shape of a vegetated bank with water levels close to the ground surface or they can be aquatic in the shape of a shallow water body forming part of the greater river-floodplain system. From a hydrological perspective, the WBZ acts as an interface for dynamic groundwater surface water interactions by regulating physical and biogeochemical processes along the river-buffer zone ecotone (Smith et al., 2008). WBZs purify waters of riverine ecosystems by removal and retention of nutrients present in waters moving from terrestrial to riverine ecosystems (or from an upper course of a river to its lower course). Nutrient removal describes the processes facilitating the export of specific nutrients from incoming waters to the atmosphere, often through chemical transformations of the nutrients for instance through denitrification and loss of nitrogenous gases (i.e.  $N_2O$  and  $N_2$ ). Nutrient retention is the process of retaining or taking up nutrients dissolved in transfer water and storing them in the soil or biomass within the WBZ. As simplification the term 'retention' is used in the following text solely unless removal processes like denitrification are explicitly considered. Several processes drive the nutrient filter function of WBZs, such as i) N removal by biotic and abiotic processes such as heterotrophic denitrification or chemical catalysed oxidation of ferrous iron, ii) nutrient uptake by vegetation, iii) P precipitation and/or P sorption reactions and iv) deposition and/or sedimentation processes. For a WBZ to act as a nutrient sink, hydrological connectivity with incoming waters is a prerequisite to allow biological, chemical and physical processes to take place in order to lower the nutrient content in the water leaving the WBZ system. The character of water transfer within wetlands differs depending on the type of the WBZ and its particular eco-hydrological conditions; for instance, soil and vegetation characteristics may vary both spatially and seasonally. WBZ types can be classified based on their dimensions, soil composition, hydrology and vegetation, favouring different nutrient mitigation management measures as described in detail in Jabłońska et al. (2020). The sites selected for extraction of the data used in this study were categorized in three groups (Fig. 1, see supplement for individual categorization):

- (1) **Riparian mineral soil wetland** — narrow strips of “wet land” including river banks along streams or smaller rivers created by periodically changing river water levels. A higher water level in the stream may result in inundation of the land in its proximity but this is not common. Typically, the width of the flooded zone is several metres and the most frequent soil type is sandy deposits. Large floodplains are not included in this category.
- (2) **Fen** — peat-accumulating wetlands, typically developing in groundwater discharge sites. Fens are usually dominated by sedges or occasionally by reedbeds, shrubs and trees. While the groundwater flowpath is often the main water source, the hydrology can be much more complex than the simple lateral groundwater flow. Full water balances including all flow paths have rarely been determined, let alone the complex interactions and exchange occurring between the various flow paths (Petersen et al., 2020a). Typically, the investigated fens consist of spring mires at the valley's edge, wider percolation mires dominated by groundwater flow and strips of flood mires (Joosten, 1997). Undrained fens in an original near-natural state must be distinguished from rewetted fens that have previously been drained and usually utilised for agricultural purposes, and which have a surface layer of heavily mineralised peat, so-called “moorsh soil”. Rewetted fens can occasionally appear as shallow lakes due to significant soil subsidence (Zak et al., 2018a)



**Fig. 1.** Wetland buffer zones in this study: riparian mineral soil wetland (A), fen (B) and floodplain (C).

- (3) **Floodplains** — larger areas adjacent to streams or rivers. They may be up to several km wide (at least in a natural state) and are usually flooded by water for several weeks at the end of the cold seasons but also at other times of the year. Typically, floodplains are characterized by mineral soils, but organic soils may also occur.

It must be noted that it was sometimes challenging to clearly attribute one of these categories to the sites included in the present study because of the following reasons: (i) These WBZ types are broadly defined and rather constitute a gradient than clearly distinct categories, (ii) study areas often represented more than one WBZ type, and (iii) a clear description of the sites was not always available in published studies used for this work. Taking these limitations into account, the analysis in this work did not solely focus on the three WBZ types. In addition, soil types, hydrology and vegetation patterns were used for further classification as described below (see Section 2.3).

### 2.2. Literature collection and inclusion criteria

For the literature review, we searched the ISI Web of Science (<http://apps.webofknowledge.com/>) and Scopus database (<https://www.scopus.com/>) using the following keywords: 'minerotrophic peatlands' OR 'fen' OR 'percolating mire' OR 'buffer zone' OR 'buffer strip' OR

'restored wetland' OR 'filter strip' in combination with 'nutrient retention', 'nutrient capture', 'nutrient balance', 'nutrient budget', 'nitrate', 'nitrogen', 'phosphate' and 'phosphorus'. A total of 82 individual wetland studies were identified based on information extracted from 51 publications (for details see supplement). Searches were conducted in English only due to difficulties with translation from other languages, with the exception of some studies published in Danish and German, included by native speaking co-authors. Strict criteria were applied to the search results to define their suitability for inclusion in the review:

1. The literature search was restricted to WBZs in the temperate (Temperate Oceanic Climate, Cfb) and continental (Subarctic, Dfc; humid Continental, Dfa and Dfb) climatic zones of the Köppen-Geiger climate classification system (Beck et al., 2018).
2. Only WBZs as characterized in Section 2.1 were analysed. Ombrotrophic peatlands (bogs) where the major hydrological input derives from rainfall were not considered in this review.
3. Studies reporting water nutrient concentrations without loading rates were excluded.
4. A prerequisite was that studies were carried out under natural climatic conditions, so studies conducted in a greenhouse or laboratory setting were excluded (both microcosm and mesocosm studies).
5. In order to obtain realistic values, seasonal variations must be accounted for, so studies not covering at least one whole year were excluded.
6. When different publications covered the same wetland in different years, averages were used so as not to overly bias the results towards only a few wetlands.
7. Constructed wetlands artificially established by earthworks to create pond systems were not included in the review. Included were artificially flooded constructed wetlands with in-situ soil and vegetation structures (representing flood event scenarios).

Where information published in a review or meta-analysis was used as a source, the initial information sources were considered. Due to data availability, for Europe, mainly studies on converted minerotrophic fens were retrieved, while principally studies on riverine buffer zones with mineral soils and variable vegetation covers were retrieved for north-eastern USA and Canada. Two wetlands from temperate New Zealand were included. The studies were sorted per region of origin and country (Fig. 2). The nutrients investigated were N in the form of nitrate ( $\text{NO}_3^-$ -N), ammonium ( $\text{NH}_4^+$ -N), dissolved organic N (DON) and total nitrogen (TN) and P in the form of soluble reactive phosphorus (SRP) and total phosphorus (TP). For studies not specifically mentioning DON, DON

was calculated as  $\text{NO}_3^-$ -N and  $\text{NH}_4^+$ -N subtracted from total dissolved N. Other information extracted from the available studies was vegetation of the WBZ, soil type (organic or mineral), degradation state of organic soils, soil nutrient status, main water source to WBZ, main flow path from WBZ to surface waters, whether incoming water originated from agricultural drainage, wetland type, wetland area or buffer strip width, mineral soil type (also those underlying fens), flooding of site, whether harvested biomass was included and calculated and if N deposition from the atmosphere was included in the load.

Nutrient fluxes were quantified as nutrient mass balances. The following fluxes as mass per area and time ( $\text{kg ha yr}^{-1}$ ) were distinguished: 1) 'Nutrient Load' flowing into the WBZ, 2) 'Nutrient Loss' flowing out of the WBZ, and 3) 'Retention' by the WBZ calculated from the difference between nutrient load and nutrient loss. Finally, 'Efficiency' was calculated as retention given as a percentage of the nutrient load. There are uncertainties involved in using the mass balance method of retention calculation. These may result from, for instance, missing information about the direct fertiliser input to the WBZ (if still active pastoral land), missing estimation of existing soil nutrient loading (remnants from previous agricultural use) or uncertainty of or missing estimation of the atmospheric deposition to land (Bach and Frede, 1998). Loss of nutrients by seepage into deeper groundwater and the loss of N via emissions of nitrous oxides were not quantified because such losses were not ascertained in the majority of the studies. If papers cited only retention or efficiency, then these values were also included in the analysis.

To obtain a more detailed picture of the nutrient cycling in WBZs, literature on the nutrient stock of plants typical for WBZs and on denitrification rates was also reviewed. Other biotic processes such as anaerobic ammonium oxidation and dissimilatory nitrate reduction or abiotic chemolithotrophic nitrate reduction were omitted from the review as the published data were mostly obtained from constructed wetlands or insignificant in natural WBZs. To obtain information on nutrients and nutrient uptake by plants, we used the search terms 'wetland', 'wetland buffer zone', 'peatland' AND 'nitrogen stock', 'nitrogen uptake' OR 'phosphorus stock', 'phosphorus uptake' AND 'alnus' OR 'salix' OR 'quercus' OR 'phragmites' OR 'typha' OR 'sedges' OR 'helophytes'. In addition, unpublished data on plant communities monitored in 11 rewetted wetlands of the River Odense catchment in Denmark were included in the study. For an overview of denitrification rates, two recent extensive scientific meta-analyses were identified (MA-1: Alldred and Baines, 2016; MA-2: Zhou et al., 2017) using a structured search in ISI Web of Science, and literature herein was used for further analysis (MA-1: Kaplan et al., 1979; Groffman et al., 1992;

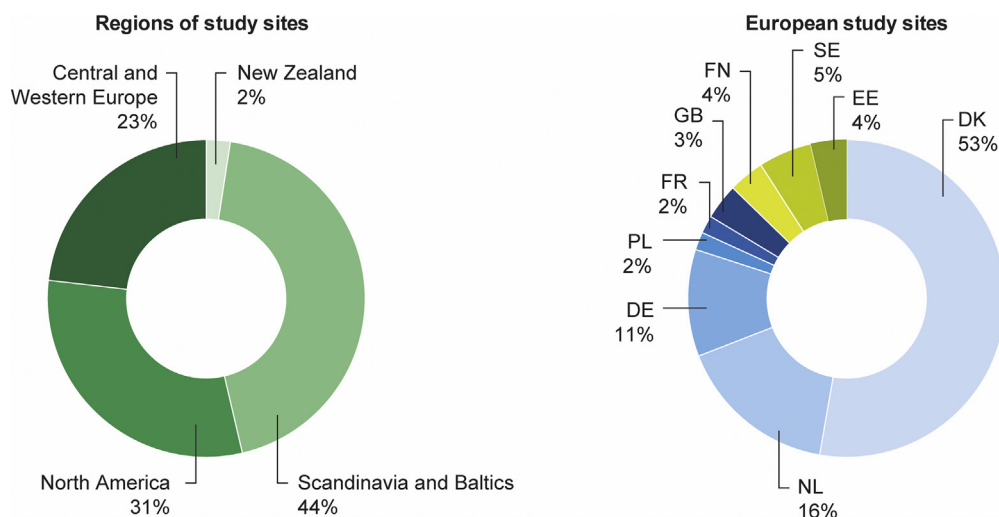


Fig. 2. Origin of reviewed study sites (DK: Denmark, NL: The Netherlands, DE: Germany; PL: Poland, FR: France, GB: Great Britain, FN: Finland, SE: Sweden, EE: Estonia).

Hanson et al., 1994; Bodelier et al., 1996; Lusby et al., 1998; Otto et al., 1999; Ottosen et al., 1999; Verhoeven et al., 2001; Findlay et al., 2003; Windham and Ehrenfeld, 2003; Windham and Meyerson, 2003; Davis et al., 2004; Wigand et al., 2004; Olde Venterink et al., 2006; MA-2: Thomas and Christian, 2001; Antheunisse et al., 2007) (for details see supplement). Similar criteria as those used for the main literature survey were applied (criteria nos. 1, 2, 4 – but with mesocosms included, and 7).

### 2.3. Data processing and analysis

Data obtained from the review (both on general removal and denitrification rates) displayed a large variability in soil, hydrological and vegetation types that may affect nutrient retention differently, so subdivisions relative to 1) morphological setting (based on soil types), 2) hydrological pathways and 3) vegetation cover were created to aid the analysis of data:

- 1) WBZs grouped as 'Organic' were represented almost entirely by drained, degraded fens in Europe, while those grouped as 'Mineral' mainly covered riparian mineral soil wetlands and floodplains of mineral soil with low organic matter contents (usually <5% of dry mass). It should be noted that organic soils from peatlands (histosols) are defined as soils with an organic matter content of at least 30% (Mitsch and Gosselink, 2000); however, occasionally the term organic soil is used also for soils with a lower organic matter content than 30% but not lower than 10% of dry mass. Mineral soil types, including those below organic layers (where indicated), were silt-sand-gravel complexes, i.e. mainly fluvisols, varying in composition, or glacial moraines (poorly sorted clay to boulder, low permeability) with sand overburden.
- 2) Four conceptual hydrological pathways for the water discharge into the WBZs were distinguished in this paper: i) lateral flow of groundwater through the soil ('groundwater'), ii) precipitation, if coupled with nutrient analysis ('precipitation'), iii) overland flow into the riparian zone, which also includes nutrient-enriched, tile-drained waters entering the wetland through disconnected drains ('surface runoff/drain discharge'), and iv) irrigation and inundation of the buffer zone by river water ('river inundation').
- 3) Vegetation cover causes differing nutrient uptake rates and a differing presence of organic matter in soils to aid in denitrification (Osborne and Kovacic, 1993; Zak et al., 2014). To account for this, vegetation was subdivided into three categories or plant types: 'arborescent' (wooded plus an understorey), 'herbaceous' (semi-

natural herbaceous communities, mainly Poaceae) and 'aerenchymous' (mainly helophytes, i.e. grouping of reeds and sedges) (see Table 1 for single plant species). Mixed assemblages, where mentioned, refer to helophytes and grasses together.

Data analysis of sub-groups and comparison of nutrient load with the efficiency of removal for each nutrient type were performed using the R programming software version 3.5.2 (R Core Team, 2018). Individual datasets per nutrient type were analysed for significant correlations ( $p$ ) using the Pearson's test for non-monotonic datasets and the Spearman's test for monotonic datasets. Significance was tested against a level of 0.05. Unfortunately, neither hydraulic residence time, water holding capacity nor redox-gradients were given in the majority of the studies, so that their importance in nutrient removal could be not considered in detail.

## 3. Results and discussion

Overall, WBZs were found to work as effective barriers for diffuse nutrient pollution from agriculture. Of the 82 individual study sites included in this review, 27 (33%) received nutrient input mainly from river water, 26 (32%) from lateral groundwater discharge, and 22 (27%) from agricultural runoff through either tile drainage or surface overland flow. Precipitation was a substantial flow pathway at 7 sites (9%) (for details see supplement). All reviewed studies reported positive retention of  $\text{NO}_3^-$  and only one study was negative for TN, while negative retention was found for  $\text{NH}_4^+$ , DON, SRP and TP in 32%, 71%, 41% and 29% of the studies, respectively. Standard deviations (SD) were high in all studies due to the variability of natural settings (Table 2).

Previous research has shown that WBZs should retain 60% of  $\text{NO}_3^-$  from incoming waters (Tiemeyer et al., 2006) and this general value was accordingly used as an indication of retention efficiency at some stages of the analysis. Surprisingly, the three WBZ types categorized in the Section 2.1 of this study showed only minor differences with respect to retention and retention efficiencies for each of the investigated nutrient forms. There was a trend that TP retention is highest in floodplains, and almost all fens were characterized as DON sources (Fig. 3). The importance of soil types, hydrological pathways and vegetation forms will be considered in detail in the following sections.

### 3.1. Are WBZs on organic soils more effective nutrient filters than WBZs on mineral soils?

When looking separately at the different soil types, the average removal efficiencies for  $\text{NO}_3^-$  for organic and mineral soils were 52% and 51%, respectively (Fig. 4). Mean  $\text{NO}_3^-$  load and retention in the studies of organic soils were 588 and 173  $\text{kg NO}_3^- \text{-N ha}^{-1} \text{ yr}^{-1}$ , respectively, while the mean load and retention in the studies of mineral soils were 516  $\text{kg NO}_3^- \text{-N ha}^{-1} \text{ yr}^{-1}$  and 182  $\text{kg NO}_3^- \text{-N ha}^{-1} \text{ yr}^{-1}$ , respectively. Generally, we expected that rewetted fens with organic soils are better at removing  $\text{NO}_3^-$  from incoming waters than mineral soils. However,

**Table 1**

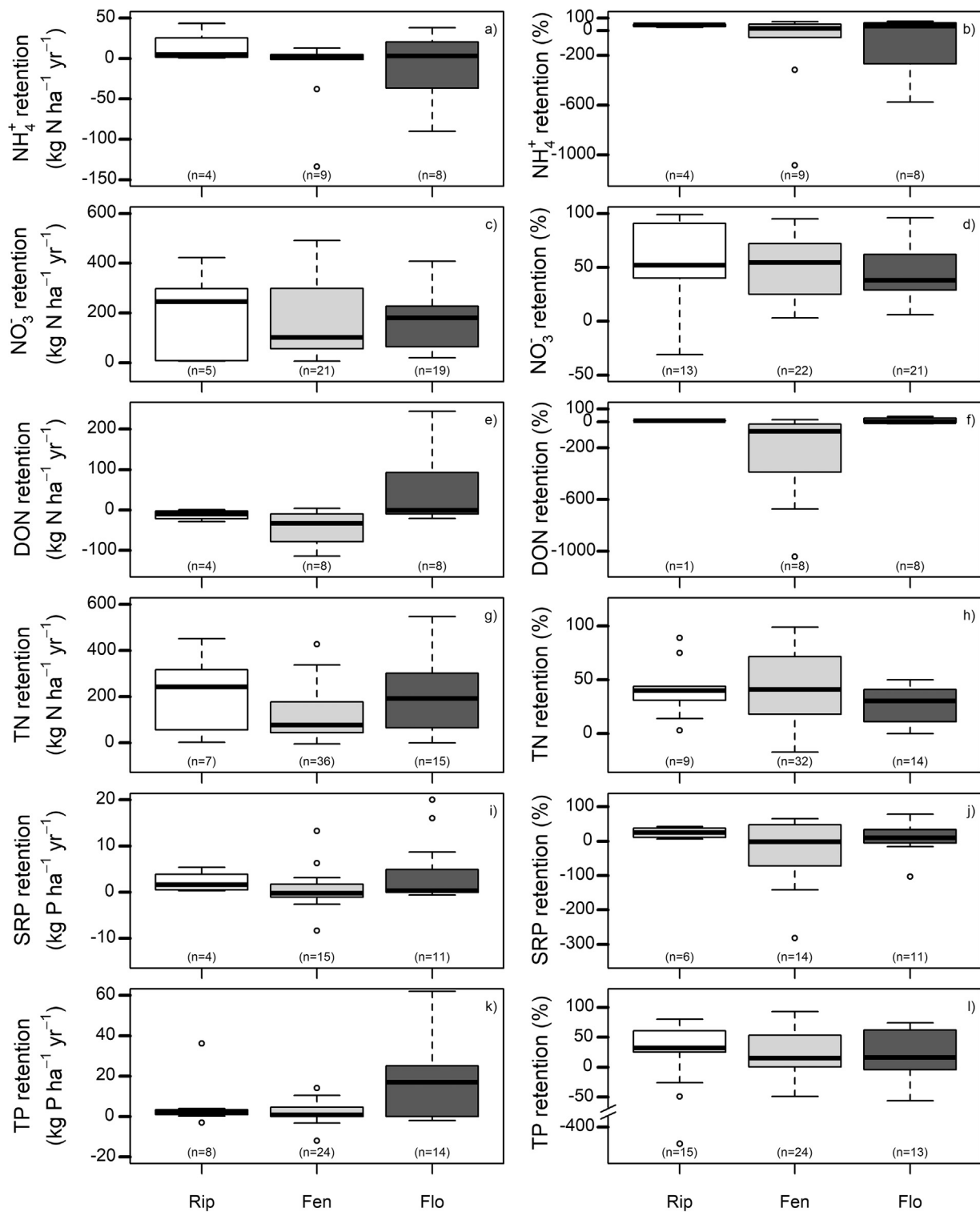
Plant species related to the plant types used in the review.

Plant types	Plant species
Arborescent	<i>Acer rubrum</i> , <i>Alnus glutinosa</i> , <i>Alnus rugosa</i> , <i>Populus</i> spp., <i>Quercus</i> spp., <i>Salix cinerea</i> , <i>Salix viminalis</i> , <i>Salix</i> spp.
Herbaceous	<i>Impatiens pallida</i> , <i>Impatiens</i> sp., <i>Limonium serotinum</i> , <i>Lythrum salicaria</i> , forage grasses ( <i>Agrostis stolonifera</i> and <i>Poa trivialis</i> ), <i>Symlocarpus foetidus</i>
Aerenchymous	<i>Carex acutiformis</i> , <i>Carex riparia</i> , <i>Glyceria maxima</i> , <i>Juncus effusus</i> , <i>Juncus maritimus</i> , <i>Littorella uniflora</i> , <i>Nuphar advena</i> , <i>Phragmites australis</i> , <i>Potamogeton pectinatus</i> , <i>Potamogeton perfoliatus</i> , <i>Schoenoplectus pungens</i> , <i>Scirpus olneyi</i> , <i>Scirpus</i> spp., <i>Spartina alterniflora</i> , <i>Spartina alterniflora</i> (short), <i>Spartina alterniflora</i> (tall), <i>Spartina patens</i> , <i>Typha angustifolia</i> , <i>Typha latifolia</i> , <i>Typha orientalis</i> , <i>Zostera marina</i>
Mixed aerenchymous/herbaceous	<i>Distichlis spicata</i> , <i>Spartina patens</i> , <i>Juncus</i> spp., mix of helophyte and semi-natural grassland vegetation (e.g., <i>Agrostis stolonifera</i> , <i>Mentha aquatica</i> , <i>Myosotis scorpioides</i> , <i>Poa trivialis</i> , and <i>Potentilla anserina</i> )

**Table 2**

Overall results of the literature review (mean  $\pm$  standard deviation, number of studies in brackets). Abbreviations:  $\text{NH}_4^+$  = ammonium,  $\text{NO}_3^-$  = nitrate, DON = dissolved organic nitrogen, TN = total nitrogen, SRP = soluble reactive phosphorus, TP = total phosphorus.

	Load $\text{kg N/P ha}^{-1} \text{ yr}^{-1}$	Loss	Retention	Efficiency (%)
$\text{NH}_4^+$	18 $\pm$ 19 (23)	26 $\pm$ 39 (21)	-8 $\pm$ 42 (21)	-90 $\pm$ 294 (21)
$\text{NO}_3^-$	560 $\pm$ 523 (46)	255 $\pm$ 253 (37)	177 $\pm$ 137 (45)	51 $\pm$ 31 (56)
DON	148 $\pm$ 196 (20)	142 $\pm$ 148 (21)	-1 $\pm$ 76 (20)	-112 $\pm$ 291 (17)
TN	523 $\pm$ 557 (56)	383 $\pm$ 495 (56)	149 $\pm$ 133 (63)	43 $\pm$ 30 (58)
SRP	8 $\pm$ 8 (28)	4 $\pm$ 5 (20)	2 $\pm$ 6 (30)	-5 $\pm$ 74 (31)
TP	20 $\pm$ 30 (48)	14 $\pm$ 24 (48)	7 $\pm$ 14 (49)	21 $\pm$ 72 (55)

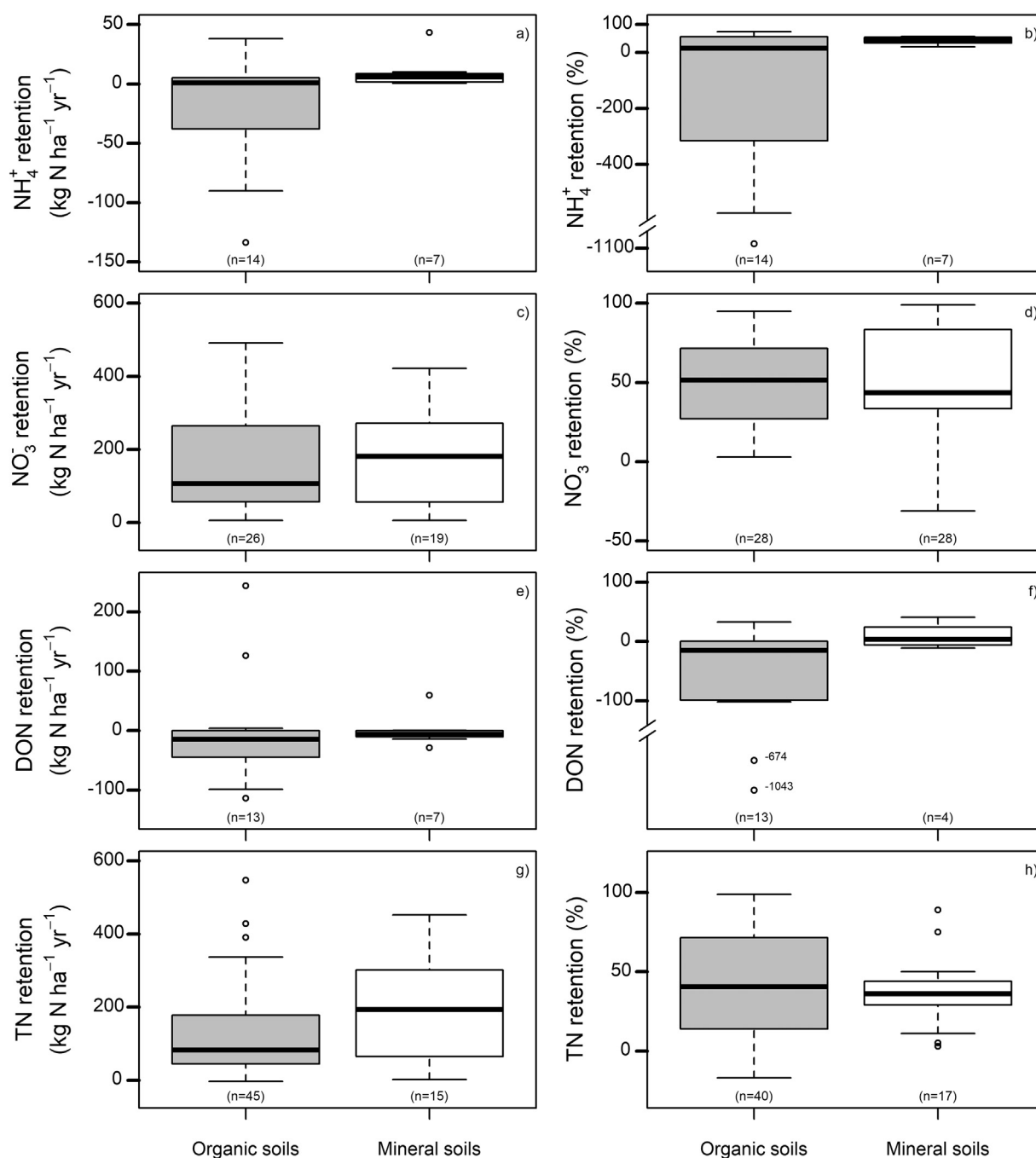


**Fig. 3.** Comparison of absolute nutrient retention and retention efficiencies of three WBZ types: riparian mineral soil wetland (Rip), ground-water charged peatland (fen) and floodplain (flo). Abbreviations:  $\text{NO}_3^-$ : nitrate,  $\text{NH}_4^+$ : ammonium, DON: dissolved organic nitrogen, TN: total nitrogen, SRP: soluble reactive phosphorus, TP: total phosphorus. In the box plots, the bottom and top of the box are the 25th and 75th percentiles, and the band within the box is the 50th percentile (the median); the ends of the whiskers represent the minimum and maximum of all the data and open dots are outliers.

variations in load have a stronger influence than soil type on  $\text{NO}_3^-$  retention in WBZs (see Section 3.2.1). Still, previous studies support the conclusion that organic soils have a greater ability to retain  $\text{NO}_3^-$  compared with adjacent mineral soil WBZs (Cooper, 1990; Schipper et al., 1993).

Only nine studies reported  $\text{NH}_4^+$  mass balances for fens, and three of these reported release to waters. Of these three sites, two were placed in

an experimental site in Sweden where release rates were very high ( $-1084\%$  and  $-313\%$ ) due to intense organic N mineralisation caused by flooding in the first year of the experiment (Leonardson et al., 1994), resulting in poor  $\text{NH}_4^+$  retention efficiency. Mean retention and efficiency values for fens were  $-16 \text{ kg NH}_4^+ \text{-N ha}^{-1} \text{ yr}^{-1}$  and  $-135\%$ . When extreme outliers were removed, a mean retention of



**Fig. 4.** Comparison of absolute retention and retention efficiencies of organic and mineral soils for different nitrogen species ( $\text{NO}_3^-$ : nitrate,  $\text{NH}_4^+$ : ammonium, DON: dissolved organic nitrogen, TN: total nitrogen). In the box plots, the bottom and top of the box are the 25th and 75th percentiles, and the band within the box is the 50th percentile (the median); the ends of the whiskers represent the minimum and maximum of all the data and open dots are outliers.

$3.7 \text{ kg NH}_4^+\text{-N ha}^{-1} \text{ yr}^{-1}$  and an efficiency of 26% were obtained, providing a better reflection of the overall values. Seven WBZs on mineral soils demonstrated means of  $10 \text{ kg NH}_4^+\text{-N ha}^{-1} \text{ yr}^{-1}$  retention and 42% efficiency.

The efficiencies of wetlands in removing DON varied. The average retention of DON was  $0.35 \text{ kg DON ha}^{-1} \text{ yr}^{-1}$  (9%) on mineral soils and  $-1.33 \text{ kg DON ha}^{-1} \text{ yr}^{-1}$  (-150%) on organic soils. Indeed, the average DON load on DON-exporting wetlands was low ( $122 \text{ kg DON ha}^{-1} \text{ yr}^{-1}$ ) compared with the average load on wetlands removing DON ( $285 \text{ kg DON ha}^{-1} \text{ yr}^{-1}$ ). Thus, when DON loads are small, wetlands on both mineral and organic soils may act as net sources of DON. Conspicuously, DON retention/release was not reported for any of the sites that mainly received groundwater inputs. Studies reporting large

releases of DON were primarily conducted in WBZs receiving surface runoff or tile drainage from upland areas, while reports of positive DON retention mainly came from floodplain WBZs (Fig. 3).

Fens and floodplains with organic soils were slightly more effective in removing TN than WBZs on mineral soils (45% and 36%, respectively). Fifteen studies from Denmark reported only retention within the range of 13 to  $428 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ . This is indicative of correspondingly high N loads as commonly found in other studies of rewetted WBZs from Denmark (Brüsch and Nilsson, 1993; Hoffmann et al., 2011). Mean loads on mineral soil WBZs were slightly higher than on fens ( $587$  against  $511 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ ). A single study showed release of TN, reportedly due to a high flux of DON during the first year of fen rewetting (Kieckbusch and Schrautzer, 2007).

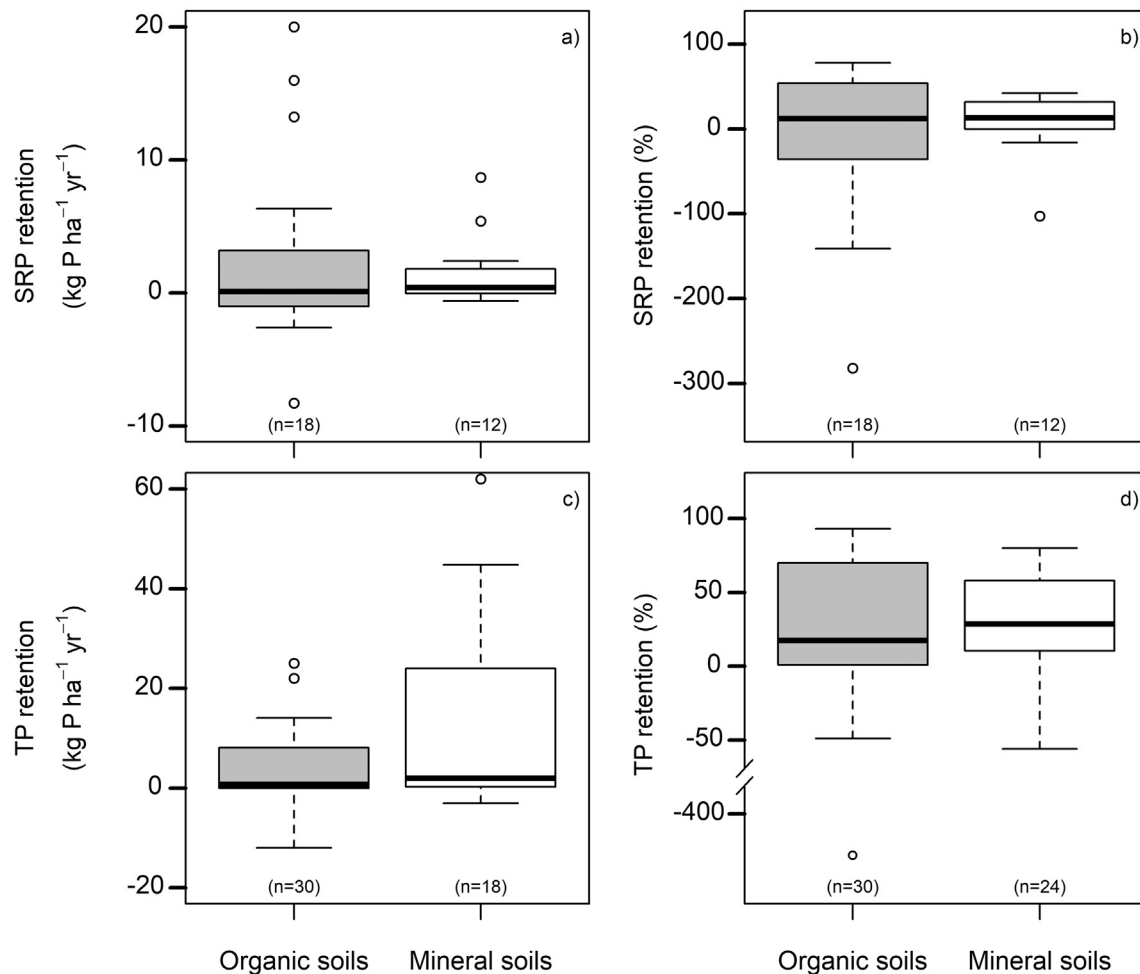
Mean retention of TP in fens with organic soils was  $2.1 \text{ kg P ha}^{-1} \text{ yr}^{-1}$  with an efficiency of 6.4% (24 studies). All but five studies revealed positive retention values. Two wetlands from the same study area exhibited releases of  $-49\%$  and  $-429\%$  (the latter from a very small load) (Kieckbusch and Schrautzer, 2007). Excluding the outlier of  $-429\%$ , the mean retention efficiency in fens reached 25%. Wetlands on mineral soil (18 studies) showed positive retention ( $12.9 \text{ kg P ha}^{-1} \text{ yr}^{-1}$ ) and a mean efficiency of 27% (24 studies) (Fig. 5), with only 5 of 24 studies reporting a release. Mineral soil WBZs with higher loads ( $>10 \text{ kg P ha}^{-1} \text{ yr}^{-1}$ ) exhibited mean retention efficiencies of 34% ( $n = 8$ ) at mean loads of  $53.6 \text{ kg P ha}^{-1} \text{ yr}^{-1}$ , while organic soils WBZs had 16% efficiency at mean loads of  $15 \text{ kg P ha}^{-1} \text{ yr}^{-1}$ . This indicates that P retention can still be high after flooding events and with higher inputs. The SRP results were those that varied most, with 39% of organic soils studies (7 of 18) and 25% of mineral soil studies (3 of 12) reporting a release. Organic soils WBZ had a mean removal efficiency of  $-15\%$  (median 12%) and a positive retention of  $2.6 \text{ kg SRP ha}^{-1} \text{ yr}^{-1}$ , while mineral soils had a mean efficiency of 7.8% and a retention of  $1.5 \text{ kg SRP ha}^{-1} \text{ yr}^{-1}$ . A study of two experimental wetlands in Germany (Lenz and Wild, 2001) demonstrated high retention from very high loads, which increased removal rates and efficiencies significantly, while three studies of fens had efficiencies of more than  $-100\%$  (release). Our results support the suggested risk of SRP release to water bodies by desorption due to redox changes in degraded organic soils (Zak et al., 2004).

### 3.2. What processes and factors influence a WBZ's ability to retain nitrogen?

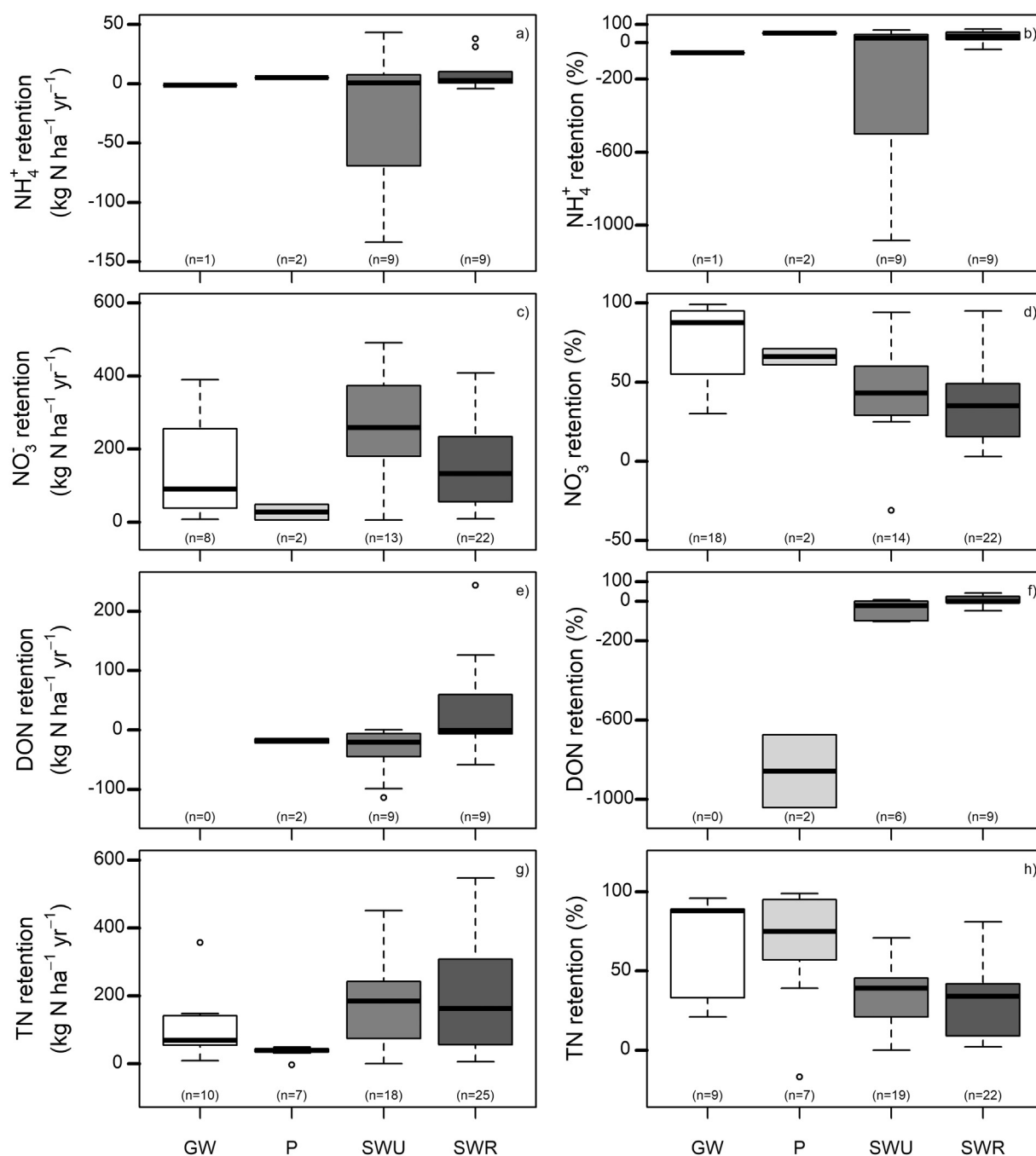
Degraded, mineralised peat materials may release high levels of mobile dissolved N ( $\text{NH}_4^+$ ,  $\text{NO}_3^-$ , DON) (Geurts et al., 2008), while past agricultural application of mineral fertilisers and animal manure (Verhoeven et al., 2006; Kronvang et al., 2007) or groundwater influx from surrounding agricultural areas can cause a nutrient build-up in both organic and mineral soils (Tiemeyer and Kahle, 2014). Relative to water sources,  $\text{NO}_3^-$  removal efficiencies were, on average highest for the ground water path ( $76 \pm 25\%$ ) and lowest if river water was the main water source ( $35 \pm 24\%$ ). The largest absolute removal was found for surface runoff/tile drainage from uplands (Fig. 6). These areas received surface runoff/tile drainage but also had the largest release or lowest retention of  $\text{NH}_4^+$  and DON, causing reduced TN removal efficiency. Thus, the largest  $\text{NO}_3^-$  retentions were counterbalanced by the largest releases of  $\text{NH}_4^+$  and DON, resulting in only slight differences in TN retention and removal efficiency between the different water sources (Fig. 6). The effects of other abiotic and biotic factors often linked with the highly temporally and spatially variable hydraulic regime will be discussed below.

#### 3.2.1. Effect of load saturation

TN and  $\text{NO}_3^-$  retention efficiency was inversely correlated with the load into the WBZ, with high loads reducing the efficiency of removal (Fig. 7). Load saturation was found to be the most important factor



**Fig. 5.** Comparison of absolute retention and retention efficiencies of organic and mineral soils for soluble reactive phosphorus (SRP) and total phosphorus (TP). In the box plots, the bottom and top of the box are the 25th and 75th percentiles, and the band within the box is the 50th percentile (the median); the ends of the whiskers represent the minimum and maximum of all the data and open dots are outliers.



**Fig. 6.** Comparison of absolute retention and retention efficiencies of four different water sources: groundwater (GW), precipitation (P), surface water runoff/drain discharge from upland (SWU), and surface water from the river (SWR) for different nitrogen species ( $\text{NO}_3^-$ : nitrate,  $\text{NH}_4^+$ : ammonium, DON: dissolved organic nitrogen, TN: total nitrogen). In the box plots, the bottom and top of the box are the 25th and 75th percentiles, and the band within the box is the 50th percentile (the median); the ends of the whiskers represent the minimum and maximum of all the data and open dots are outliers.

influencing nutrient retention for TN and  $\text{NO}_3^-$ . It commonly occurred at high loads of  $\text{NO}_3^-$  (Hefting et al., 2006) but was also recorded in a study of a catchment with unusually low discharges over multiple seasons (Tiemeyer et al., 2006). This indicates that the high load of incoming nitrate can effectively ‘use up’ the short-term retention potential of a soil, primarily via denitrification, allowing excess nutrients to pass through the buffer zone. Retention of  $\text{NO}_3^-$  under such circumstances may still be high and ranged between 102 and 359  $\text{kg NO}_3^- \text{N ha}^{-1} \text{yr}^{-1}$ . However, related efficiencies decreased to values lower than 30% (Leonardson et al., 1994; Mitsch et al., 2005; Hefting et al., 2006; Kieckbusch and Schrautzer, 2007; Hoffmann et al., 2012). The trend of substantially reduced retention at higher loads was observed

independently of WBZ type, dominant hydraulic regime and vegetation type. A comparison of N loads with efficiency values implies a N saturation effect (Fig. 7). Two outliers were removed, though; one from a reportedly unique, high organic nitrogen flux post-rewetting, and one from a fen with a geological terrain differing completely from the other studies (Devito et al., 1989; Kieckbusch and Schrautzer, 2007). The regression line crossed the previously mentioned 60% efficiency mark for TN and  $\text{NO}_3^-$  at about 160  $\text{kg N ha}^{-1} \text{yr}^{-1}$ . However, the efficiency differed widely, ranging between 50 and 100% at this but also at higher loading rates (Fig. 7). Still, the regression analysis showed a strong correlation and negative trend ( $p < 0.001$ ;  $r^2 = 0.35$ ). Table 3 displays the retention efficiencies at loads higher and lower than

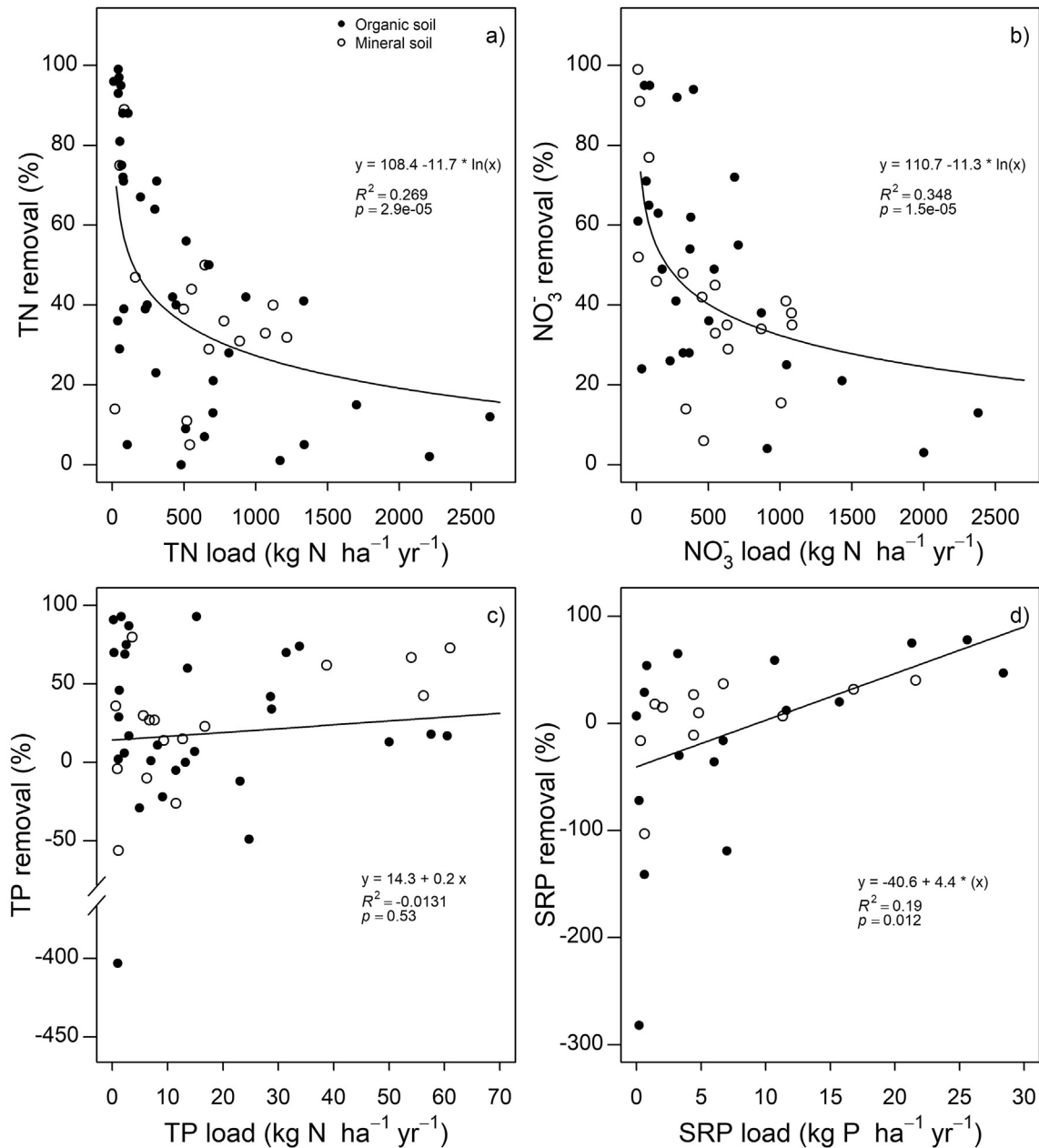


Fig. 7. Relation between load and removal efficiency for total nitrogen (TN), nitrate ( $\text{NO}_3^-$ ), total phosphorus (TP) and soluble reactive P (SRP), respectively for organic and mineral soils.

$160 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ . The discrepancy is clear, with average efficiency rates for TN and  $\text{NO}_3^-$  of 80% and 70%, respectively, for loads below the boundary value and of only 31% and 38%, respectively, for loads above the boundary.

Across all scales, an increase in discharge will coincide with an increase in  $\text{NO}_3^-$ -N loading (or loss from the catchment) (Tomer et al., 2003), which may sometimes be disproportionate (Tiemeyer et al., 2006). One catchment exhibited a low annual retention of  $16 \text{ kg NO}_3^-$ -

$\text{N ha}^{-1} \text{ yr}^{-1}$  with 24% efficiency due to multiple years of low rainfall. In this catchment, N saturation still occurred; however, increased rainfall might have inflated the effect (Tiemeyer et al., 2006). Experimental wetlands that were irrigated with river water and where discharge into the WBZ often occurred were consistently associated with higher nutrient loads and lower retention (Lenz and Wild, 2001; Koskiaho et al., 2003; Mitsch et al., 2005, 2014). High discharge events may promote pulses of nutrients to waterways, for instance of  $\text{NH}_4^+$  and SRP (Reddy

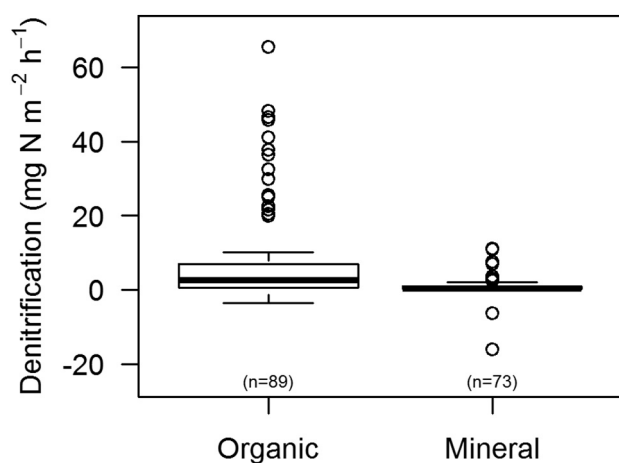
**Table 3**  
Overall results for total nitrogen (TN) and nitrate ( $\text{NO}_3^-$ ) separated by a load of  $160 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  (mean  $\pm$  SD; number of studies in brackets) in kg nutrient per hectare per year.

	Load		Loss	Retention	Efficiency
	kg N $\text{ha}^{-1} \text{ yr}^{-1}$				
TN load	>160	$742 \pm 270$ (12)	$491 \pm 189$ (12)	$205 \pm 190$ (12)	$31 \pm 26$ (12)
	<160	$64 \pm 34$ (15)	$21 \pm 38$ (15)	$48 \pm 21$ (15)	$80 \pm 15$ (15)
$\text{NO}_3^-$ load	>160	$739 \pm 508$ (33)	$381 \pm 229$ (24)	$220 \pm 126$ (32)	$38 \pm 22$ (32)
	<160	$64 \pm 49$ (12)	$21 \pm 24$ (12)	$43 \pm 32$ (12)	$70 \pm 23$ (12)

and D'Angelo, 1994; Kieckbusch and Schrautzer, 2007), which could be related to nutrient buildup in the soils but not directly to fertiliser practices in the catchment (Vinten et al., 1994). The average retention from 36 rewetted fens in agricultural catchments in Denmark was  $122 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  (Hoffmann and Baatrup-Pedersen, 2007; Audet et al., 2020; Petersen et al., 2020a, b). Conversely, two forested buffer zones on mineral soils in North Carolina exhibited 99% and 91%  $\text{NO}_3^-$  removal efficiencies, reportedly with high denitrification removal due to very low inputs of 8 and  $21.5 \text{ kg NO}_3^- \text{ N ha}^{-1} \text{ yr}^{-1}$ , respectively (Jacobs and Gilliam, 1985). However, studies with loads below  $100 \text{ kg NO}_3^- \text{ N ha}^{-1} \text{ yr}^{-1}$  did not always achieve such high retention rates. The reasons for reduced denitrification rates can be unfavourable redox conditions due to decreasing soil water tables, low temperature and/or short hydraulic residence time (Martin and Reddy, 1997; Syversen, 2005; Hill, 2019). Buffer zones with narrower widths allow for shorter hydraulic residence times in soils, although the majority of nitrate is commonly removed from discharging groundwater on the upland side of the WBZ (Peterjohn and Correll, 1984; Cooper, 1990; Schipper et al., 1993; Hoffmann et al., 2006).

### 3.2.2. Denitrification potential in soils

Nitrogen removal has been shown to be generally more efficient in subsurface flow than in surface flow, however N removal efficiencies can vary widely within these two categories (Mayer et al., 2007). Some of this variance may be due to differences in the amount of exchange between the two flow domains – infiltration/exfiltration and diffusional exchange (Petersen et al., 2020b) or due to differences in substrate quality (Lowrance, 1992; McClain et al., 2003). The retention efficiency for  $\text{NO}_3^-$  was higher in organic soils WBZs than in mineral soil WBZs at loads above  $160 \text{ kg NO}_3^- \text{ N}$  (means: 42% ( $n = 19$ ) and 32% ( $n = 13$ ), respectively). This is likely due to increased denitrification rates in degraded peat soils where the organic soils provide sufficient available carbon for microbiological processes (Cabezas et al., 2012). Denitrification is the conversion of  $\text{NO}_3^-/\text{NO}_2^-$  to gaseous  $\text{N}_2/\text{N}_2\text{O}$  occurring under reduced conditions, where facultative, anaerobic bacteria use  $\text{NO}_3^-$  instead of oxygen in the respiration process (Nichols, 1983; Starr and Gillham, 1989), microbially available carbon serving as an electron donor (Cabezas et al., 2013). The comparison of denitrification rates determined under in-situ conditions (Allred and Baines, 2016; Zhou et al., 2017) revealed that average values were about 10 times higher for organic soils ( $8.0 \pm 13.5 \text{ mg N m}^{-2} \text{ h}^{-1}$ ,  $n = 73$ ) compared with mineral soils ( $0.8 \pm 3.1 \text{ mg N m}^{-2} \text{ h}^{-1}$ ,  $n = 89$ , Fig. 8). However, this significant difference ( $p < 0.05$ ) only appeared if organic soils having an organic matter content lower than 30% were



**Fig. 8.** Denitrification rates in organic soils compared to mineral soils. In the box plots, the bottom and top of the box are the 25th and 75th percentiles, and the band within the box is the 50th percentile (the median); the ends of the whiskers represent the minimum and maximum of all the data and open dots are outliers.

included. When the threshold for organic peat soils of 30% organic matter content (Mitsch and Gosselink, 2000) was used in the statistical analyses, no significant differences emerged (results not shown). On the other hand, a  $^{15}\text{N}$ -isotope study showed lack of denitrification for water-saturated soil layers having an organic matter content below 3% (Hoffmann et al., 2000). Accordingly, it can be concluded that also at lower  $\text{NO}_3^-$  loading rates, denitrification would be carbon limited if the organic matter content of soils is lower than 3%. Similarly, a threshold of 2% was recommended in integrated buffer zones for mitigating  $\text{NO}_3^-$  pollution from agricultural runoff (Zak et al., 2018b).

It is difficult to quantify in-situ denitrification rates, but occurrence of denitrification hotspots with amplified rates is presumed (Murray et al., 1995). Denitrification will be carbon limited in mineral soils, but upon interaction with a horizon rich in organic matter (e.g. peat layer or buried river channel) it will become more  $\text{NO}_3^-$  limited, beyond the initially high rates of  $\text{NO}_3^-$  consumption (Hill et al., 2000). Therefore, in degraded peat soils fed by laterally flowing groundwaters,  $\text{NO}_3^-$  retention above 90% should be expected (Hoffmann et al., 2011). A continual  $\text{NO}_3^-$  supply will decrease the loss of both N and DOC to waterways (Aulakh et al., 2000; Davidsson et al., 2002).

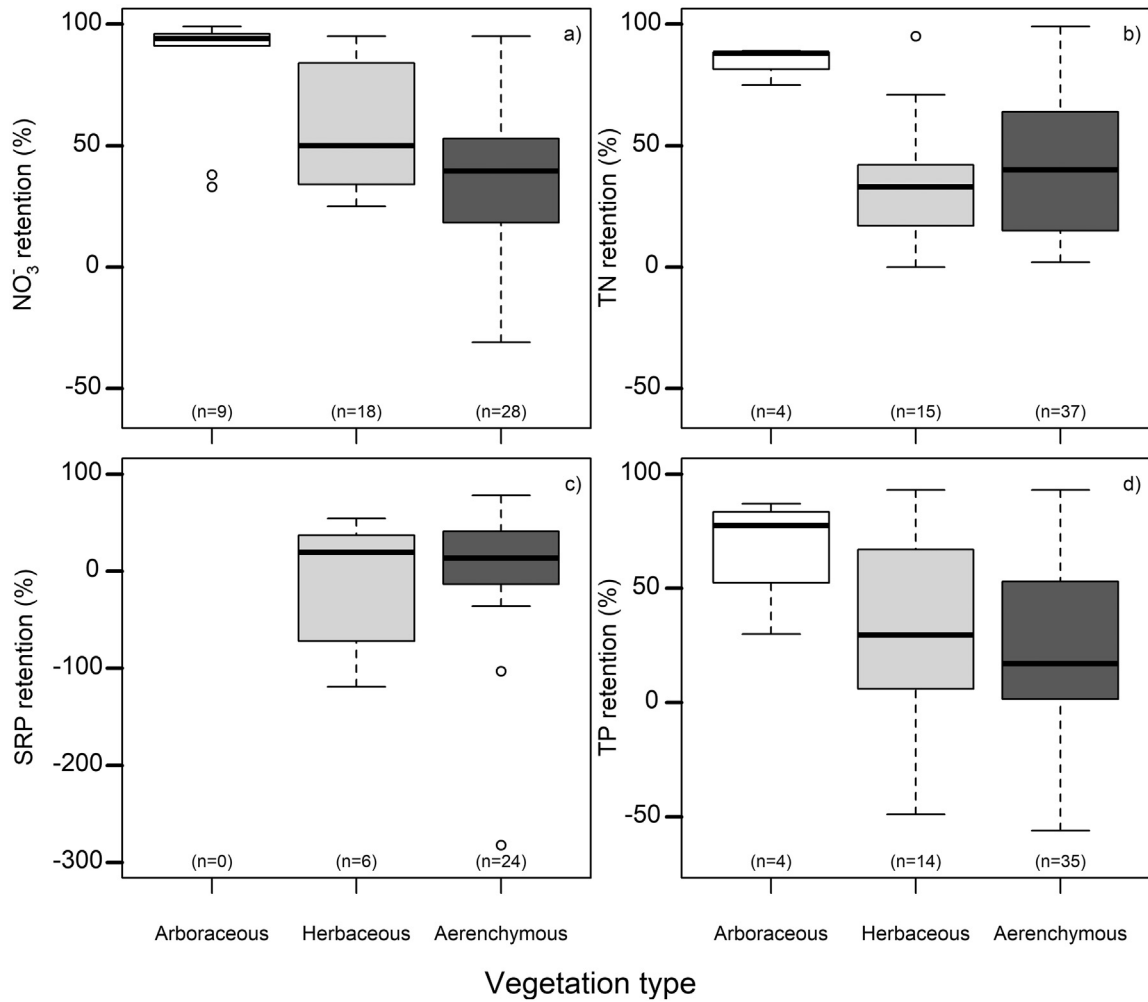
Mineral soils with a low organic matter content and increased aeration create oxic conditions in the upper soil horizons (Pinay et al., 1993), which can lower denitrification rates four- to six-fold (Aulakh et al., 2000) in the surface horizons. Surface horizons often exhibit higher soil organic matter contents than subsurface horizons (Starr and Gillham, 1989), so the combination of oxic conditions and lack of electron donors impedes denitrification in both surface and sub-surface horizons of mineral soils. This can be compounded by lower hydraulic residence times through a more porous medium, in which preferential or rapid diffusion through the subsurface can limit the retention efficiency (Willems et al., 1997). In the absence of organic matter, ferrous iron found in incoming waters can act as an electron donor for denitrification (Hefting et al., 2006). Contrastingly, some subsurface horizons have been found to support significant denitrification (Smith and Duff, 1988; Willems et al., 1997).

Furthermore, seasonality of incoming nutrients and varying hydraulic gradients will result in fluctuating denitrification potentials (Syversen and Borch, 2005; Hoffmann et al., 2006). Winter seasons and higher water tables may lead to reduced soil conditions in the upper soil and promote higher denitrification rates (Pinay et al., 1993). But, contrastingly, high flow rates (i.e. due to snowmelt) can also cause formation of preferential pathways through soils (Väänänen et al., 2008) or they may channelise and direct the water over the buffer zone (Väänänen et al., 2006), thereby significantly reducing the water residence time in the WBZ. Plant structure can also impact denitrification rates, which, as affirmed by the results of this review, are expected to be higher in forest soils than in non-forested soils due to greater organic contents and rooting depths (Osborne and Kovacic, 1993).

### 3.2.3. Effect of vegetation structure

The role of vegetation uptake is uncertain, with some studies questioning its importance (Peterjohn and Correll, 1984) and others showing vegetation to be the primary factor in nutrient retention (Silvan et al., 2004; Syversen, 2005). The results of our review show that WBZs with an arboraceous vegetation structure had consistently higher retention of TN and  $\text{NO}_3^-$  than herbaceous and aerenchymous vegetation types (Fig. 9). Two separate studies (Haycock and Pinay, 1993; Osborne and Kovacic, 1993) reported more effective removal of  $\text{NO}_3^-$  from forested WBZs than from herbaceous WBZs on mineral soils (93% against 83% and 99% against 84%, respectively). In these studies, loads were not reported, but they should be similar among sites of different vegetation classes as the study plots were adjacent.

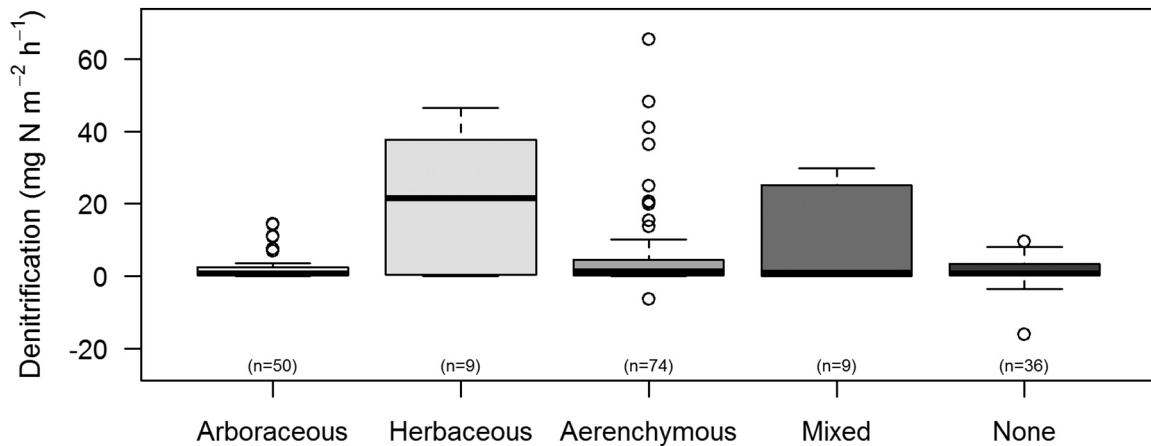
It has previously been reported that forested buffer zones can act as an important nutrient sink (Lowrance et al., 1984). Trees have greater rooting depths, which would help to introduce labile organic matter



**Fig. 9.** Retention efficiency according to three vegetation types (see Table 1 for plant species) for nitrate ( $\text{NO}_3^-$ ), total nitrogen (TN), phosphate ( $\text{PO}_4^{3-}$ ) and total phosphorus (TP). In the box plots, the bottom and top of the box are the 25th and 75th percentiles, and the band within the box is the 50th percentile (the median); the ends of the whiskers represent the minimum and maximum of all the data and open dots are outliers.

into subsurface horizons (Ghestem et al., 2011), particularly those fractions that are located below the water table (Hill, 1996). During the growing season when water tables are low, plant uptake can be dominant in  $\text{NO}_3^-$  removal (Haycock and Pinay, 1993), whereas it will be

negligible in the non-growing season (Osborne and Kovacic, 1993), indicating that elevated water tables enhance denitrification rates (Pinay et al., 1993). As reported for experimental wetlands, depending on the season, the proportion of TN removal by denitrification is



**Fig. 10.** In-situ denitrification rates determined in soils with different vegetation stands (see Table 1 for plant species) extracted from literature of two meta-analyses [Allred and Baines, 2016; Zhou et al., 2017]. In the box plots, the bottom and top of the box are the 25th and 75th percentiles, and the band within the box is the 50th percentile (the median); the ends of the whiskers represent the minimum and maximum of all the data and open dots are outliers.

typically 60–95% relative to the 1–34% assimilated by plants and algae (Lee et al., 2009), although denitrification rates determined in-situ in different vegetation covers did not spot any significant difference (Fig. 10). However, the lowest average rates were found in bare soils ( $1.3 \pm 4.0 \text{ mg N m}^{-2} \text{ h}^{-1}$ ,  $n = 36$ ), while ca. 20 times higher average rates were determined for herbaceous stands ( $20.5 \pm 20.6 \text{ mg N m}^{-2} \text{ h}^{-1}$ ,  $n = 9$ ) (Aldred and Baines, 2016; Zhou et al., 2017). In all vegetation types, there is a tradeoff between lifespan and growth rates, where trees grow slower and herbaceous plants faster (Aerts et al., 1999). Trees have a much longer lifespan than herbaceous plants; however, established trees and mature forests have a lower nutrient requirement than young forests (Emmett et al., 1993), so forest age also affects nutrient uptake rates. Eight of twelve studies in this review on forests on mineral soils showed >90% retention efficiencies for  $\text{NO}_3^-$ , but information on stand age was not included.

### 3.2.4. Other N-related processes within the soil

Some processes are important for rewetted fens but non-significant for natural fens or WBZs on mineral soils. Rewetted fens have greater nutrient accumulation rates than natural fens (Cabezas et al., 2014). Due to altered hydrological and soil conditions, the rewetting process of degraded fens often results in the formation of a novel, shallow lake ecosystem and nutrients are retained in a new 'muck' soil layer (Cabezas et al., 2014; Zak et al., 2018a). The accumulation of nutrients in the sediment is often closely related to net ecosystem production (Chapin et al., 2006). Keeping in mind that helophytes (grouped as 'aerenchymous' vegetation in this study) will rapidly succeed to be the dominant vegetation type in these novel ecosystems (Roth et al., 1999; Zerbe et al., 2013), the major part of the nutrient input into the muck layer will derive from decomposition of helophytes (Cabezas et al., 2014). Wetlands vegetated with helophyte species (e.g., reeds, sedges, cattails) have higher dieback rates than mosses (Aerts et al., 1999), so short-term storage of nutrients in plant tissues will be returned to the surface soil layer where they can be re-mineralised and potentially re-mobilised. Consequently, nutrients are available for re-uptake by growing vegetation or potential release to surface waters.

This dominance of helophytes and the formation of a nutrient-rich detritus mud layer inhibit the formation of typical fen bryophyte communities inter alia via competition for light (Olde Venterink et al., 2002a). Higher microbial activity in the newly formed mud is characterized by high availability of degradable organic carbon and results in higher rates of mineralisation and dissolved inorganic N (DIN) release in the soil (Verhoeven et al., 1987). The mud layer can, however, act as a novel form of nutrient storage within the wetland (Cabezas et al., 2014). Physico-chemical changes of soil characteristics and altered hydrology, such as isolation from natural groundwater flow or frequent water table fluctuations (Kieckbusch and Schrautzer, 2007; Tiemeyer and Kahle, 2014), mean that a return to nutrient-poor, peat-forming conditions will not occur within a short time frame, unless the degraded topsoil becomes removed prior rewetted (Zak et al., 2017).

The process of mineralisation of organic N to  $\text{NO}_3^-$  and  $\text{NH}_4^+$  increases DIN availability and occurs in wetlands with higher primary productivity and a higher organic N (Verhoeven and Arts, 1987) or TN (Reddy, 1982) availability in the soil. Mineralisation rates are commonly up to three times higher in well-aerated soils (Bridgham et al., 1998), so plant available DIN species will be more commonly found in drained or aerated wetland soils with a higher organic content. Higher mineralisation, in combination with reduced denitrification would mean that aerated soils have a higher DIN content than flooded soils or those with a low organic matter content (Olde Venterink et al., 2002b). Previous studies have found that mineralisation rates were higher in forested fens than in herbaceous fens (Verhoeven et al., 1990), and mineralisation and release of  $\text{NO}_3^-$  were found to be higher in forested than in grassed buffer strips, which relates the soil organic content directly to  $\text{NO}_3^-$  release through mineralisation (Osborne and Kovacic, 1993). In addition, oxidation of  $\text{NH}_4^+$  to  $\text{NO}_3^-$  can be a relevant

process in WBZs with high  $\text{NH}_4^+$  load from, for instance, agricultural land fertilised with animal manures. Respiratory reduction of  $\text{NO}_3^-$  to  $\text{NH}_4^+$  and  $\text{N}_2$  under anoxic conditions is a process where denitrifying bacteria reduce available  $\text{NO}_3^-$  (Sørensen, 1978). This can effectively diminish the  $\text{NO}_3^-$  flux from flooded organic soils but increases the  $\text{NH}_4^+$  flux in the process.

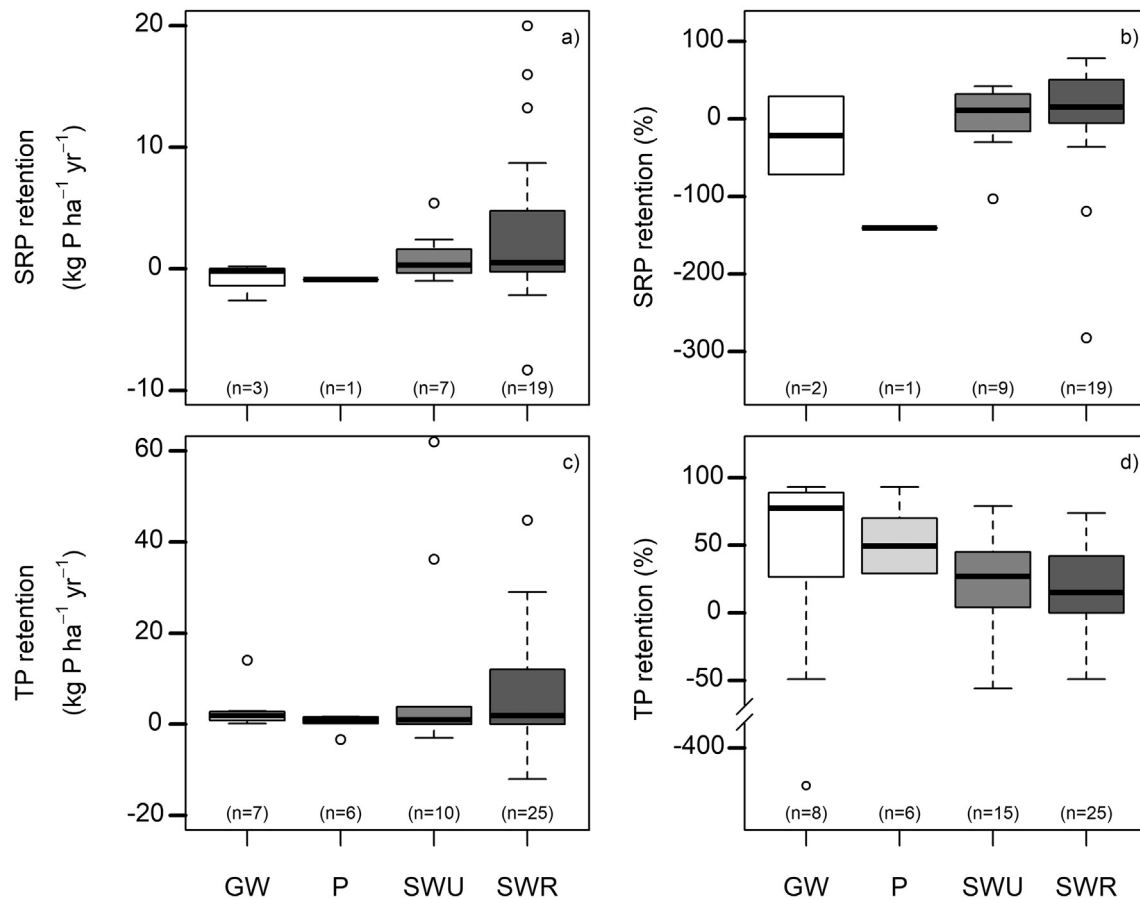
### 3.3. What factors and processes influence a WBZ's ability to retain phosphorus?

It is difficult to predict the retention response of P, especially of SRP in WBZs, due to complex interactions between hydrological and biogeochemical processes of the individual wetland setting (Martin and Reddy, 1997). As 90% of both organic and mineral soil WBZ studies showed positive TP retention, it is suggested that deposition of particulate P through sedimentation has a strong effect on retention (Hoffmann et al., 2009). Other studies assert a direct correlation between WBZ size and TP retention (Uusi-Kämppä et al., 2000; Väänänen et al., 2008). In rewetted fens, such as the 'Karlsmosen Fen' in Denmark, high TP retention occurred during periods of high flow, indicating that the fen was flooded and that sedimentation of particulate P was the main process of retention (Hoffmann et al., 2011). Indeed, the highest TP retention values were reported for sites receiving river water, partly because these areas receive the largest loads (Fig. 11). However, overall, no significant differences appeared for either SRP or TP in relation to the water source. Still, as for N, water saturation coupled with water table fluctuations and redox gradients as well as vegetation are important factors in determining the retention and release of P as will be unraveled in the following sections.

#### 3.3.1. Do WBZ types have different potential phosphorus pollution risks?

Several studies have shown that the recurrence of anoxic conditions in degraded organic soils after rewetting of fens can initiate release of SRP formerly bound to iron III ( $\text{Fe}^{3+}$ ) hydroxides (or other reactive metal oxides) through reduction processes (Lenz and Wild, 2001; Olde Venterink et al., 2002b; Tiemeyer et al., 2007; Zak et al., 2008). Moreover, SRP in pore water of degraded fens has been found to be up to three orders of magnitude higher than in natural settings (Zak et al., 2004, 2010). Furthermore, it has been shown that a high SRP flux risk may persist several years after wetland restoration (Audet et al., 2020). In total, 8 of the 15 studies of rewetted WBZs on organic soils in our review exhibited release of SRP, implying that 53% of rewetted, degraded fens acted as SRP source. WBZs on mineral soils had positive retention in seven out of nine studies, the two studies with negative retention observations coming from shallow flooded WBZs. Riparian buffer zones on mineral soil are expected to have oxic topsoil conditions, effectively negating the SRP flux risk. Further investigation showed that WBZs with low loads (below  $1 \text{ kg SRP ha}^{-1} \text{ yr}^{-1}$ ) exhibited a tendency to negative retention efficiency. Of eight such studies, five showed SRP release. For both organic and mineral soils, the reported losses were small, but due to the low loads the negative retention efficiency was highly exaggerated. For example, mean load was  $0.41 \text{ kg SRP ha}^{-1} \text{ yr}^{-1}$  and mean retention was  $-0.20 \text{ kg SRP ha}^{-1} \text{ yr}^{-1}$ , but efficiencies ranged from  $-282$  to 54%. In contrast, almost all sites with high SRP loads ( $>10 \text{ kg ha}^{-1} \text{ yr}^{-1}$ ) in mineral and organic buffer zones showed positive retention (Fig. 7).

In shallow, flooded fens with oxic surface water conditions ( $<1 \text{ m}$  water depth), soils rich in Fe can create a P barrier at the surface where SRP resorption by metal-hydroxides renders it immobile (Zak et al., 2004). These 'iron-rich fens' have soil Fe:P ratios  $>10$  (Geurts et al., 2008; Zak et al., 2010; Forsmann and Kjærgaard, 2014). Increased sulphate ( $\text{SO}_4^{2-}$ ) input into flooded wetland soils can lead to a further decrease in the Fe binding ability as the reduction of  $\text{SO}_4^{2-}$  to sulphide subsequently increases iron sulphide or pyrite ( $\text{FeS}_x$ ) formation and diminishes Fe availability for binding with P (Lucassen et al., 2004; Zak et al., 2009). In a longer-term



**Fig. 11.** Comparison of absolute retention and retention efficiencies of four different water sources: groundwater (GW), precipitation (P), surface water runoff/drain discharge from upland (SWU), and surface water from the river (SWR) for soluble reactive phosphorus (SRP) and total phosphorus (TP). In the box plots, the bottom and top of the box are the 25th and 75th percentiles, and the band within the box is the 50th percentile (the median); the ends of the whiskers represent the minimum and maximum of all the data and open dots are outliers.

perspective, the trapping of SRP at Fe-rich soil surfaces effectively retains the P in the system in a bioavailable form; so, although it reduces the immediate P flux, it inhibits the overall reduction of nutrients in the system and the succession to nutrient-poor ecosystems (Emsens et al., 2017).

Seasonal variations in load can affect P retention processes. During periods of high inflow, hydraulic contact and residence times are reduced, diminishing the soil's ability to retain nutrients through plant uptake, and in these periods chemical sorption to soil mineral complexes is likely to be the dominant form of retention (Väänänen et al., 2006). Longer water residence times within the WBZ correlate with higher nutrient retention rates and can contribute to high P retention rates, especially in soils with a high P sorption capacity, provided that the soils do not become anaerobic (Koskiahho et al., 2003). Thus, in the Vechtplatten area of the Netherlands, one rewetted fen was fed with minerotrophic groundwaters and had an annual retention rate of 0.16 kg SRP ha<sup>-1</sup> and a retention efficiency of 29%, while an adjacent fen fed by ditch water released 0.48 kg SRP ha<sup>-1</sup> yr<sup>-1</sup> with a retention efficiency of -280% (Koerselman et al., 1990). In the first example, the incoming groundwater was rich in Fe, Mg and Ca, which negated higher P inputs through a higher sorption capacity (Verhoeven and Arts, 1987), and 98% of the groundwater-derived P was removed (Koerselman et al., 1990). Interestingly, five WBZ fed artificially with river water showed a positive retention efficiency for SRP (mean = 47%) with high loads (Mitsch et al., 2014). This indicates that higher amounts of incoming SRP in non-fen buffer zones may lead to a greater potential for removal.

### 3.3.2. Effect of phosphorus load saturation

TP showed no correlation (Pearson's test:  $p = 0.53$ ;  $r^2 = -0.01$ ) between load and efficiency (Fig. 7), with the majority of studies reporting positive TP retention. However, SRP showed a significant positive correlation between retention and load (Pearson's test:  $p = 0.012$ ;  $r^2 = 0.012$ ). The regression line crossed 0% efficiency at about 10 kg SRP ha<sup>-1</sup> yr<sup>-1</sup> (Fig. 7). This is due to the influence of the experimental sites pumped with P-rich river water and showing good retention compared with minerotrophic peatlands fed primarily with groundwater where loads were lower but the potential for P release higher (Hoffmann et al., 2009, 2011). Organic soil WBZ were slightly less efficient in retaining TP than mineral soil buffer zones at lower loads (7% against 16%), but similar at loads over 10 kg P ha<sup>-1</sup> yr<sup>-1</sup> (Fig. 7). Higher retention at higher loads is likely related to increased sedimentation and hence particulate P deposition at higher flow rates.

### 3.3.3. Effect of vegetation structure on phosphorus retention

Most studies found a higher mean retention efficiency for TP in arboraceous (68%) than in herbaceous wetlands (26%). This contrasts the findings of Osborne and Kovacic (1993), who found higher concentrations of TP (and total dissolved P) throughout the year on the stream side of arboraceous WBZs (on organic soils) compared with herbaceous WBZs (on mineral soils), indicating temporary storage of P as well as release over the longer term. This release is related to potentially higher P mineralisation rates in the forest mineral soils, which have a higher organic matter content. The contrasting findings of our review might be due to a difference in the number of studies, arboraceous wetlands being included in only four studies and herbaceous wetlands in 14,

creating great diversity of results. The mean loads for forests were four-fold lower than for grasslands and three-fold lower than for helophyte dominated wetlands, which would further resulting in varying retention efficiency.

The formation of a muck soil layer in shallow, flooded fens is also important for P retention. Helophytes remove P from the soil water, incorporate it into growing parts and deposit it on the soil surface upon dieback, where it is then decomposed (Cabezas et al., 2014). A term previously used for this process was “smuggling of nutrients” above the redox barrier, which leaves nutrients available for remobilisation and release to surface waters, allowing SRP to effectively bypass the soil surface (Zak et al., 2014). Upon plant death, the litter is returned to the soil surface and between 35% (Richardson and Marshall, 1986) and 72–90% (Zak et al., 2014) of the P can be made available in surface waters through leaching or decomposition. Sedimentation will also introduce particulate P into this new soil layer from flooding and surface water input (Hoffmann et al., 2009).

Load concentrations of SRP are an important factor in determining retention rates in biomass. An important study by Richardson and Marshall (1986) found that at low levels of P input (5–10 kg SRP ha<sup>-1</sup> yr<sup>-1</sup>), much of the available P is retained in the litter-microorganism compartment (LMC) through chemical sorption. Uptake from plant growth was of limited importance for retention at low input levels. Higher levels of P input (20–170 kg ha<sup>-1</sup> yr<sup>-1</sup>) led to a decrease of the LMC removal efficiency to 22%, with *Carex spec.* removing up to 61% of the added P, of which 81% was stored in the roots and rhizomes. Uptake in the LMC, primarily by fungi, reflects the short-term responses, and if the impact of LMC decreases, chemical sorption will immobilise the remaining available P if geochemical conditions are suitable. Furthermore, if there are seasonal variations in nutrient loads and the majority of nutrient input occurs outside the growing season, then the plant uptake will be almost negligible (Richardson and Marshall, 1986). Plants have lower uptake rates over longer time scales, so a low abundance of P in soils will restrict the uptake ability of the plants due to the rapidly diminished nutrient supply (Richardson and Marshall, 1986). Plant and microorganism uptake is considered a transient P pool as the P is either eventually released back into the soil via decomposition or is dependent on equilibria in the soil (Verhoeven et al., 1990). Only peat formation (with bryophytes or roots from helophytes) under permanently reduced conditions or stable geochemical compound formation like vivianite (Fe<sub>3</sub>(PO<sub>4</sub>)<sub>2</sub>·8H<sub>2</sub>O) are considered viable modes of long-term P storage (Rydin and Jeglum, 2006; Rothe et al., 2016).

#### 3.4. Can harvesting successfully alter the nutrient budget of a WBZ?

If the denitrification potential is limited by nitrate saturation and the restored buffer zones are enriched with P from past fertilising practices, harvesting of vegetation could become an important source of export of N and P from the wetland. Koerselman et al. (1990) stated that “Harvest operations provide an important tool for manipulating the nutrient budget of fens”, and they reported TN uptake and removal rates in a rewetted, groundwater fed fen of up to 66 kg N ha<sup>-1</sup> yr<sup>-1</sup> by harvesting of *Carex sp.* and *Phragmites sp.* This particular fen had an N load of 75.6 kg N ha<sup>-1</sup> yr<sup>-1</sup> (including N fixation and atmospheric N deposition), so harvesting effectively balanced inputs. Despite the high removal through harvesting, this fen still exhibited an output of 21.2 kg N ha<sup>-1</sup> yr<sup>-1</sup>, for which there are four possible explanations: 1) some of the incoming N passed uninterrupted through the WBZ and the excess N uptake was removed from the soil N pool, 2) soil processes, for example mineralisation, released DIN to the water, 3) mineralisation was ineffective and organic N was released, and 4) elevated atmospheric N deposition. The other fen in this study fed by surface water returned a harvest of 37.6 kg N ha<sup>-1</sup> yr<sup>-1</sup> at a load of 53 kg N ha<sup>-1</sup> yr<sup>-1</sup>. Three other studies involving harvesting of

vegetation in the Netherlands reported TN retention efficiencies of 93%, 97% and 99% (Koerselman, 1989; Wassen and Olde Venterink, 2006).

The two fens from the Koerselman et al. (1990) study had low TP inputs but high harvest results, with loads of 1.3 and 1.1 kg P ha<sup>-1</sup> yr<sup>-1</sup> and harvests of 5.6 and 3.9 kg P ha<sup>-1</sup> yr<sup>-1</sup>, respectively. This might have led to depletion of the available P in the soil. A fen study in north-eastern Germany indicated a post-rewetting release of P from degraded peat to pore water of approximately 2.7 ± 0.6 g P m<sup>-2</sup> (median ± standard error, n = 30) within a growing season of 150 days matching the nutrient uptake rates between 1.1 and 3.0 g P m<sup>-2</sup> by the helophyte species *Phragmites australis*, *Glyceria maxima* and *Typha latifolia*, respectively (Zak et al., 2014). This comparatively high P uptake concurred with findings from other European wetlands (Dykyjová, 1978; Tanner, 1996; Headley et al., 2003). High plant removal rates can result in P release from desorption and incoming waters and contribute as well to depletion of the P soil pool. An overview of nutrient stocks, which can be used as a rough estimate of the net nutrient uptake by selected dominant plant species, and by mixed plant assemblages occurring at various succession stages of wetland rewetting, is given in Table 4.

In the initial fen rewetting stages, N will be the key limiting nutrient for plant growth (Koerselman, 1989). If N inputs to the buffer zone are low, then the primary productivity of the WBZ will be correspondingly low, meaning that harvesting primarily accounts for the atmospheric N deposition and not for the soil N pool (Olde Venterink et al., 2002a). WBZs with high productivity have the ability to remove surplus nutrients from the soil pools, sometimes in the order of 3% annually of the soil N pool in the topsoil (Olde Venterink et al., 2002a). As outlined in the previously mentioned studies, plant growth will remove considerable amounts of available P and potassium from the soil (Koerselman, 1989; Koerselman et al., 1990), so continued harvesting of biomass from the WBZ will eventually make P the greater growth-limiting nutrient (Verhoeven and Schmitz, 1991). These studies targeted fens, but the same theorem applies to WBZs on mineral soils exhibiting nutrient enrichment. Harvesting can be employed to reduce nutrient buildup in the topsoil; however, management and continued reduction of nutrient inputs from the catchment are important in the longer term to help restore targeted plant communities and more natural soil conditions.

Estimates of the return to nutrient-poor conditions required for revegetation with original peat-forming plant species range between 20 and 50 years for rewetted fens (Zak et al., 2014). Harvesting is most effective at the beginning of flowering period when biomass and P concentrations are at their peak (Meulemann et al., 2002). Early harvesting may pose difficulties in mixed species assemblages, but dominance of a single species in larger areas (>1 ha) with low abundances of other species is often observed (Zak et al., 2014). Nutrients are taken up by plants at a slower rate than water-flow occurs and thus, will not affect short-term nutrient fluctuations (Richardson and Marshall, 1986). In the longer term, harvesting of vegetation contributes to reducing soil trophy, and it will prevent succession into forested ecosystems (Koerselman et al., 1990; Grootjans et al., 2006). As consequence the buffer zone structure remains open promoting favorable ground-light conditions inter alia for mosses or low sedges (Middleton et al., 2006). On the banks of regularly maintained brooks and rivers, elevated accumulation of organic material due to deposition of mown plants is often observed. Removal of this “topsoil” not only improves the nutrient status of buffer zones but also increases the potential of flooding.

#### 4. Summary and recommendations

The efficient removal of TN and TP demonstrated by almost all the studies investigated in this review suggests that the role of WBZs ought to be recognised in large-scale, long-term pollution management. Removal of nitrate was found to be similar at lower load rates when

**Table 4**  
Nutrient stocks (in  $\text{g m}^{-2}$ ) of dominating plant species and plant communities of wetland buffer zones. Plant species data represent (range of) mean values ( $n$  = number of study sites) derived from literature. Data of plant communities represent ranges ( $n$  = number of sampling spots) and are obtained from 10 rewetted wetland buffer zones in the Region of Southern Denmark in the Kratholm catchment (unpublished data). The harvesting time was during the growing season, mostly before of the flowering time (May until August).

Species/communities	Nitrogen stock		Phosphorus stock	
	Organic	Mineral	Organic	Mineral
Hybrid poplar/stems*		13.7–48.0 ( $n = 4$ ) <sup>1</sup>		1.4–5.7 ( $n = 4$ ) <sup>1</sup>
Hybrid poplar/branches*		9.1–36.4 ( $n = 4$ ) <sup>1</sup>		1.1–4.4 ( $n = 4$ ) <sup>1</sup>
<i>Salix viminalis</i>		20.1–45.8 ( $n = 2$ ) <sup>2</sup>		3.0–7.0 ( $n = 2$ ) <sup>2</sup>
<i>Alnus glutinosa</i>		1.9–2.0 ( $n = 2$ ) <sup>2</sup>		0.2 ( $n = 2$ ) <sup>2</sup>
<i>Phalaris arundinacea</i>	7.1–26.9 ( $n = 23$ ) <sup>3,4,5,6</sup>	4.5–10.9 ( $n = 5$ ) <sup>7,8</sup>	1.6–3.4 ( $n = 23$ ) <sup>3,4,5,6</sup>	0.8–1.5 ( $n = 2$ ) <sup>8</sup>
<i>Glyceria maxima</i>	12.3 ( $n = 5$ ) <sup>4</sup>	11.5 ( $n = 1$ ) <sup>9</sup>	2.8 ( $n = 5$ ) <sup>4</sup>	
<i>Typha latifolia</i>	18.7 ( $n = 5$ ) <sup>4</sup>		2.9 ( $n = 5$ ) <sup>4</sup>	
<i>Carex riparia</i>	9.7–11.6 ( $n = 7$ ) <sup>3,4</sup>		1.3–1.5 ( $n = 7$ ) <sup>3,4</sup>	
<i>Carex acutiformis</i>	8.0 ( $n = 5$ ) <sup>4</sup>		1.1 ( $n = 5$ ) <sup>4</sup>	
Herbaceous buffer**		2.1–4.9 ( $n = 4$ ) <sup>1</sup>		0.3–0.6 ( $n = 4$ ) <sup>1</sup>
Reed beds	3.3–29.5 ( $n = 18$ )		0.4–4.7 ( $n = 18$ )	
Dry fallow fields	4.7–17.9 ( $n = 5$ )		0.6–1.8 ( $n = 5$ )	
Fen-sedge beds	5.7–25.1 ( $n = 5$ )		0.7–3.8 ( $n = 5$ )	
Humid tall herb fringes	8.2–28.1 ( $n = 5$ )		2.0–4.7 ( $n = 5$ )	
Moist fallow fields	3.8–14.4 ( $n = 6$ )		0.8–1.3 ( $n = 6$ )	
Rich fens	6.1–21.3 ( $n = 6$ )		0.7–3.1 ( $n = 6$ )	
Humid grasslands	14.8–26.0 ( $n = 2$ )		2.7–3.7 ( $n = 2$ )	
Mesophile grasslands	4.7–25.1 ( $n = 3$ )		0.7–2.1 ( $n = 3$ )	

\* *Populus deltoides* × *nigra* (DxN-3570); *P. canadensis* × *maximowiczii* (DNxM-915,508); *P. maximowiczii* × *balsamifera* (MxB-915311).

\*\* *Phleum pratense*, *Agropyron repens*, *Agrostis* spp., *Vicia cracca*, and *Solidago* spp.

<sup>1</sup> Fortier et al. (2015).

<sup>2</sup> Zak et al. (2019).

<sup>3</sup> Schulz et al. (2011).

<sup>4</sup> Steffenhagen et al. (2008).

<sup>5</sup> Saijonkari-Pahkala (2001).

<sup>6</sup> Mander et al. (2012).

<sup>7</sup> Landström et al. (1996).

<sup>8</sup> Heinsoo et al. (2011).

<sup>9</sup> Nijburg et al. (1997).

comparing WBZs on mineral and organic soils, while at higher load rates, WBZs on organic soils were more efficient at removal. The detailed consideration of water sources indicated that average nitrate removal efficiencies were approximately twice as high for ground water compared to river water. No significant pattern for P retention emerged; however, the highest absolute removal appeared if the P source was river water. Denitrification is the dominant factor in the removal of  $\text{NO}_3^-$  inputs into the buffer zone, while high loads and saturation of this process are the main factors behind reduced rates of N retention. On mineral soils with high  $\text{NO}_3^-$  input, denitrification is limited by carbon availability and restricted by seasonality and the depth of the water table. In contrast, in restored fens and restored wetland settings rich in organic matter, denitrification is limited by the  $\text{NO}_3^-$  supply and the entire soil is generally saturated (Davidson and Swank, 1986). As high loads reduce nutrient retention efficiency in wetlands, close management of agricultural practices and optimisation of fertiliser use should be considered in conjunction with restoration using WBZs.

Understanding nutrient inputs, hydrology, soil characteristics, plant succession and setting realistic restoration goals, are necessary to determine the required steps in a WBZ restoration program. Assessing the tradeoff between lifespan and growth rates of herbaceous and arboraceous vegetation cover or the higher nutrient use efficiency and lower nutrient input back into the soil from evergreen trees compared with deciduous trees is important (Aerts et al., 1999). Reed and sedge plants can be harvested and used as building materials, for paper production or for bioenergy (Thevs et al., 2007) in a process called ‘wet agriculture’ (on mineral soils) or ‘paludiculture’ (on organic peat soils), where yields can be reintroduced into energy, construction or fodder as circular economy value chains (Ziegler, 2019). This may, at least partly, cover harvesting costs and provide compensation for flooded land that is no longer available for conventional agricultural uses (Wichtmann and Schäfer, 2007). However, it should be noted that conflicts may arise if ecologically driven measures meet economic

ambitions (Turner et al., 2000). Thus, from the perspective of nature conservation, a different harvesting regime regarding time, extent and techniques may be better than that used to fulfill economic interests, but this requires further evaluation. The risk of limited N and P retention in WBZs with large loads also shows that restoration options have to be integrated with good agricultural practices to promote cross-compliance with water protection legislation such as the EU Water Framework Directive.

Despite long-term research on the processes and factors driving P and N turnover in wetlands, knowledge gaps still exist. Briefly summarised, these are:

- on nutrient balances for both drained and natural wetlands,
- on the importance of hydrology and specific biogeochemical processes and their changes over time in the course of the wetland restoration,
- and on the long-term effects of harvesting management on certain soil nutrient fractions like organic bound N or P, including legacy P, and plant litter quality.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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### Author contributions

CW and DZ conceptualized the study. CW, JA, JL and RJP performed the data analyses. CW and DZ compiled the manuscript and all co-authors contributed to the text.

### Data availability statement

The datasets generated and analysed for this study can be found in an electronic supplement to this article (Data\_Supplement-Walton\_et\_al.xlsx; link to STOTEN website).

### Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2020.138709>.

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