



Mitigating agricultural nitrogen load with constructed ponds in northern latitudes: A field study on sedimental denitrification rates

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ABSTRACT

Constructed agricultural ponds and wetlands can reduce nitrogen loading from agriculture especially in areas where warm climate predominates. However, in cold climate temperature-dependency of microbiological processes have raised the question about the applicability of constructed wetlands in N removal. We measured in situ denitrification rates in a constructed agricultural pond using ¹⁵N-isotope pairing technique at ambient light and temperature throughout a year as well as diurnally. The field IPT measurements were combined with a wide set of potentially important explanatory data, including air temperature, photosynthetically active radiation, precipitation, discharge, nitrate plus other water quality variables, sediment temperature, oxygen concentration and penetration depth, diffusive oxygen uptake and sediment organic carbon. Denitrification varied, on average, diurnally between 12 and 314 $\mu\text{mol N m}^{-2} \text{h}^{-1}$ and seasonally between 0 and 12409 $\mu\text{mol N m}^{-2} \text{d}^{-1}$. Light and oxygen regulated the diel variation of denitrification, but seasonally denitrification was governed by a combination of temperature, oxygen and turbidity. The results indicated that the real N removal rate might be 30–35% higher than the measured daytime rates, suggesting that neglecting the diel variation of denitrification we may underestimate N removal capacity of shallow sediments. We conclude, that by following recommended wetland:catchment – size ratios, boreal agricultural ponds can efficiently remove nitrogen by denitrification in summer and in autumn, while in winter and in spring the contribution of denitrification might be negligible relative to the loading, especially with short residence time.

1. Introduction

Humans have transgressed the planetary boundaries in the fixation of N₂ (Rockstrom et al., 2009) by doubling the global amount of reactive nitrogen (N_r) (Fowler et al., 2013; Gruber and Galloway, 2008; Sutton et al., 2011). In Europe, the amount of N_r has been tripled and it is estimated that 40–70% of the fertilizer N_r applied for cereal production is lost to the environment (Sutton et al., 2011). Excess N leaching in receiving waterways has resulted in eutrophication and reduced water quality for drinking, agricultural, recreational, and other purposes (Galloway et al., 2013; Robertson and Vitousek, 2009). Only about half of the European surface waters met the Water Framework Directive (WFD) objective of good ecological status in 2015 and 25% of the ground waters investigated suffered from excess nitrate-N (NO₃⁻) mainly caused by agriculture (EEA, 2015). Moreover, at the same time with the increased amount of N_r, the globe has lost more than half of its natural wetlands (Davidson, 2014).

Constructed wetlands (CWs) have been successfully applied to remove excess nitrogen (N) from agricultural runoffs (e.g. ÓGreen et al.,

2010; Strand and Weissner, 2013; Vymazal, 2017). CWs have been shown to remove N in warm climates, but whether they work in cold conditions, has still been called into question (Arheimer and Pers, 2017; Wang et al., 2018). In Finland, some non-remunerative investments, e.g. agricultural CWs, are economically supported by the EU and national legislation, and over a thousand wetlands have been built since 1995. Furthermore, the number of smaller pond systems, e.g. individual or chains of sedimentation ponds is likely to be even greater. Agricultural wetlands and ponds are often reasonably non-vegetated, due to highly turbid agricultural waters (e.g. Tikkanen et al., 1985) limiting light penetration, and routine management practices like mechanical excavation. Boreal sedimentation ponds are considered to exhibit insignificant and/or highly variable N-retention, being more efficient in removing solids and phosphorus (P) (e.g. Häikiö, 1998; Vuollekoski et al., 2015). However, study reports from Sweden conclude that CWs can have a high N removal, although it varies considerably (Strand and Weissner, 2013). Whether the reports of low N-retention results from insufficient retention time, or negligible N removal processes in cold climate, is poorly understood.

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Three natural processes contribute to overall N retention in freshwater wetlands: denitrification, sedimentation, and assimilation by aquatic biota. However, denitrification is the only pathway that removes N entirely from the aquatic ecosystems. Denitrification is an anaerobic microbial process, where nitrate nitrogen (NO_3^-) is reduced into gaseous form, either into nitrous oxide (N_2O) or nitrogen gas (N_2) (Mitsch and Gosselink, 2015). These gases are transferred into the atmosphere, balancing the natural and anthropogenic N input. Denitrifiers utilize organic carbon (C) (heterotrophic denitrification) or reduced inorganic compounds (e.g. sulfides; autotrophic denitrification) as energy sources. Autotrophic denitrification is typical to marine environments (Shao et al., 2010), while heterotrophic denitrification is considered to be the dominating process in freshwater ecosystems (Mulholland et al., 2008).

Denitrification can be based on the NO_3^- from the water above the sediment (D_w), and/or from the coupled nitrification-denitrification process (D_n), occurring in the oxic layers of the upper sediment. In shallow sites with benthic primary production, higher sediment oxygen concentration followed by increased photosynthetically active radiation (PAR) can promote D_n (An and Joye, 2001; Lorenzen et al., 1998; Risgaard-Petersen et al., 1994), while D_w can be lower during light hours (Christensen et al., 1990; Risgaard-Petersen et al., 1994). In boreal environments, where PAR amount changes significantly between seasons (Lakkala et al., 2016), light-induced changes in the N removal of shallow wetlands may be highly important. Besides changes in PAR and accompanying oxygen conditions, temperature has been found to govern denitrification activity, explaining variable denitrification rates in boreal lakes (Holmroos et al., 2012; Rissanen et al., 2011), temperate wetlands (Bastviken et al., 2007; Hernandez and Mitch, 2007) and temperate stream sediments (de Klein, 2008; Veraart et al., 2014).

Current estimates of denitrification rates in agricultural wetland and stream sediments (e.g. Castaldelli et al., 2015; Pinardi et al., 2009; Roach and Grimm, 2011) are based on laboratory incubations conducted in dark at constant temperature. Because of this the results do not necessarily reflect the real, in situ denitrification rates. Moreover, sampling has been targeted only on certain seasons like summer and on daytime. This paper reports in situ denitrification rates at ambient light and temperature conditions in a constructed agricultural pond of northern Europe, Finland. The field measurements were performed throughout the year, as well as diurnally. Using the direct ^{15}N -isotope pairing technique (IPT, Nielsen, 1992) simultaneously with sediment oxygen profiling, allowed us to study the role of different environmental factors controlling denitrification rates at different temporal and spatial scales. We expected ambient light and temperature regime being important drivers of denitrification on annual basis in boreal agricultural ponds, which typically have high nutrient concentrations throughout the year. Furthermore, we hypothesized that denitrification rates may also show similar variation diurnally. On the basis of diel results, we recalculated the measured seasonal N removal in the sediment of a boreal agricultural pond.

2. Materials and methods

2.1. The catchment and the study site

The study was conducted in an agricultural watershed in southern Finland (61°04'97"N, 25°02'89"E) (Fig. 1). Koiransuolenoja is an approximately 4 km long brook flowing through typical agricultural catchment (6.8 km²) into Lake Pääjärvi. The brook is approximately 1–2 m wide with an average depth less than 0.5 m. The stream is heavily impacted by farming, as agricultural land covers up to 24% of the drainage area (Arvola et al., 2015). Nearly half of the catchment surface area soil is easily erodible material e.g. sand and silt (Tikkanen et al., 1985). During the study period (July 10th 2014–June 25th 2015), the average $\text{NO}_x\text{-N}$ (indicating the NO_2^- plus NO_3^-) concentration in the brook was 194 $\mu\text{mol l}^{-1}$ (SD $\pm$ 58 $\mu\text{mol l}^{-1}$, n = 48).

An agricultural sedimentation pond had been built one year earlier in March 2013. Aquatic vegetation had not yet developed into the littoral, and no shading was provided by trees or shrubs. Surface area of the pond was 320 m² and volume 226 m³ (mean depth 0.7 m, max. depth 1.6 m). Average discharge in Koiransuolenoja was 0.058 m³ s⁻¹ resulting in theoretical residence time approximately one hour in the pond. The real residence time varied from 15 min to 4.5 h depending on the discharge. Water flow in different parts of the pond was measured with a flow meter (MiniAir2, Schiltknecht) and the flow rate in the pond littoral was 0 m s⁻¹ on each experiment date throughout the study. We investigated the grain size of the homogenized and dried (48 h, 60 °C) top sediment (0–3 cm) with a vibratory sieve shaker (Analysette 3, Fritsch, Germany). The shallow littoral where denitrification was measured (Fig. 1) consisted of accumulation sediment having the highest amounts (91%) of fine materials, fine sand (grain size 0.063–0.125 mm) and silt (0.002–0.062 mm). In the deepest part of the pond, 64% of the top sediment was fine materials. Two species of non-nitrogen-fixing benthic algae *Spirogyra* and *Planktothrix* sp. was almost every time observed on the sediment surface.

2.2. Sampling, field and laboratory analyses

Sediment samples and water $\text{NO}_x\text{-N}$ samples for denitrification measurements were taken manually from the littoral zone of the constructed pond (Fig. 1). Quality of the stream water (turbidity, dissolved organic carbon (DOC), $\text{NO}_x\text{-N}$, ammonium ($\text{NH}_4^+\text{-N}$) and total N (TN)) was investigated weekly from water samples taken at site K1 (Fig. 1), representing the water quality at catchment scale. Discharge was calculated from the discharge curve based on the measured water level and flow rate. Data for air temperature and precipitation measured at Lammi biological Station (Fig. 1), 800 m NE from the study site, were obtained from The Finnish Meteorological Institute. Photosynthetically active radiation (PAR) was measured with a quantum sensor (PQS1, Kipp & Zonen) at 10-min intervals, located 600 m NE from the study site (Fig. 1). In laboratory, $\text{NO}_x\text{-N}$, $\text{NH}_4^+\text{-N}$, and TN concentrations were analyzed using standard laboratory methods (see Arvola et al., 2015). Samples for DOC were filtered through pre-rinsed cellulose ester filters (pore size 0.45 μm , Millex-HA, Merck Millipore) and analyzed using a carbon analyzer (Ordior TOC-V, Shimadzu). Organic C content of the study littoral sediment (0–3 cm) was calculated from the loss on ignition (LOI%) of oven dried material (550 °C, 2 h). Temperature was measured from the top of the sediment using the flow meter. Oxygen (O_2) penetration depth in the sediment (OPD) and O_2 concentration on top of the sediment were measured with Clark-type microelectrode (tip $\varnothing$ 100 μm) in the laboratory from three replicate intact sediment-water cores within one hour of sampling (OX100-sensor, PA-2000, Unisense). Sediment diffusive O_2 uptake (DOU) was calculated from the flux through the diffusive boundary layer above the sediment from the oxygen profiles (Jørgensen and Revsbech, 1985; Revsbech et al., 1980).

2.3. Sediment core incubations and isotope analysis for denitrification

Before the field incubations, the validity of the method at the study site (independency of denitrification of ambient $\text{NO}_x\text{-N}$ and positive dependency of denitrification of ^{15}N -labeled nitrate on the added tracer amount) and the possible presence of anammox (Nielsen, 1992; Risgaard-Petersen et al., 2003), was investigated in a pre-experiment with a concentration series in the laboratory using ^{15}N -labeled potassium nitrate (50, 100, 200, 400, and 600 μmol of K^{15}NO_3 , 98 atom%, Sigma-Aldrich).

Denitrification experiments were performed at the pond littoral at water depth of 30–40 cm. Sediment cores were incubated in situ under ambient light and temperature conditions, using ^{15}N -isotope pairing technique (IPT) by Nielsen (1992). Diel denitrification rates were measured twice, on the 28th and 25th of August in 2014 and 2015, respectively. Three undisturbed replicate sediment samples were

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