Evaluating the potential for denitrification in permeable interlocking concrete pavements

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This research explored the denitrification potential of a submerged zone incorporated into the present permeable interlocking concrete pavement (PICP) design. The following three main factors controlling denitrification were investigated through a laboratory study: (i) detention time, (ii) inclusion of a carbon source (newspaper), and (iii) submerged zone depth. The study used 10 columns, each fully packed with 50– 63 mm washed aggregate and including a 1.5 m deep submerged zone. Columns were paired according to detention time (1, 2, 5, 10 and 'varied' days) with one set including newspaper, the other set not. All columns were loaded with synthetic stormwater over a 4-month period. Samples were taken from different submerged depths (0, 300, 600, 900, 1200 and 1 500 mm) and analysed for concentrations of ammonia (NH₃), nitrate (NO₃⁻) and phosphate $(PO₄³⁻)$ every 10 days. This study found that 10 – or more – days detention and the provision of a carbon source had the most significant impact on denitrification – providing an overall mean NO₃⁻ and nitrogen removal above 41% and 59%, respectively. Moreover, a submerged depth of 300 mm was sufficient to achieve a minimum NO₃[−] removal of 41% in columns which included a carbon source and had 10 days detention. Generally, an increase in detention time resulted in an increase in NH₃ and PO₄^{3−} removal with overall mean values of 86% and 30%, respectively, achieved with 10-day detention periods.

INTRODUCTION

The United Nations (2018) has estimated that the global population living in urban areas will have increased by 4.7% from the 2018 figure by the year 2030 – an additional one billion people. This will lead to increasing demands for infrastructure and safe water sources in many urban areas, particularly in Asia and Africa. However, the provision of these are unsustainable with current development models. The 2030 Sustainable Development Goals (particularly 'Water and Sanitation Goal 6' and 'Sustainable Consumption and Production Goal 12') were thus developed to promote solutions. Authorities and regulators are now being pressed to protect existing water sources within cities and promote water reuse. There is a greater focus on minimising the negative effects associated with urbanisation.

Urbanisation has led to an increase in the number of impermeable surfaces, such as roadways, which act as large waterways during a rainfall event. Fertilizers, animal excrement, atmospheric deposition, sewage, motor-oil, and vehicle emissions, inter alia, settle and accumulate on these surfaces (Collins et al., 2010a). They contaminate stormwater with nitrogen compounds, sediments, heavy metals, pathogens, hydrocarbons, and organics, which are then washed into nearby rivers, streams and dams or infiltrate into groundwater (Bean et al., 2007). Elevated concentrations of nitrate ${\rm (NO_3^-)}$ can result in eutrophication and toxic algal growth (Kim et al., 2003; Kuruppu et al., 2019). Moreover, elevated $NO₃⁻ concentrations (above regulation limit) in drinking water are a concern to many authorities as$ regularly ingesting nitrates can increase one's risk of developing cancer and cause methemoglobinemia in infants (Ward et al., 2018).

Historically, stormwater control measures (SCMs) have been primarily implemented as hydrological management tools used to optimise the removal of runoff and reduce flooding (Collins et al., 2010a, b). Recent research on SCMs has focused on their pollutant removal capabilities, but SCMs have not always been effective at removing nitrogen compounds (e.g., NO_3^- and NH_3) present within stormwater. This has led many researchers to investigate the treatment efficacies of permeable pavements (PPs), which have been successful at removing NH₃, total suspended solids, hydrocarbons, ammonium, total Kjeldahl nitrogen, total phosphorus, biochemical oxygen demand, chemical oxygen demand (COD), heavy metals and pathogenic bacteria (Collins et al., 2010a, b; Tota-Maharaj and Scholz, 2010; Kuruppu et al., 2019). However, most researchers consider PPs to be ineffective at reducing NO₃⁻ concentrations – which were often found to be elevated in PP effluent (Collins et al., 2010a; Kuruppu et al., 2019).

Few studies have investigated the potential for removing $\mathrm{NO_3}^-$ with the inclusion of a submerged zone within a PP structure to facilitate denitrification. Within this zone, denitrifying bacteria can convert NO_3^- to nitric oxide (NO), nitrous oxide (N₂O) and nitrogen gas (N₂), which escape to the surrounding atmosphere. The efficiency of denitrification is dependent on numerous factors including: (i) detention time, (ii) inclusion of a carbon source and (iii) submerged (anoxic) depth (Kim et al., 2003). This study aimed to provide insight into how detention time, the inclusion of a carbon source (newspaper in this case), and submerged depth impact $\rm NO_3^-$, $\rm NH_3$ and $\rm PO_4^{3-}$ removal with a view to improving PICP design (the most widely used PP structure) and thus improving the quality of stormwater entering water sources (e.g., dams, rivers, etc.).

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METHODS

Ten column reactors were filled with 50–63 mm washed aggregate (Fig. 1) and loaded with synthetic stormwater to a depth of 1.5 m over 4 months in a laboratory column study. Columns 1P, 2P, 5P, 10P and 10MP were provided with newspaper as a carbon source to support denitrification, while Columns 1N, 2N, 5N, 10N and 10MN were not. Five different detention times were evaluated (1, 2, 5, 10 and 'varied' days) in pairs of columns with one column with newspaper and the other not. The 'varied' days columns (Columns 10MP and 10MN), with an overall mean detention time of 10 days, were subjected to drying and wetting cycles that mimicked Cape Town's rainfall patterns.

The pollutant concentrations (NH_3 , NO_3^- and PO_4^{3-} – the latter added as an additional nutrient of concern) were measured at different submerged depths (300 mm depth intervals) of each column every 10 days. The pH, temperature, dissolved oxygen, and soluble COD concentrations present in each column were also measured to monitor submerged zone conditions.

Column reactor design and experimental setup

Ten columns were designed and constructed specifically for this study (Fig. 1). Each column weighed an estimated 250 kg with aggregate and synthetic stormwater included. A steel frame was thus constructed to support all 10 columns with removable sections that allowed columns to be safely placed or removed.

A notional 50−63 mm aggregate was chosen as this is what is typically used in the sub-base layer of PICP to provide the reservoir volume as well as support against vehicle loads (Biggs, 2016). The aggregate also provides the surface area for biofilm formation.

Apart from a 100 mm depth that was kept free of aggregate to reduce the risk of overflow, a 400 mm deep unsubmerged zone was left at the top of each column to mimic the upper PICP layers that are generally never submerged and thus provide aerobic conditions for nitrification. The total submerged depth of 1 500 mm was maintained using a raised underdrain (Fig. 1b).

Compared with previous studies (Kim et al., 2003; Lynn et al., 2016; Kuruppu et al., 2019), this study incorporated a greater submerged depth of 1 500 mm with sample ports located at depths of 0, 300, 600, 900, 1 200 and 1 500 mm (Fig. 1b). 1 500 mm was chosen as the maximum submerged depth likely to be implementable in the field. The sampling ports were made by threading taps into the wall of the 315 mm diameter uPVC column and sealing them with marine glue to prevent leaks and air from entering the submerged zone.

Newspaper was the chosen carbon source in this experiment as it was concluded in a study by Kim et al. (2003) to be the best overall electron-donor substrate for providing efficient $\mathrm{NO_3^-}$ removal. The newspaper was first torn into strips before approximately 950 g of it was packed into the 75 mm diameter uPVC pipes located in the centre of each of Columns 1P, 2P, 5P, 10P and 10MP so that the entire 1 500 mm submerged depth of each column was filled with newspaper. Four holes of 8 mm diameter were drilled around the central pipe at 100 mm intervals along its length to allow the synthetic stormwater and the relevant organisms to encounter the newspaper in the inner pipe. Since newspaper remained at the end of the experiment and the soluble COD concentrations remained reasonably consistent through the study, it was assumed that there was sufficient newspaper. No microbiological analysis was undertaken.

Figure 1. (a) Sectional view through a column; (b) photo of a column supported by the steel frame

Synthetic stormwater preparation and loading

Table 1 presents the concentrations of the nutrients used in the synthetic stormwater feed for this study. These were determined by Liu (2020) who considered them 'worst-case' concentrations of pollutants in stormwater according to literature. The synthetic stormwater feed was made in 500 L batches in a 500 L low-density polyethylene tank covered in black plastic to reduce algal growth (Fig. 2a). The procedure for the preparation of the synthetic stormwater is provided in Appendix A.

A new batch of synthetic stormwater was required every 2–3 days. The first 5 batches of synthetic stormwater were tested for consistency and once it had been established that the nutrient concentrations were as per Table 1, no further testing was carried out on subsequent batches. The synthetic stormwater in the 500 L tank was stirred directly before loading the columns to ensure homogeneity of the solution.

The full submerged volume in Columns 1P, 2P, 5P, 10P, 1N, 2N, 5N and 10N was replaced with fresh synthetic stormwater using a watering can to mimic rainfall (Fig. 2b), in accordance with the loading schedule presented in Table 2. While most of the detention times were kept constant throughout the experiment, Columns 10MP and 10MN were subjected to an irregular loading designed to represent the variable detention times in the field more realistically.

Table 1. Synthetic stormwater nutrient concentrations

	Stormwater constituent Targeted influent concentration (mg/L)
NH ₃	2.00
NO ₃	0.60
$POa3-$	0.80

Table 2. Detention time and carbon source for each column

The determination of the loading pattern for Columns 10MP and 10MN was informed by Cape Town's rainfall patterns (Table B1, Appendix B) using daily rainfall data for the months May to August (124 days) – the rainy season – from Cape Town International Airport (CTIA) for 2002, a year in which the total rainfall depth closely matched the long-term mean for this station. The rainfall depths were then increased by a factor of 5 thereby mimicking a run-on factor (ratio of the area of the catchment contributing to the flow onto the permeable pavement to the area of the permeable pavement) of 4 – typical of many PICP designs (Winston et al., 2016). Daily rainfalls of 5 mm or less were ignored as negligible discharge from PICPs has been recorded for small rainfalls such as these (Pratt et al., 1989; Drake and Bradford, 2013). The daily rainfall data in millimetres was then multiplied by a factor of 0.45 L/mm to generate a loading schedule in litres for both columns that incidentally had a mean 10-day detention time over the 124-day experimental period (Table B2, Appendix B).

Chemical analysis

Samples of the stormwater in the 10 columns were analysed every 10 days over 4 months. Samples from the bottom of each column were tested for soluble COD concentrations. A sample was also taken from each of 6 depths (0, 300, 600, 900, 1 200 and 1 500 mm) in the submerged zone of all 10 columns – a grand total of 60 samples every 10th day. These were then tested at the University of Cape Town's Water Quality Laboratory for NH₃, NO₃⁻ and PO₄^{3−} concentrations using a Thermo Scientific Gallery Discrete Analyser. The DO, pH and temperature readings were taken for each sample using hand-held OHAUS probes.

A total of 13 batches of samples were analysed over the 4-month period, which came to a total of 780 samples tested – excluding soluble COD. A total of 130 samples of soluble COD were tested over this same period.

RESULTS AND DISCUSSION

NH₃

Aerobic conditions were created as oxygen $(O₂)$ was introduced into the column's submerged zone as it was loaded with synthetic stormwater, allowing nitrification to take place. During nitrification, NH₃ is reduced to NO_2^- and NO_3^- in a 2-step biological process. The first step (Eq. 1) involves the oxidation of $NH₃$ to form $NO₂^-$, followed by Step 2 (Eq. 2) which is the conversion of NO_2^- to NO_3^- . NO_3^- is a particularly mobile anion,

Figure 2. (a) creating the synthetic stormwater solution; (b) columns being loaded using a watering can

which easily percolates through filter media and soil (Collins et al., 2010a; Kuruppu et al., 2019).

$$
NH_3 + O_2 \rightarrow NO_2^- + 3H^+ + 2e^-
$$
 (1)

$$
NO_2^- + H_2O \rightarrow NO_3^- + 2H^+ + 2e^-
$$
 (2)

Figure 3a presents the overall mean NH₂ concentration (mean across submerged depth and time) for each column over the duration of the study. All columns reduced $NH₃$ by a mean concentration of at least 1.210 mg/L (60% NH₃ removal) over the duration of the study. Moreover, columns with longer detention times displayed greater reduction in $NH₃$ concentration than those with shorter detention times, except for Columns 10P and 10MP. This is displayed in Fig. 3a where mean $NH₃$ concentration decreased from Columns 1P to 5P (1, 2- and 5-days detention) and Columns 1N to 10N (1-, 2-, 5- and 10-days detention) by 0.220 mg/L and 0.350 mg/L, respectively. Out of these columns, Column 1P with the shortest detention time of 1 day provided the lowest $NH₃$ reduction of 1.374 mg/L (69% $NH₃$ removal), and Column 10N with the longest detention time of 10 days provided the highest NH_3 reduction of 1.725 mg/L (86% NH_3 removal). The longer the detention time of a column, the more time the nitrifying microorganisms had to reduce NH₃. In addition, columns both with and without carbon performed similarly in terms of their ability to reduce NH₃. This is supported by having conducted a one-way analysis of variance (ANOVA) test which indicated that the inclusion of a carbon source had no significant effect on $NH₃$ concentration at the 5% significance level ($p = 0.13$).

In general, Fig. 3b shows that the $NH₃$ concentration increased with an increase in depth from 600 to 1 500 mm and most columns displayed their greatest increase in NH₂ concentration between 1 200 and 1 500 mm. This could possibly be because an increase in depth resulted in a decrease in DO concentration, which inhibited the nitrifying microorganism's ability to reduce NH₃ due to limited O₂ availability (Collins et al., 2010a; Kuruppu et al., 2019).

NO3 −

After nitrification had occurred in the columns, $\rm NO_3^-$ was further reduced to nitrogen oxides in a process called denitrification. Biological denitrification occurs in the anoxic zone through 4 main pathways: the reduction of $NO₃⁻$ to $NO₂⁻$ (Eq. 3), the

conversion of NO_2^- to NO (Eq. 4), the production of N_2O from NO reduction (Eq. 5), and lastly the formation of $N₂$ via further reduction of N_2O (Eq. 6) (Kuruppu et al., 2019).

 $NO₃⁻ + 2H⁺ + 2e⁻ \rightarrow NO₂⁻ + H₂O (NO₃⁻ reductase)$ (3)

$$
NO2- + 2H+ + e- \rightarrow NO + H2O (NO2- reductase)
$$
 (4)

 $2NO + 2H^+ + 2e^- \rightarrow N_2O + H_2O$ (nitric oxide reductase) (5)

 $N_2O + 2H^+ + 2e^- \rightarrow N_2 + H_2O$ (nitrous oxide reductase) (6)

Figure 4a shows that an increase in detention time – for columns which included newspaper – allowed for a decrease in $NO₃^$ concentration as denitrifying microorganisms had more time to reduce $NO₃^-$. With reference to columns which included newspaper, the shorter detention times of 1 and 2 days provided an increase in overall mean $NO₃⁻$ concentration by 1.690 and 1.350 mg/L (282 and 225%), respectively. The longest detention time of 10 days (Column 10P) provided the greatest decrease in overall mean NO_3^- concentration, of 0.421 mg/L (70% $NO_3^$ removal). Detention times of 1, 2 and 5 days resulted in an increase in $NO₃⁻$ concentration, possibly due to the denitrifying microorganisms not having enough time to reduce $NO₃⁻$, causing $\rm NO_3^-$ accumulation. Column 10MP suggests that an overall mean $NO₃⁻$ removal of 41% is achievable in the field if PICP includes newspaper in its submerged zone and allows for a mean 10-day detention. The overall mean NO_3^- removal of 70% (Column 10P) and 41% (Column 10MP) in this study was far greater than that reported in other PP studies. Collins et al. (2010a) found that in most cases studies reported negative $NO₃⁻$ removal (production of $NO₃⁻$) in PPs.

The inclusion of newspaper had a significant effect on $NO₃^$ concentration. Columns 1N to 10MN shows that the absence of newspaper resulted in an increase in overall mean $NO₃$ ⁻ concentration by over 200%. This was possibly due to denitrifying microorganisms not having the carbon source (electron donor) they needed to reduce $NO₃⁻$ to $N₂$ (Kuruppu et al., 2019). Instead, nitrifying microorganisms continued to convert $NH₃$ to $NO₃$ without the ability for denitrifying microorganisms to reduce $NO₃⁻$, thus resulting in NO₃⁻ accumulating in these columns. This indicates that short detention time (1 and 2 days) and exclusion of a carbon source both hindered the ability for denitrifying microorganisms to reduce $NO₃⁻$ to a similar extent.

Figure 3. (a) Overall mean (± standard deviation) NH₃ concentration for each column; (b) mean NH₃ concentration as a function of submerged depth

Figure 4. (a) Overall mean (± standard deviation) NO₃-concentration for each column; (b) mean NO₃-concentration as a function of submerged depth

Figure 5. (a) Overall mean (± standard deviation) PO $_4$ ^{3–} concentration for each column; (b) mean PO $_4$ ^{3–} concentration as a function of submerged depth

Figure 4b shows that, generally, $NO₃⁻$ concentration decreased with an increase in depth from 0 to 1 500 mm and most columns displayed their greatest decrease in $NO₃⁻$ concentration at depths between 1 200 and 1 500 mm. This could possibly be because an increase in depth resulted in a decrease in DO concentration which improved anoxic conditions and allowed denitrifying microorganisms to reduce $NO₃⁻$ at a higher rate (Knowles, 1982; Sperling, 2007; Kuruppu et al., 2019). Columns 10P and 10MP, with the highest overall mean $NO₃⁻$ removal of 70% and 41%, respectively, remained relatively unaffected by an increase in depth greater than 300 mm. This was possibly due to there being limited availability of ${\rm NO_3^-}$ remaining for denitrifying microorganisms to further reduce.

The mean DO, pH and temperature readings for all columns ranged from 1.5 to 2.0 mg/L, 6.29 to 6.83, and 23.2 to 23.3°C, respectively. These values are within the necessary range for denitrification (Knowles, 1982; Volokita et al., 1996; Xu et al., 2009). Moreover, mean soluble COD concentrations ranged from 2 to 100 mg/L for all columns over the duration of the

study – indicating the availability of a soluble carbon substrate for denitrification.

PO4 3−

Figure 5a shows that Columns 1P, 2P, 1N and 2N, with detention times of 1 and 2 days, provided the highest overall mean PO_4^{3-} concentration at a range of 0.696 to 0.671 mg/L (13% to 16% PO_4^{3-} removal). Columns 5P, 10P, 10MP, 5N, 10N and 10MN, with longer detention times of 5, 10 and 'varied' days, provided the lowest mean PO_4^{3-} concentrations with a range of 0.626 to 0.535 mg/L (22% to 33% PO_4^{3-} removal). This indicates that an increase in detention time allowed for a decrease in PO_4^{3-} concentration as the microorganisms had more time to reduce PO_4^3 ⁻. A one-way ANOVA test confirmed that the inclusion of a carbon source had no significant effect on PO_4^{3-} concentration at the 5% significance level ($p = 0.45$). Columns both with (Columns 1P, 2P, 5P, 10P) and 10MP) and without (Columns 1N, 2N, 5N, 10N and 10MN) newspaper were able to reduce overall mean PO_4^{3-} concentrations by 0.104 to 0.265 mg/L (13% to 33% PO_4^{3-} removal).

Figure 6. Percentage mass of nitrogen in each column

Figure 5b shows that, generally, PO_4^{3-} concentration decreased with an increase in depth from 0 to 900 mm. Thereafter, an increase in PO_4^{3-} concentration occurred in most columns when the depth increased from 900 to 1 500 mm. It is not known why PO₄^{3–} concentration increased in columns between depths of 900 and 1 500 mm. There is very little published research on PO43− concentrations at such depths in PICP.

Nitrogen mass balance

Figure 6 presents the distribution of the overall mean mass of nitrogen (%) in each column. Nitrogen compound 'X' represents nitrogen oxides such as NO, N₂O and N₂ which can escape to the surrounding atmosphere and are therefore assumed to be removed from the system. No assessment could be made of nitrogen incorporated within the biomass of organisms growing within the columns, which was assumed to be negligible – but may not be. With these assumptions, all columns appeared to remove between 41% and 72% of nitrogen from the influent synthetic stormwater. Although Column 10P with a 10-day detention provided the second highest overall mean nitrogen removal of 69%, it performed the best in terms of $NO₃⁻$ removal. Column 10MP indicates that an overall mean nitrogen removal of 59% is achievable in the field if PICP includes newspaper and has a mean 10-day detention.

CONCLUSIONS

This study concludes that the inclusion of a submerged zone in PICP in the field has the potential to promote denitrification. A submerged depth of 300 mm was sufficient to achieve a minimum $\rm NO_3^-$ removal of 41% in columns which included a carbon source (newspaper in this instance) and had a 10-day detention time. 59% nitrogen removal is possible. An increase in detention time is associated with a decrease in both NH_3 and $\mathrm{PO_4^{3-}}$ concentrations, with the longest detention time in the laboratory of 10 days resulting in an overall mean removal of 86% and 30%, respectively. However, the inclusion of a carbon source had no significant impact on NH_3 and PO_4^{3-} removal. In most cases an increase in submerged depth resulted in an increase in NH₃ concentration from 600 to 1 500 mm, and a decrease in PO_4^{3-} concentration from 0 to 900 mm. PICP thus has the potential to significantly reduce $NH₃$, NO₃⁻ and PO₄³⁻ compounds present in stormwater through the incorporation of a submerged zone, ultimately improving the quality of runoff entering the natural environment.

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AUTHOR CONTRIBUTIONS

Luke Brown was responsible for the conceptualisation and methodology of the study; the setting up and running of the laboratory experiment; data collection, analysis, and interpretation; and writing the first draft of the paper. Neil Armitage supervised the study and edited the paper with the assistance of Luke.

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APPENDIX A

Procedure for the preparation of the synthetic stormwater

Stoichiometric principles were used to determine that approximately 7.64 mg/L of ammonium chloride (NH4Cl), 4.33 mg/L of potassium nitrate $(KNO₃)$, and 4.50 mg/L of di-potassium hydrogen phosphate $(K, HPO₄)$ were required to produce nutrient concentrations of 2.00 mg/L NH₃, 0.60 mg/L $NO₃⁻$ and 0.80 mg/L PO₄³⁻. The following steps were followed for the preparation of the synthetic stormwater:

- (i) A 500 L tank was filled with municipal tap water.
- (ii) A 1 L volumetric flask with half-filled with de-ionised (DI) water.
- (iii) 3.819 g NH₄Cl, 2.166 g KNO₃ and 2.249 g K₂HPO₄ were weighed out using an analytical balance and added to the 1 L volumetric flask.
- (iv) The 1 L volumetric flask was then filled with DI water and stirred with a magnetic stirrer until all solids had dissolved.
- (v) The 1 L concentrated solution was added to the 500 L tank and stirred using a paddle for 10 minutes.
- (vi) The tank lid was closed. The synthetic stormwater was now ready for immediate use.

APPENDIX B

Determination of the loading rates for the 'varied' loading regime

