

Treatment efficiency of a wet detention pond combined with filters of crushed concrete and sand: a Danish full-scale study of stormwater

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Abstract Traditional wet detention ponds and sand filters remove particles efficiently, whereas only a minor part of the dissolved and bioavailable load is removed. To improve the retention of dissolved substances, we tested crushed concrete as a filter material simultaneously with a traditional sand filter placed after an existing wet pond. The particulate fractions (particles, organic matter, phosphorus, and heavy metals) were removed efficiently in the pond and both filter materials, with the concrete filter often being best seen over a year. Dissolved heavy metals (lead (Pb), nickel (Ni), copper (Cu), chromium (Cr), and cadmium (Cd)) were largely retained, though a washout was observed from the pond (Ni and Cu), concrete filter (Cr), and sand filter (Ni) during the first month. The pond only retained total dissolved phosphorus (TDP) during summer. Crushed concrete and sand had a high (>70 %) retention of TDP within the first months of operation, but the retention dropped in both filters due to a large oil load into the system (4 kg impermeable ha^{-1} in 1 month). The poor retention might to some degree be due to mineralization

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M. J. Sønderup · T. Bochdam Arwos A/S, Forsyningsvejen 2, 6200 Aabenraa, Denmark processes turning particulate phosphorus (PP) into TDP. The massive oil load was retained efficiently (99.3 %) in the pond and both filters, clearly illustrating that both filter materials were able to retain either oil or TDP. An additional pilot study showed that at residence times of 1 h, crushed concrete bound 90 % TDP whereas sand only bound 22 % TDP. Retention of TDP and PP decreased with shorter residence time in both materials, but fastest in sand.

Keywords Urban runoff \cdot Neutralization \cdot Retention \cdot Phosphorus \cdot Wet detention pond

Introduction

Stormwater runoff contains dissolved and particulate organic matter, nutrients, and xenobiotics, where the composition mostly depends on the land use in the catchment (e.g., Birch et al. 2005; Gobel et al. 2007; Egemose et al. 2014; Sønderup et al. 2015). Phosphorus (P) tends to accumulate in receiving water bodies, whereas nitrogen (N) and organic matter (OM) can either be released to the atmosphere as N_2 and CO_2 during mineralization or accumulate in the sediment, depending on the inlet concentration and the mineralization rate (Cooke et al. 2005). As discharged particulate nutrients and OM are easily removed by sedimentation, focus should be on the dissolved fraction as it is difficult to remove and directly bioavailable.

Applying active measures like stormwater ponds support retention of suspended solids (SSs), nutrients, and xenobiotics by buffering the hydraulic load (Hvitved-Jacobsen et al. 2010). Dry ponds are preferred in arid or semiarid climate zones, because the high evaporation rate does not allow permanent water level. Infiltration to the surrounding soil will recharge the groundwater but, at the same time, create a risk of groundwater pollution (Barbosa and Hvitved-Jacobsen 2001). Wet ponds are widely used in moist climate zones like Denmark, and they are often constructed with a clay membrane to avoid infiltration and contamination of the groundwater and to ensure a permanent minimum water level. This pond type mainly retains large and heavy particles and contaminants sorbed to these. As retention of particulate matter depends on the particle sizes and the residence time (Reinhardt et al. 2005), the size of the wet volume (and hence the residence time) is important. This is illustrated by a comparison of a highloaded (40 m³ red ha⁻¹ (reduced or impermeable hectare)) and low-loaded (240 m^3 red ha^{-1}) wet pond, where the high-loaded pond was consistently observed to have the lowest retentions of, e.g., SS of 42 and 84 %, respectively (Hvitved-Jacobsen et al. 2010).

Wet ponds, on the other hand, do not support sufficient time to remove the lighter particles and the dissolved fraction from the water column (Muthukrishnan and Selvakumar 2006; Egemose and Jensen 2009; Hvitved-Jacobsen et al. 2010). Unfortunately, these fractions are the most nutrient rich (Stone and English 1993) and contain a higher fraction of heavy metals (Sansalone and Buchberger 1997). In addition, already settled matter and retained nutrients might over time be released from the pond, due to wind/wave-induced resuspension activities, mineralization of organic particles, and/or reduced conditions.

Combining a traditional wet or dry pond with filter media (often sand) improves the retention of the particulate fraction but also the dissolved fraction. This is due to filtration, adsorption, and precipitation processes (Arias et al. 2001). The particulate loading will gradually clog the porous structure of every filter (Siriwardene et al. 2007; Knowles et al. 2011), but by ensuring satisfactory residence time in the pond before infiltration, the coarse particulate fraction is removed and the lifetime of the filter is prolonged (Barbosa and Hvitved-Jacobsen 2001; Birch et al. 2005). Eventually, the filter material though has to be replaced to keep a satisfying hydraulic conductivity (Nivala et al. 2012).

Sand filters efficiently remove SS (e.g., Birch et al. 2005; Siriwardene et al. 2007; Hvitved-Jacobsen et al. 2010) and often have a high N removal due to coupled nitrification-denitrification processes (Arias et al. 2001). However, in filters with a permanent water level, the limited oxygen availability inhibit nitrification (Vohla et al. 2007). Retention of dissolved inorganic phosphate (DIP) depends on the adsorption to iron (Fe) and aluminum (Al) and precipitation with calcium (Ca) (Arias et al. 2001). In a study of 13 Danish sand types, of which some were impacted by the Weichsel glaciation (clayey and calcareous soils) and some were not (sandy and leached out soils), Brix et al. (2001) found that some sand types would use up the P-binding capacity within a few months while, e.g., the Ca-rich types would last for years. Coating the materials with P removal agents like Ca, Fe, and/or Al improves the efficiency (Hvitved-Jacobsen et al. 2010). Ayoub et al. (2001) found that coating of silica sand and olivine with a combination of Fe and Al hydroxyl coating increased the DIP retention dramatically from a limited DIP capacity to >90 % DIP removal.

Alternative filter materials with good DIP removal capacities have been tested in several studies (e.g., Ballantine and Tanner 2010; Vohla et al. 2011), including natural, industrial, and man-made materials. Beside their DIP removal capacity, the materials were evaluated due to economy, local availability, non-toxicity, reusability, and safe disposal (Ballantine and Tanner 2010). Berg et al. (2005) and Oguz et al. (2003) showed that crushed gas concrete removes DIP efficiently (85-99 %), and studies of Molle et al. (2003) and Egemose et al. (2012) showed the same for crushed concrete, which is a cheap $(10 \in t^{-1})$ residue from demolition of buildings. The high DIP-binding capacity is due to the cement content of Ca oxides (≈ 63 %), Al oxides (≈ 5 %), and Fe oxides (≈ 3 %) (Molle et al. 2003; Mechling et al. 2009), inducing adsorption to Fe and Al and precipitation with Ca as calcium phosphate on the concrete surface (Oguz et al. 2003). The natural release of calcium hydroxide from concrete induces a pH increase in the water, often to pH about 10 (Molle et al. 2003; Egemose et al. 2012). As calcium phosphate gradually covers the concrete surface, the Ca release is reduced and, at a certain limit, the DIP-binding capacity is reached (Berg et al. 2005). This process also leads to a gradual decline in pH towards neutral conditions (Berg et al. 2005). If crushed concrete is used as a filter material, the treated stormwater needs to be diluted or neutralized before being discharged, as the high pH can be harmful to living organisms.

The aim of this study was to investigate the treatment efficiency of a demonstration facility comprising an existing wet stormwater pond followed in parallel by a traditional sand filter and a filter of crushed concrete, allowing comparisons of simultaneous measurements during the same conditions. Crushed concrete was chosen, as it showed a great potential for P removal in several short-term laboratory studies. Consequently, we tested the material in a pilot study (≈ 1 month), before making the full-scale experiment at natural precipitation conditions with varying intensity, duration, and inlet concentrations and with natural pH, oxygen, and temperature. The full-scale experiment lasted 1 year. Here, we tested the retention of P, N, SS, OM, Ca, Fe, Al, heavy metals, and hydrocarbons in runoff from an industrial catchment. We followed pH not only in the concrete filter to test the effect on P removal over time but also in the effluent after neutralization to ensure good conditions downstream.

Methods

Pilot study

The pilot study was conducted to gain experience with flow rates, retention efficiencies, and kinetics supporting the design of the full-scale demonstration facility.

Water from a wet stormwater pond in the city of Aabenraa was used, as the concentrations of hydrocarbons and heavy metals in the runoff were low while the concentration of total dissolved phosphorus (TDP) was high at about 200 μ g P L⁻¹. Water was pumped into two intermediate bulk containers (1000 L each), filled with 0.2 m^3 stones (8–16 mm) in the bottom and 0.5 m^3 of either crushed concrete (0-40 mm) or sand (0-8 mm) on top. Water was percolated vertically through the filter materials and ran out from a tap placed in the bottom of each container. Inflow and outflow were adjusted to 2.4, 4.8, and 9.5 mL min⁻¹, where each flow scenario lasted 3 days and was followed by a still water period of 3 days. The filters were kept saturated during the entire period in order to calculate residence times. Prior to the experiment, water was pumped through the filter materials (4 days at 2.4 mL min⁻¹) to wash out loosely bound particles and thereby avoid interference during the experiment. During each 3-day flow campaign, water samples for TDP and particulate phosphorus (PP) were taken every third hour the first day and then with 6 and 12 h of temporal resolution the second and third days, respectively.

Demonstration facility

An existing wet stormwater pond (man-made for stormwater treatment) in the city of Padborg, Denmark, was in winter 2012/2013 converted into a wet pond followed by a traditional sand filter and a filter of crushed concrete to allow comparison (Fig. 1). The facility receives runoff from an industrial catchment of 18.2 ha (impermeable area of 11.8 ha). Point measurement of P conducted in January 2012 (before renovation) showed that approx. 450 µg total phosphorus (TP) L^{-1} entered the pond while approx. 150 µg TP L^{-1} was discharged and that P was equally distributed between dissolved and particulate P. The relative high P concentration in the outlet made the pond well suited as a test facility. The catchment mainly consists of huge parking lots for trucks, and the runoff is therefore very oil containing. This is why the existing system had an oil separator attached, which is still in use. We also discovered that the parking area was short of toilets for the many truck drivers (<1000 at a time) and that they therefore urinated along the fence during summer and in portable toilet facilities emptied in the rainwater grids during winter. This resulted in high concentrations of TDP (primarily organic) and ammonium (NH_4^+) in the runoff, especially during winter.

Due to the truck driver's regular (but illegal) oil spill into the sewer system during 17 years, the existing pond was more or less full of oil sludge that had to be removed. During this process, the facility was enlarged to the current pond (0.8 m depth, 450 m³ wet volume) and storage capacity (1.8 m depth, 1850 m³ max volume). Existing pipes and wires in the ground restricted the size of the enlargement, and the pond therefore ended up being undersized (38 m³ red ha⁻¹), compared to optimal conditions between impermeable area and pond volume (>250 m³ red ha⁻¹ (Hvitved-Jacobsen et al. 2010)).

A joint water brake (maximum capacity of 18.2 L s^{-1}) placed in the pond outlet controls the filter inlets and hence the minimum residence time (0.9 h) in each filter. The water is distributed into the two filters through a Y-pipe (Fig. 1). This design made it possible to test the two filter materials simultaneous during the same conditions of rain intensity, pollution load, dry



Fig. 1 Schematic overview of the demonstration facility with indication of the main waterways into and out of the system. From the grit chamber, the water flows to the pond but, when the water level in the pond >1.2 m (i.e., 0.4 m above normal water level), 3 L s⁻¹ is discharged to the oil separator. From the outlet of the

pond, the water is discharged to the two filters and/or accumulates in the pond and, only when the water level is >1.8 m, the overflow become effective. The cross-sectional view shows the normal and maximum water tables

periods, etc. The piping construction ensured that 5 cm of water always would cover the filter surface (cross-sectional view in Fig. 1) to ensure an equal water distribution on the surface and thereby prevent formation of channels. The water percolates vertically through the filters (1 m depth, 93 m³ filter material) into a drainage system below the filter. Here, the water is collected and discharged to a nearby stream. Vertical filtration was chosen rather than horizontal filtration, as the first has a better distribution of water and load and hence a lower risk of clogging (Hvitved-Jacobsen et al. 2010).

Grain sizes for the concrete filter and sand filter were 3–35 and 0–4 mm, respectively, providing pore volumes of 30 m³ per filter. Unfortunately, the sand filter turned out to behave like quicksand and, because of safety issues, it was covered by a crop protection fleece and 10 cm of stones (16–32 mm) at the end of March 2013. This changed the infiltration capacity, and the flow through the concrete filter became three times higher than through the sand filter (see the "Results and discussion" section). Capital costs for construction of the concrete filter and sand filter were 23,000 and 18,000 \in , respectively. The higher cost for the concrete filter is due to extra costs for setup of neutralization equipment. Notice that the capital costs for both filters include extra wells and pipelines for sampling and documentation of the study, which a normal stormwater pond would not include. Operational costs are the same for the two filters and are estimated to 2000 \notin year⁻¹. This cost includes replacement of the filter materials after 10 years.

Neutralization well

Due to expected high pH in the discharge from the concrete filter, a pH-monitoring pump was placed in the effluent well (\emptyset 1250 mm) to ensure neutralization by acid dosing. We chose this acid-dosing method (concentrated hydrochloric acid (HCl)) as it is a reliable neutralization method widely used in the wastewater industry.

Laboratory studies showed that pH in the concrete filter would decrease over time. This was also observed in the demonstration facility. After 8 months, pH was <8.5 and neutralization was stopped. A total of approx. 1500 L concentrated HCl was used.

Monitoring

The pond was monitored during 1 year, starting in the middle of March 2013. The monitoring program was designed to study the processes in the sedimentation pond and to compare the two filter materials. During the sampling period, precipitation data was retrieved from a local rain gauge situated next to the pond (accuracy 1 %, resolution 0.2 mm tip⁻¹).

Manual flow measurements (triplicates) were conducted on a regular basis in the inlet and outlet pipes with an OTT Kleinflügel Propeller instrument, converting rotations into velocity and then into flow using the wetted cross-sectional area of the pipe. Water level loggers (accuracy 0.3 %, resolution <0.02 kPa) were installed in the inlet and outlet from the pond, in the inlet to the filters, and in each of the two filters (Fig. 1). The sampling frequency of water levels was every 5 min. Pressure was converted into depth using a reference logger and manual water level measurements for calibration. These depths were then converted into flow using the manual flow measurements (supplementary information). As the loggers did not measure flow directly, an effort was put into the process of converting and calibrating water levels into flows, conducted by frequent flow measurements throughout the year. This ensured good relations between manual flow measurements and logged water levels, and it prevented loggers from drifting.

Oxygen (accuracy 0.5 mg L^{-1} , resolution 0.02 mg L^{-1}), temperature (accuracy 0.2 °C, resolution 0.02 °C), and pH (accuracy 0.1, resolution 0.1) were monitored with a temporal resolution of 5 min by loggers installed in the inlet and outlet from the pond and in the nearby stream (receiving water). In addition, pH and water level were monitored continuously in the neutralization well to control the acid dosing and thereby pH in the effluent and data was logged every 15th min (Fig. 1). For more information about the loggers and Kleinflügel, e.g., calibration, operation, and control, see the product sheets from Onset[®] HOBO[®] Data loggers (water level, oxygen, temperature, and rain gauge), TruTrack Data Logger (pH; Intech Instruments Ltd), and OTT Hydromet (flow).

Water samples and analyses

Water samples were collected at the inlet and outlet wells from the pond and in the outlet wells from the concrete and sand filters (Fig. 1) with automatic samplers (Teledyne ISCO sampler with GSM modules). This sampling equipment made it possible to take water samples with varying temporal resolution, depending on the precipitation intensity and history. Sampling was conducted regularly throughout the year, with 1 sample per day during the first 2 months and 1 per week afterwards. In addition, ten campaigns were conducted during different runoff scenarios with varying precipitation intensity and duration, antecedent dry periods, and season variations. Samples were taken before rain, hourly during rainfall and each 3–6 h until 2 days after the rain event.

Known volumes of each water sample were filtered in replicates on pre-weighted Whatman GF/C filters (pore size 1.2 µm) for SS, OM, PP, particulate iron (PFe), particulate calcium (PCa), and particulate aluminum (PAl). SS was measured in replicates using both filters (105 °C, 24 h), whereas OM was measured using one of the filters (520 °C, 2 h). PP and PFe were measured by boiling the combusted filter in 1 M HCl (1 h) followed by determination of DIP and dissolved iron (DFe) in the extract (Andersen 1976). DIP was measured using the molybdenum blue method (Koroleff 1983). DFe was measured using the ferrozine method (Gibbs 1979). The remaining filter was solubilized by digestion in 6 mL of 65 % HNO₃ using a microwave digester for 30 min at 1600 W by heating the solution to 180 °C. After dilution, the PCa and PA1 concentrations were measured in the liquid phase by inductively coupled plasma with optical emission spectroscopy (ICP-OES; Optima 2100 DV, PerkinElmer).

The filtrated water was analyzed for nitrate (NO₃⁻), NH₄⁺, TDP, DFe, dissolved calcium (DCa), and dissolved aluminum (DAl). NO₃⁻ and NH₄⁺ were analyzed spectrophotometrically on a flow injection system (QuikChem 8500 Series, Lachat Instruments, QuikChem method 10-107-041-C). TDP, DCa, and DAl were measured by ICP-OES.

Total nitrogen (TN) was measured as NO_3^- on an unfiltered sample after digestion with potassium peroxodisulfate and sodium hydroxide (QuikChem method 31-107-04-3-B).

Dissolved and particulate copper (Cu), chromium (Cr), cadmium (Cd), lead (Pb), and nickel (Ni) were

measured on selected samples by ICP-OES (similar to measurements of Al and Ca). Hydrocarbons were measured on a few unfiltered samples by extraction with pentane (ISO 9377-2 mod. GC/FID) as different fractions (C6–C10, C10–C25, C25–C35). The short chains (C6–C10) are mainly gasoline, the longer chains (C10–C25) are mainly diesel and fuel oil, and the longest chains (C25–C35) are mainly petroleum products. Water samples were collected for heavy metals and hydrocarbons for the entire year but were not analyzed yet due to high analysis costs.

Small lab study

The DIP adsorption capacity of concrete with and without oil was tested in triplicates. Two grams of crushed concrete (grain size <18 mm) was either mixed with 7.5 mL of waste oil and shaken for 48 h or left unaffected. The concrete samples were then added to 50 mL of 2 mM NaHCO₃ (to mimic natural alkalinities) with 500 μ g DIP L⁻¹ and shaken for 24 h, and after which, they were analyzed for DIP.

Calculations

The flow into the grit chamber, oil separator, and pond and out from the pond either by outlet or overflow and out from the two filters was calculated every 15 min from their respective water level changes (converted into flow) and the manual flow measurements (supplementary data). This 15-min interval was chosen to limit the amount of data points while keeping a high resolution. The flow into the grit chamber, i.e., runoff, consisted of precipitation data and additional sources, e.g., groundwater, infiltration, other catchment activities, and evaporation (Eq. 1, calculation of the total runoff entering the grit chamber). Additionally, changes in pond and filter volumes were calculated from the water level changes. Based on these calculations, the water balance was made for periods of 3 months.

$$Q_{\text{runoff}} = \text{Rain} \cdot A_{\text{catchment}} + Q_{\text{groundwater}} + Q_{\text{infiltration}}$$
(1)
+ Q_{other} -Evaporation $\cdot A_{\text{catchment}}$

All substances, except heavy metals and hydrocarbons, were sampled daily, weekly or intensively (1035 samples in total) during the studied year. The measured concentrations were interpolated between sampling times, with one sample covering the period (15-min intervals) until the next sample. Masses were then calculated for the 15-min intervals, from concentrations and flow into and out from the pond and out from both filters, and summarized in mass balances for periods of 3 months. As the experiment lasted from the middle of March 2013 to the middle of March 2014, the last quarter of the experiment seems to cover 4 months in Table 3, though it is actually only 3 months.

The same principle was applied for heavy metals and hydrocarbons, based on daily and weekly samples, respectively, though only for the first month. Retentions were calculated for the pond and for each of the two filters, as differences in mass import and export.

Results and discussion

Pilot study

The pilot study was made to test the P adsorption at different infiltration rates. The average inlet concentrations of TDP and PP were 218 ± 137 and $37\pm23 \ \mu g \ L^{-1}$, respectively. The retention of TDP and PP in the filter materials was decreasing with shorter residence times for both filters (Table 1). The concrete filter showed a high retention of TDP (90 %) at residence times of 1 h. However, at residence times <16 min, the contact time became too small for adsorption and the flow became high enough for washout of newly adsorbed P. The sand filter was only able to bind TDP (23 %) at residence times of 1 h. The significant difference between TDP removals in the concrete and sand filter is ascribed to the much higher content of Ca, but also Fe and Al in concrete, contributing to adsorption and/or precipitation

Table 1 Pilot study of TDP and PP retention in a 0.5-m³ concrete filter and a 0.5-m³ sand filter at different water residence times

Residence time (min)		Concrete filter		Sand filter	
		TDP	PP	TDP	РР
63	%	90	69	23	82
	$\mu g \ L^{-1}$	23	18	183	10
31	%	65	53	-4	39
	$\mu g \ L^{-1}$	58	15	168	19
16	%	-10	52	-7	11
	$\mu g \ L^{-1}$	94	9	92	17

The retention is given in percent along with the mean outlet concentrations in micrograms per liter

of TDP. Regarding PP, the sand filter had the highest retention at residence times of 1 h (82 %) but it decreased rapidly with decreasing residence time (39 % at 31 min and 11 % at 16 min), while the concrete filter had a more constant retention of PP independent on residence time (52–69 %). This difference between sand and concrete was probably related to the different grain size distributions of the filters.

Although laboratory studies showed that crushed gas concrete and crushed concrete can adsorb >90 % DIP within 15 min (Oguz et al. 2003; Sønderup, unpublished data), it was not the case in the pilot study. This was probably because of the much higher grain size (0– 40 mm) in the pilot study compared to the laboratory studies (<2 mm). Egemose et al. (2012) showed that smaller grain sizes are more efficient than larger ones regarding DIP retention, as the higher surface-tovolume ratio increases the adsorption and precipitation. Grain sizes up to 30–50 mm are common standards in demolition companies and are therefore cheap and easy to obtain and use in future full-scale concrete filters. The minimum residence time should therefore at least be 30 min, but preferably 1 h.

Demonstration facility

Flow and water balance

Precipitation during the investigated year (867 mm) was close to an average year (822 mm) and well within the normal variations (639-990 mm) in this region (data from a national rain gauge situated in the city of Aabenraa). This means that it was an ordinary year regarding precipitation and that the results therefore are comparable with similar studies. Apart from stormwater runoff, additional contributions came from groundwater and infiltration (visible seepage into the pipelines), plus activities in the catchment, like car wash and emptying of portable toilets. These contributions corresponded to 52 % of the total runoff and occurred even during dry weather periods. High groundwater seepage into the sewer system is not unusual in Denmark, as the large concrete pipes for transporting stormwater do not seal tightly, and as big aquifers lay just below the ground level. This condition is anticipated to be similar in other sites out of Denmark, though it is often not recognized.

Most of the yearly runoff from the catchment (72 %) occurred during autumn and winter (September-

March), due to a higher precipitation and groundwater flow in this period. This was reflected in the entire pond system (Table 2). A small part of the runoff was discharged directly through the oil separator (13 %), while the rest entered the pond. From here, most of the water ran to the filters while 1 % evaporated and 24 % was discharged during overflows, primarily in autumn and winter. The average residence time in the pond was 20 ± 2 h. Some water (<0.1 %) evaporated from the filter surfaces, but most percolated through the filters before discharge.

The inflow to the two filters should ideally be equal, but because the sand filter was covered by a crop protection fleece and because of different grain sizes, the infiltration rate also differed, resulting in an average flow of 3.3 ± 2.8 L s⁻¹ (maximum 8.9 L s⁻¹) from the concrete filter and 1.3 ± 0.6 L s⁻¹ (maximum 2.2 L s⁻¹) from the sand filter. This affected the residence times, which in average were 5.9 ± 5.6 h in the concrete filter and 8.8 ± 5.5 h in the sand filter. The unequal distribution of water caused different loadings on the filters, and the concrete filter experienced shorter time for percolation and also for adsorption and biological retention processes compared to the sand filter.

A large rain event with respect to millimeters may result in a lower retention in the pond (Sharma et al. 2011), due to shorter residence time. For SS and PP (Table S1), there were significant negative correlations between the measured precipitation and retention in the pond during rain events (p=0.036 for both parameters). The duration of each event may also impact the residence time and thereby the retention in the pond. Significant negative correlations were found between the duration of an event and the retention of SS and PP in the pond (p<0.001 and p=0.005, respectively). TDP showed similar decreasing trends for precipitation and duration, without being significant (Table S1).

Temperature, oxygen, and pH

Continuous logger measurements showed that the largest variations in temperature occurred during summer, whereas it was more stable during winter. Mineralization and retention are expected to be higher during summer (20-25 °C) compared to winter (<10 °C), as mineralization rate increases with increasing temperature (Candfield et al. 2005). Independent of season, the temperature decreased in the system during rain events, when the stagnant water masses, heated by

Water balance		March-May	June-August	September-November	December-March	Total
Runoff						
Precipitation	mm	106	174	327	260	867
From catchment	m ³	13,211	21,596	40,729	32,319	107,855
Other ^a	m ³	19,352	8129	36,157	51,445	115,081
Grit chamber						
In	m ³	32,564	29,725	76,886	83,764	222,938
Out oil separator	m ³	794	1045	7394	19,631	28,864
Pond						
In ^b	m ³	31,770	28,680	69,492	64,134	194,075
Overflow	m ³	2134	4829	24,251	15,091	46,305
Out	m ³	28,642	22,843	44,946	49,030	145,462
Difference ^c	m ³	994	1008	294	12	2309
Filters						
In concrete	m ³	19,832	16,766	33,500	35,012	105,110
In sand	m ³	8810	6077	11,446	14,018	40,352
Out concrete	m ³	19,827	16,760	33,463	35,015	105,066
Out sand	m ³	8799	6065	11,407	14,021	40,292
Difference ^d	m ³	16	18	77	-7	104

Table 2 Water balance calculated from the middle of March 2013 to the middle of March 2014 for intervals of 3 months

^a Additional runoff from the catchment, e.g., groundwater, other sources, infiltration, and loss by evaporation

^b Inlet to the pond=inlet to the grit chamber-outlet to the oil separator

^c Retention and/or evaporation

^d Total difference between inlet and outlet from the two filters, which is made up by retention and/or evaporation

the sun, were flushed out by the colder runoff. These variations did not seem to impact the retention during rain events. The temperature tended to be warmest in the filters, due to the shallow water level above the filter surfaces (0.05–0.5 m), and coldest in the receiving stream, due to cooling in the pipeline (179 m) and shading provided by trees and shrubs close to the stream.

The pond was usually high in oxygen during dry summer periods, despite of high oxygen consumption from oil-containing water and high temperatures. Periodically, the pond water became oversaturated. This might be explained by a high primary production and the oil film preventing degassing of oxygen. The oxygen-rich pond water is beneficial for mineralization (Hvitved-Jacobsen et al. 2010) and reduction of the internal loading of, e.g., P, Fe, and NH₄⁺ (Cooke et al. 2005). In dry winter periods, the oxygen content was typically low (<20 %), which again might be explained by the oil content contributing to a constantly high oxygen consumption, even in the cold water (<10 °C),

while reaeration from the atmosphere was prevented by the oil film. Anoxic conditions might cause a solubilization of metals, leading to a release of nutrients (Cooke et al. 2005) and hence to a reduced efficiency of the system. Oxygen content in the pond varied during rain events. Rain during summer caused decreasing oxygen concentrations due to inflow of oil-containing water and washout of oversaturated water. On the other hand, rain events during winter caused increased oxygen concentrations, when oxygen-rich runoff entered the pond. The tendencies with high/low retention at high/low oxygen concentration were observed during rain events, though less pronounced. The changing oxygen conditions in the pond were reflected with a delayed effect in the stream.

As concrete is an alkaline material, pH was measured continuously (monitoring sites in Fig. 1) and acid dosing was considered necessary. pH in the outlet from the pond and hence in the inlet to the two filters differed between 6 and 9 throughout the investigated year (Fig. S1), with photosynthesis explaining the high pH (Hvitved-Jacobsen et al. 2010). At first, pH in the outlet

from the concrete filter was 10-11 but it decreased over time, as expected from Berg et al. (2005) and Egemose et al. (2012). Acid dosing was stopped after 8 months, as pH had been stable (pH <8.5) for a month and the concrete filter was therefore considered neutralized. During the year, pH varied from 6 to 8 in the stream, indicating that acid dosing was an efficient neutralization method.

Particles and organic matter

The periods March-May and June-August were the driest with regard to runoff (Table 2) and made up only 15 and 13 %, respectively, of the yearly inlet. Nevertheless, more than half (65-66 %) of SS and OM ran off to the pond within this half year (Fig. 2). This might be due to a large deposition of particles in the catchment (primarily roads and parking areas) during spring and summer, due to increased traffic intensity (Vejdirektoratet 2001). A total of 693 kg red ha⁻¹ SS and 321 kg red ha⁻¹ OM entered the pond. The high organic content is unusual in stormwater and is probably related to catchment activities. Organic particles are typically smaller and lighter than inorganic particles, and combined with a high mineralization rate and oxygen demand, the retention potential may be decreased. Considering the high organic load, the annual mean retention of SS and OM was good in the pond (60 and 65 %, respectively) and even better in the concrete filter (78 and 84 %, respectively) and sand filter (77 and 75 %, respectively). The high retentions meant that only 101 kg red ha⁻¹ SS and 38 kg red ha⁻¹ OM were discharged to the stream, of which half came from the overflow. Considering the low pond-to-catchment ratio and the massive groundwater seepage, the retention of SS in the pond was fairly high, though lower than in other wet ponds (80–90 %) with optimal volume-to-catchment ratios of 250 m³ red ha⁻¹ (Hvitved-Jacobsen et al. 1994). This must be due to the large detention volume that, to some degree, compensates for the small wet volume (Sønderup et al. 2014).

The retention in the pond was best when the loading was low (small and/or short events) and decreased with increasing loading (Table S1). This is because the increased runoff loading is directly proportional to increased rain intensity, which leads to a shorter residence time in the pond and hence to a decreased retention (Hossain et al. 2005; Sharma et al. 2011). This was in particular a problem for the examined pond, as it was much smaller than the recommended 250 m³ red ha⁻¹ (Hvitved-Jacobsen et al. 2010) and therefore more affected by the high inflows. A rain event in May 2013 (8.6 mm in 4.7 h) can illustrate the issue. The pond volume was changed 0.8 times during the event and 1.9 times in the following 24 h (Fig. 3). During the event, a high load into the pond and a small discharge resulted in a high retention of SS in the pond (32 kg). In the following 24 h, the inlet load decreased considerably whereas the heavily loaded pond water did not have the sufficient time to settle and was therefore washed out. This resulted in a drop in retention from 32 to 5 kg in the pond, while the previously retained SS entered the filters, resulting in a heavy overloading and a poor retention. This clearly demonstrates that a large wet volume is necessary to ensure sedimentation whereas a



Fig. 2 *Left* mass balance (kg red ha⁻¹) for the inlet, outlet, and overflow from the pond and also for the inlet and outlet from the concrete filter and sand filter. All values for SS and OM are calculated from March 2013 to March 2014 for 3 months at a



time. *Right* measured concentrations of SS and OM in the inlet to the pond, outlet from the pond, and outlet from the concrete filter and sand filter during a rain event in the winter 2014



Fig. 3 Accumulation of SS into and out from the pond and of water into the pond compared to the number of bed volumes (1 bed volume= 30 m^3) during a rain event in May 2013 (8.6 mm in 4.7 h)

large detention volume empties into the receiving water after each rain event, eventually through filters. The retention in the two filters differed considerably from event to event (62–92 %) due to the varying loading of the pond. Nevertheless, the annual mean retention of SS in both filters was in accordance with retention in traditional sand filters (75–99 %) (Hvitved-Jacobsen et al. 2010), though in the lower end of the interval. Again, this can be ascribed to the small wet volume of the pond.

During dry weather periods, the mean concentrations of SS and OM before (34 and 14 mg L^{-1} , respectively) and after (32 and 14 mg L^{-1} , respectively) the pond were quite similar, though the minimum (3–6 and 0 mg L^{-1} , respectively) and maximum (98-218 and 60- 72 mg L^{-1} , respectively) concentrations differed considerably for inlet and outlet. The huge variability in concentrations is due to differences in precipitation (load, duration, and intensity), antecedent dry periods, and catchment activities (Sharma et al. 2011). The outlet concentrations of SS and OM from both filters also differed (0-81 and 0-77 mg L⁻¹, respectively) but showed low mean concentrations from the concrete filter (13 and 7 mg L^{-1} , respectively) and sand filter (6 and 4 mg L^{-1} , respectively). During rain events, the maximum concentrations increased up to 3 times compared to dry weather concentrations, resulting in increased mean concentrations of SS and OM into the pond (36 and 18 mg L^{-1} , respectively), due to particle washout from the catchment. On the other hand, the mean concentrations of SS and OM decreased out of the pond (23 and 9 mg L^{-1} , respectively) and out of both filters (4 and 1 mg L^{-1} , respectively), due to sedimentation and filtration processes.

Despite of the differing concentrations, the retention of SS and OM from intensive rain campaigns was quite stable in the pond (68 ± 20 and 73 ± 18 %, respectively), concrete filter (86±8 and 96±7 %, respectively), and sand filter (89±11 and 94±10 %, respectively). Because of this steady tendency, we have chosen to follow the concentrations of SS and OM during one of the events, a coupled rain event (Fig. 2). Most of the water came the 7th of February 2014 (14 mm in 18 h) and the rest in two parts the following 2 days (2 mm in 3 and 1 h, respectively). The concentrations of SS and OM into the pond peaked at high rain intensities, resulting in two big peaks and one smaller peak. The peaks were all rediscovered in the outlet from the pond, though more attenuated (Fig. 2). It was obvious that rain intensity and washout of particles were connected (Hossain et al. 2005; Sharma et al. 2011). After a peak, the concentrations of SS and OM decreased towards background concentrations of 10 and 5 mg L⁻¹, respectively. Throughout the event, the concentrations of SS and OM from both filters were rather constant at 2–8 and 0–2 mg L^{-1} , respectively. This demonstrates that retention in filters is independent on flow rate.

Ca, Fe, and Al

Both filter materials contained Ca, Fe, and Al, all known for their ability to adsorb P. The amount of Al and Fe was quite similar in the concrete filter (0.8-1.1 t) and in the sand filter (0.1-0.3 t), whereas the amount of Ca was 10-30 times higher in both filters. The initial content of the three adsorbents was 2-6 times higher in the concrete filter compared to the sand filter (Fig. 4). This is



Fig. 4 Initial content of Ca, Fe, and Al in the concrete filter and sand filter and the remaining content after the 1-year study. The difference between initial and remaining content is due to washout or gain

because the cement fraction in concrete contains oxides of Ca, Fe, and Al (e.g., Mechling et al. 2009). This means that crushed concrete initially had a higher P adsorption capacity than sand. After 1 year of operation, the filter content had changed due to sorption and filtration of dissolved and particulate adsorbents from the incoming water and due to washout or dissolution of the adsorbents (Fig. 4).

Ca was mainly dissolved throughout the system, including inlet (96 %) and outlet (98 %) from the filters. The concrete filter lost 140 kg, while the sand filter lost 1200 kg, corresponding to 1 and 32 % of the initial content, respectively (Fig. 4). Because of the high amount of Ca in the concrete filter (9.9 t), the net loss was insignificant for the adsorption capacity. The opposite was true for the sand filter, resulting in a highly reduced adsorption capacity. As washout of DCa from sand filters has not been addressed before, it is unsure whether it will continue in the following years, exhausting the filter regarding Ca. The distribution of Fe and Al was just opposite to Ca, as the majority was particulate, including inlet to the filters (90 and 93 %, respectively). The higher particulate fraction into the demonstration facility came from transformation of DFe (anoxic groundwater) to PFe in the sewer system due to oxidation and from erosion of surfaces in the catchment. After the first year, a net retention of Fe of 9 and 8 % was seen in the concrete filter and sand filter, respectively. Even though the percentages are similar, the concrete filter had the highest retention in kilograms, which is more important considering the binding capacity. The same was true for Al, where the concrete filter retained 28 kg (3 %) and the sand filter retained 11 kg (8%). As the ratios between the adsorbents (Ca, Fe, and Al) and P increased from the inlet to the outlet of both filters, extra binding capacity was added to the downstream environment and might therefore reduce eutrophic conditions with time.

In principle, P binds to Ca, Fe, and Al in the ratio 1:1 (de Vicente et al. 2008a). However, several studies show a higher retention by a higher dosing ratio of 1:4–1:10 (Cooke et al. 2005; de Vicente et al. 2008a, b). Using a P:Ca ratio of 1:10 and the annual P loading on the concrete filter (2.4 kg red ha⁻¹) and sand filter (1.0 kg red ha⁻¹), their remaining adsorption capacity is roughly estimated to be maximum 35 and 22 years, respectively. It should be noted that the initial adsorption capacity (using the same parameters) was 36 and 33 years, respectively, again indicating the massive loss of Ca from the sand filter. Using the same principles as for Ca, the remaining adsorption capacity for Fe and Al is estimated individually in each of the two filters. As opposed to Ca, the capacity did not decrease during the 1-year study but remained 4 (Fe) and 3 (Al)years in the concrete filter and 3 (Fe) and 1 (Al)years in the sand filter. This demonstrates that the yearly retention of Fe and Al in the filters was equivalent to an increased and important binding capacity that, with time, contributes to further P retention. The overall P capacity, from adding all three adsorbent capacities, might be smaller than estimated (e.g., 1:20) because of chemical interference of, e.g., humic acid and silicates (de Vicente et al. 2008a) and because of hidden binding sites within the coarse concrete grains.

Phosphorus

In total, 3.4 kg red ha^{-1} PP and 2.3 kg red ha^{-1} TDP entered the pond (Fig. 5). During the first weeks, a net release from the pond was observed, which could only be caused by leaching from the newly established clay membrane. In the season of growth (May to October), the first submerged macrophytes emerged on the bare membrane and, at the same time, a net retention of 12-62 % TDP (highest in July and August) was observed. This was probably due to biological uptake of P, as plants can remove up to 85 % TDP depending on the species (Bratieres et al. 2008). From November 2013 to March 2014, TDP was released moderately (14 %), probably due to mineralization (Cooke et al. 2005) and decaying vegetation (Oberts and Osgood 1991). The annual retention of TDP (10 %) was guite low, but in accordance with other published results of, e.g., the mean retention (27 %) during 65 events (Hvitved-Jacobsen et al. 2010) and the median winter retention (-2%) in 27 wet ponds (Sønderup et al. 2015). PP was retained in the pond throughout the year, though best during summer (65 %) compared to winter (18 %). This was most likely due to more frequent rain events during winter corresponding to shorter residence times insufficient for settling of PP. The annual retention of PP (39 %) was much lower than the retention of SS in the pond (60 %), demonstrating that the smallest particles are most nutrient rich (Stone and English 1993) and most difficult to remove (Ingvertsen et al. 2011). Egemose and Jensen (2009) found that 38-57 % PP in runoff settled slower than 1 m h^{-1} and that >28 % settled slower than 1 cm h⁻¹. A satisfactory retention would



Fig. 5 *Left* mass balance (kg red ha⁻¹) for the inlet, outlet, and overflow from the pond and also for the inlet and outlet from the concrete filter and sand filter. All values for P are calculated from March 2013 to March 2014 for 3 months at a time. *Right* measured

therefore demand a much higher minimum residence time than the current time of 30 min and thereby a higher pond-to-catchment ratio.

Because of the poor retention in the pond, 1.7 kg red ha^{-1} PP and 1.6 kg red ha^{-1} TDP entered the filters (Fig. 5). The annual mean retention of PP was approximately the same in the concrete filter (80 %) and sand filter (78 %), even though the retention varied throughout the year in both filters. The retention fits with the SS retention in both filters and also with the retention of SS in sand filters of 75-99 % in the literature (Hvitved-Jacobsen et al. 2010). This demonstrates that the filters efficiently removed particles independent of grain size. During the first 2 months, the retention of TDP was high (74–79 %) in the concrete filter but, during the third month, it decreased (24 %), and in the rest of the studied period, the previously retained TDP was washed out of the filter. The sand filter had an equally high retention of TDP in the first quarter (80 %) and then showed a similar but much slower decreasing trend, ending at 13 % retention in December 2013-March 2014. The decreasing trend was expected for concrete (Berg et al. 2005; Egemose et al. 2012) and sand (Arias et al. 2001), but first when the adsorption capacities were exhausted, i.e., not the first 5–10 years. The poor retention might, to some extent, be due to mineralization processes in the filter materials, turning PP into TDP. It should be mentioned that due to different infiltration rates through the filters, the concrete filter received 72 % more water, TDP, and PP, but also oil compared to the sand filter. This meant that the sand filter was favored over the concrete filter, as the latter experienced (1) shorter



concentrations of P in the inlet to the pond, outlet from the pond, and outlet from the concrete filter and sand filter during a rain event in the winter 2014

contact time between filter material and P, (2) faster use of adsorption capacity, and (3) greater impact of oil embedding the concrete particles and reducing the contact surfaces. Based on the results from the pilot study (1 h retention resulted in 90 % TDP removal), the shorter contact time in the full-scale concrete filter (minimum 0.9 h, average 5.9 h) could not affect the P retention significantly. The low adsorption could neither be ascribed to the concrete filter's P capacity, as it was estimated to last 5-10 years. The only issue left that could affect the P removal was the constant inflow of oil to the pond and hence to the two filters. After few weeks of operation, a thin oil film was visible on the filter surfaces and, during summer, the filter materials had turned blackish. A small lab experiment showed that pure and oily concrete reduced a solution of 500 µg DIP L^{-1} by 99.3±0.1 and 25.9±10.7 %, respectively. Notice the pronounced standard deviation when oil is present. Further studies are needed to identify the longterm efficiency of crushed concrete in runoff with less or no oil. Retention of TDP in concrete before oil covering was comparable to natural materials like Polonite, peat, and shell sand (Vohla et al. 2011); amended sand and zeolites (Arias et al. 2001; Ballantine and Tanner 2010); and to industrial by-products and man-made products like drinking water treatment residuals, slag, Filtralite P, Leca, and Phoslock (Ballantine and Tanner 2010; Vohla et al. 2011), all retaining >80 % TDP.

During dry weather periods, the mean concentrations of TDP and PP before (157 and 187 μ g L⁻¹, respectively) and after (165 and 227 μ g L⁻¹, respectively) the pond were quite similar, though with a massive

difference between minimum (12–37 and 35–36 μ g L⁻¹, respectively) and maximum (653-730 and 812-874 μ g L⁻¹, respectively) concentrations for inlet and outlet. Note that the concentrations of TDP and PP are in the same range. The minimum and maximum outlet concentrations of TDP and PP from both filters also differed (0–209 and 13–120 μ g L⁻¹, respectively) but showed low mean concentrations from the concrete filter (101 and 42 μ g L⁻¹, respectively) and sand filter (48 and 40 μ g L⁻¹, respectively). The relative high mean TDP concentration from the concrete filter is explained by the oil-covered filter grains. During rain events, the maximum concentrations increased up to 2.5 times compared to dry weather concentrations, resulting in increased mean concentrations of TDP and PP into the pond (224 and 243 μ g L⁻¹, respectively) but decreased concentrations out of the pond (139 and 189 μ g L⁻¹, respectively). The mean concentration of PP out of both filters was roughly the same as during dry weather, whereas the mean concentration of TDP increased out of both filters (70–128 and 35–47 mg L^{-1} , respectively), due to mineralization processes and washout of loosely bound P.

During a coupled rain event in February 2014, the concentration of PP was higher into and especially out of the pond at the beginning of the rain compared to TDP (Fig. 5). It should also be noted that PP peaked a few hours before SS and OM and several hours before TDP, indicating that PP was flushed with smaller particles than SS and OM and thereby entered the pond sooner. TDP was instead flushed with the largest runoff volume, along with the other dissolved substances. PP

out of the pond was a delayed reflection of the inlet and showed the same concentrations, whereas TDP out of the pond was changing more moderately. The concentrations of TDP and PP from the filters were more or less unaffected by the rain event, just as SS and OM. However, TDP was higher out of the concrete filter than out of the sand filter, which might be due to the higher flow into the concrete filter and thereby the lower residence time and due to release and/or mineralization processes in the filter.

Nitrogen

The inflow of N to the pond was quite high (TN 207 kg red ha⁻¹, NH_4^+ 91 kg red ha⁻¹, and NO_3^- 30 kg red ha⁻¹), which would not be expected when the inlet water consisted of groundwater and runoff (Ingvertsen et al. 2011). Normally in runoff, the concentration of NO₃⁻ would dominate due to oxic conditions in the sewer system though long dry periods could lead to anoxic conditions with NH_4^+ (Gobel et al. 2007). However, NH_4^+ dominated in the demonstration pond not only at the inlet but also in the entire system throughout the year (Fig. 6). These unusual conditions were probably caused by an extensive discharge of urine from the truck area. The lowest N discharge was seen during summer when the truck drivers presumably used the fence rather than portable toilets, obviously emptied in the rainwater grids. This is supported by the large discharge of organic P (0.5 kg red ha⁻¹ year⁻¹ corresponding to 20 % of TDP). The annual mean retention of TN and NH_4^+ in the pond was 39 % for both parameters,



Fig. 6 *Left* mass balance (kg red ha⁻¹) for the inlet, outlet, and overflow from the pond and also for the inlet and outlet from the concrete filter and sand filter. All values for N are calculated from March 2013 to March 2014 for 3 months at a time. *Right* measured



concentrations of N in the inlet to the pond, outlet from the pond, and outlet from the concrete filter and sand filter during a rain event in the winter 2014

with constant retention from May 2013 to January 2014 and a constant release in the remaining months. NO₃⁻ was retained at an equally moderate level (23 %) in the pond, with no obvious pattern in retention or release throughout the year. This is in accordance with the expected retention of TN of around 30 % in lowloaded wet ponds (Hvitved-Jacobsen et al. 2010). In the concrete filter, 8 % TN and 17 % NO3⁻ were retained while 32 % NH_4^+ was released. The sand filter only retained NO_3^- (38 %), while TN and NH_4^+ were released (34 and 128 %, respectively). The retention in both filters was smaller than the expected retention range of 45-70 % TN in sand filters (Birch, et al. 2005; Collins et al. 2010; Hvitved-Jacobsen et al. 2010). This can be explained by ammonification (microbial transformation of organic N to NH_4^+) and inhibition of nitrification by dominance of anoxic conditions in the filters (Collins et al. 2010) due to permanent water cover. A planted filter might enhance N mineralization, nitrification, and denitrification processes, as oxygen and labile organic matter are leaking from their roots (Flindt et al. 1999). Overall, 131 kg red ha⁻¹ TN, 84 kg red ha⁻¹ NH₄⁺, and 19 kg red ha⁻¹ NO₃⁻ were discharged to the stream within a year. The mass balance did not take N cycling of, e.g., nitrification, denitrification, and N₂ loss into the atmosphere into account, though these processes would change masses and thereby reductions (Cooke et al. 2005; Gobel et al. 2007).

The minimum, maximum, and mean concentrations of TN, NH_4^+ , and NO_3^- were roughly the same during dry weather periods and rain events. The variation in minimum and maximum concentrations was 1-86 mg L^{-1} for TN, 0–51 mg L^{-1} for NH_4^+ , and 0– 9 mg L^{-1} for NO₃⁻. The mean concentrations of TN, NH_4^+ , and NO_3^- were generally the same into and out of the pond (8, 4, and 2 mg L^{-1} , respectively), indicating that the N sources and sinks were equal in the pond. The mean concentration of NH4⁺, and thereby TN, out of both filters was higher (4–8 and 6–12 mg L^{-1} , respectively) than the pond concentrations, whereas the mean concentration of NO₃⁻ out of both filters was lower $(1 \text{ mg } \text{L}^{-1})$ than the pond concentrations. These values indicated a high rate of coupled nitrificationdenitrification process in the filters.

During the coupled rain event in February 2014 (Fig. 6) ,TN and NH_4^+ showed the same patterns, which were expected as NH_4^+ was made up 60–99 % of TN. In the first part of the event, the high initial concentration in the inlet (12–17 mg L⁻¹) decreased with significant

amplitudes towards 7 and 5 mg L^{-1} (TN and NH_4^+ , respectively). This resulted in four peaks all rediscovered at the outlet and from both filters, though with much lower amplitudes. It should be noted that the concentration of NH_4^+ and thereby TN out from the sand filter was high (16–19 mg L^{-1}), which could be explained by washout of NH4⁺ either accumulated during anoxic conditions or transformed from organic N (Collins et al. 2010). Anoxic conditions cannot occur during quick percolation, which inhibits denitrification but enhance nitrification (Collins et al. 2010). This was observed as the decrease in NH₄⁺ was followed by an increase in NO₃⁻ in both filters. NO₃⁻ in and out of the pond was constantly below 1.5 mg L^{-1} and changed with the same pattern as TN. At the outlet from the concrete filter and sand filter, NO₃⁻ increased from 0 to 0.8 and 0.5 mg L^{-1} , respectively.

Heavy metals

The mean concentrations of heavy metals in and out of the demonstration facility were calculated for the first month of operation (Table 3). During this period, the inlet to the pond was mainly particulate with 28 % dissolved Ni and less than 6 % dissolved Pb, Cu, and Cr. The only exception was Cd with 65 % dissolved and 35 % particulate matter, which is in accordance with Makepeace et al. (1995). Some of the *dissolved* load of all heavy metals might be bound to small particles or colloids passing through the filter size used. The runoff content of heavy metals was generally decreasing in the following order: Cu>Cr>Ni and Pb>Cd, which is in

 Table 3
 Mean concentrations in micrograms per liter in the inlet to pond, outlet from pond, and outlet from concrete filter and sand filter

		Ni	Pb	Cu	Cr	Cd
Pond in	Par.	6.4	7.3	72.3	26.1	0.5
	Dis.	2.5	0.1	4.6	0.0	1.0
Pond out	Par.	1.1	2.4	6.2	4.2	0.3
	Dis.	2.8	0.1	15.4	0.0	1.0
Concrete out	Par.	0.3	0.6	1.3	0.8	0.1
	Dis.	2.2	0.0	13.2	8.2	1.0
Sand out	Par.	0.3	0.5	1.4	0.8	0.2
	Dis.	15.0	0.0	6.7	0.0	1.0

All values are calculated based on the first month of operation *Par.* particulate fraction, *Dis.* dissolved fraction

accordance with, e.g., Egemose et al. (2014). The particulate fractions were retained efficiently in the pond and the two filters. The same was true for dissolved Pb, whereas the remaining dissolved heavy metals had a poor retention in the system or even a release from the pond (Ni and Cu), concrete filter (Cr), and sand filter (Ni). The result was that Ni, Cu, Cr, and Cd primarily were dissolved (86-96 %), while Pb still was particulate (97 %) in the outlet from the two filters. All heavy metals were though discharged to the stream in highly reduced concentrations. The concrete's ability to retain heavy metals was expected from a study of leaching from different concrete types (Egemose et al. 2012) and from knowing that heavy metals accumulate in concrete buildings in its lifetime (Berg et al. 2005). Birch et al. (2005) found that Cr and Ni leached from a sand filter whereas Cu and Pb had a mean retention of 52-53 %. In our study, both filter materials removed the particulate fraction quite efficiently, when considering that 42-92 % was removed in the pond, resulting in a lower concentration and a smaller grain size of particles to be removed in the filters.

During the coupled rain event (Fig. 7), the particulate fractions of Cu, Cr, Ni, and Pb at the inlet peaked twice during the beginning of the rain event coinciding with high rain intensities and decreased afterwards towards initial concentrations. The outlet concentrations were almost the same though with less variations. Concerning the dissolved fractions of Cu and Ni, only one peak was observed at the inlet and outlet. The concentrations of particulate Cd and dissolved Cr, Pb, and Cd were below the detection limit of 0.1 μ g L⁻¹ throughout the event. The outlet concentrations from the two filters are not shown in Fig. 7 as they were quite stable and below pond values during the event.

The outlet concentrations of dissolved heavy metals from both filters were compared to water quality standards from Makepeace et al. (1995) and Egemose et al. (2014) the first month of operation and during the coupled rain event to determine critical levels. The water quality standard for Ni of 8 μ g L⁻¹ only exceeded the first day of operation from the concrete filter, but for all the samples (8-20 μ g L⁻¹) from the sand filter the first month. The outlet concentrations of dissolved Pb exceeded the water quality standard of 0.3 μ g Pb L⁻¹ a single day $(1 \ \mu g \ L^{-1})$ for each filter, during the first month of operation, but were well below toxic levels (LC₅₀, EC₅₀, and acute toxicity of 1000 μ g L⁻¹, according to Makepeace et al. 1995). Discharge from the concrete filter violated the water quality standard for Cu (12 μ g L⁻¹) for almost all the samples (8– 40 μ g L⁻¹), while discharge from the sand filter was constantly below $(0-11 \ \mu g \ L^{-1})$. As some of the measured dissolved fractions might be colloids, the actual dissolved levels were most probably less critical. According to Makepeace et al. (1995), Cr is a concern for aquatic life at 2–20 μ g L⁻¹, which was largely exceeded during the first month for the concrete filter (4–15 μ g L⁻¹), probably due to colloids. Cr was not detected from the sand filter the first month and neither during the rain event for both filters. The outlet concentrations of dissolved Cd from both filters did not exceed the standard of 5 μ g L⁻¹.



Fig. 7 Measured concentrations of particulate and dissolved heavy metals in the inlet and outlet from the pond. Concentrations of particulate Cd and dissolved Cr, Pb, and Cd were below the detection limit of 0.1 μ g L⁻¹; therefore, data is not shown

Hydrocarbons

During the first month of operation, 0.01 kg red ha^{-1} of C6–C10 entered the pond, along with 3.2 kg red ha^{-1} C10–C25 and 0.7 kg red ha⁻¹ C25–C35. The short chains (C6-C10) were almost not retained in the pond (3 %) and not in the filters, whereas the longer chains (C10-C35) were retained with 96-98 % in the pond. Hereby, the inlet of C10-C25 and C25-C35 to the filters was reduced to 0.06 and 0.03 kg red ha^{-1} , respectively. The concrete filter and the sand filter removed 61 and 53 %, respectively, of C10-C25 and none of C25-C35. The massive hydrocarbon pollution of 4.0 kg red ha^{-1} was reduced to 0.03 kg red ha⁻¹ in the demonstration facility during the studied month. It was mainly diesel, fuel oil, and petroleum products that were retained, whereas gasoline was hardly retained. This demonstrates that reduced discharge of gasoline is most important for the quality in downstream environments. The high retention is comparable to a 5 times reduction of PAH from runoff in wet ponds, and as this retention primarily was due to sedimentation (Wium-Andersen et al. 2011) so is the high removal in the demonstration facility and it is therefore expected to continue. There is little information about the retention of hydrocarbons in filters, but as crushed concrete and sand had an equal retention of the different fractions, this is expected to apply to most other filter materials.

The concentration of C10–C35 during a coupled rain event followed the pattern of particulate heavy metals, with two peaks at the beginning of the event (Fig. 8).



Fig. 8 Measured concentrations of hydrocarbons in the inlet and outlet from the pond. Concentrations of C6–C10 were below the detection limit of 10 μ g L⁻¹; therefore, data is not shown

The highest concentration (1.6 mg L^{-1}) was observed for C10-C25. After 1.5 days, the concentration of C10-C35 had almost decreased to the initial concentration $(<0.1 \text{ mg L}^{-1})$. The concentration of C6–C10 was below the detection limit of 10 μ g L⁻¹ at the inlet and outlet from the pond and from the two filters. In addition, the outlet concentrations of C10-C35 and C25-C35 from both filters were below the detection limits of 40 and 50 μ g L⁻¹, respectively, throughout the event. All measured concentrations were in the lower end of the range (0.6-19.7 mg L⁻¹) of typical hydrocarbon concentrations in stormwater (Makepeace et al. 1995). During the first month of operation, a single extreme inlet concentration of total hydrocarbon (130 mg L^{-1}) was discovered, though with corresponding outlet concentration of 0.07 mg L^{-1} . This indicates an occasional high oil spill from the catchment.

Conclusion

Regarding retention of the particulate fractions, the results clearly show that a filter of either sand or crushed concrete improves the retention of SS, OM, and P. During 1 year, 40-65 % was retained in the wet pond and, from the discharged fraction, 75-90 % was retained in both filters, resulting in an overall retention of 90-95 % in the treatment facility. Note that the particles to be removed in the filters are smaller and of lower concentration due to sedimentation of large particles in the pond. Concerning the filters, it is clear that a filter of crushed concrete is just as efficient, if not better, than one of sand. This is true for SS, OM, TN, P, Ni, Cu, Cr, and Cd. The sand filter was only more efficient regarding retention of particulate Pb. Crushed concrete retained all examined particulate and dissolved heavy metals, except dissolved Cr, while the sand filter released dissolved Ni to the receiving stream. This demonstrates that even though some loosely bound heavy metals are washed out from the filters during the first month of operation, the retention of dissolved and particulate heavy metals are generally improved, when filters are attached to the ponds.

It is more difficult to conclude on the retention of the remaining dissolved substances in the pond and both filter materials. The pond was only able to retain TDP during summer, and the TDP retention in both filters dropped significantly after the first months of operation. The sand filter seemed better at retaining NO_3^- than the pond and concrete filter, and only the pond was able to retain NH_4^+ . The three substances are though subjected to uncertainty, as the calculations do not account for mineralization processes and losses to the atmosphere (only N). As both filters are permanently covered by water, they are dominated by anoxic conditions, inhibiting nitrification and Fe-P adsorption. Therefore, the retention of N and P could benefit from a planted filter, with oxygen leaching from the roots. The poor TDP retention in both filters was not reflected by the content of Ca, Fe, and Al. Despite a small washout of Ca in the concrete filter, the content of P adsorbents increased, thereby prolonging its lifetime regarding P retention. The small increase of Fe and Al in the sand filter could not make up for the massive loss (32 %) of Ca, resulting in a decreased content of adsorbents.

The different grain sizes and infiltration rates for the two filters caused more water and i.a. P and oil to enter the concrete filter compared to the sand filter. Additional studies showed that oil-covered concrete grains were far less efficient than oil-free concrete, which explained the low TDP retention after the first months. On the other hand, the pond and both filters retained 99.3 % of the oil though retention of the short carbon chains was inefficient (<3 %). Another study without oil showed that at equal flow rates, the concrete filter had a better retention of TDP than the sand filter and a good retention of PP, independent of residence time. These findings suggest that a filter of crushed concrete is able to retain either oil or P, dependent of the oil content in the runoff.

A wet pond filter and/or a sand filter is good at removal of the particulate fraction, but when it comes to removal of the dissolved contaminants, other filter materials are needed. This could be crushed concrete, as the high content of Ca, Fe, and Al results in a high retention of TDP but also dissolved heavy metals. Before commercialization of concrete as a filter material, further studies regarding the long-term retention of TDP without massive oil influence are desirable. Crushed concrete is a cheap waste material, and by obtaining a second application as a filter material, the sustainability of the concrete products is improved. If the concentration of xenobiotics is low, concrete might afterwards be used as soil amendment or for P recovery. Acknowledgments Thanks to Arwos (owner of the pond) for the renovation of the existing pond and construction of the two filters and for fruitful cooperation throughout the project. We thank the lab technicians and student assistants at the University of Southern Denmark for their help with chemical analysis. Thanks to Grontmij, Aquasense, Cowi, and BHC Miljø for stimulating collaboration and/or funding for the project. The study was supported by (1) the industrial/commercial PhD project (Melanie J. Sønderup) granted by the Danish Ministry of Science, Innovation and Higher Education FI case number 11-+109519; (2) the Centre for Lake Restoration, a Villum Kann Rasmussen Centre of Excellence; and (3) the project granted by the Danish Eco-innovation program from the Danish Ministry of the Environment.

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