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Evaluation of a sand filter material for road runoff treatment– pilot-scale field trial focused on copper and zinc removal

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ABSTRACT

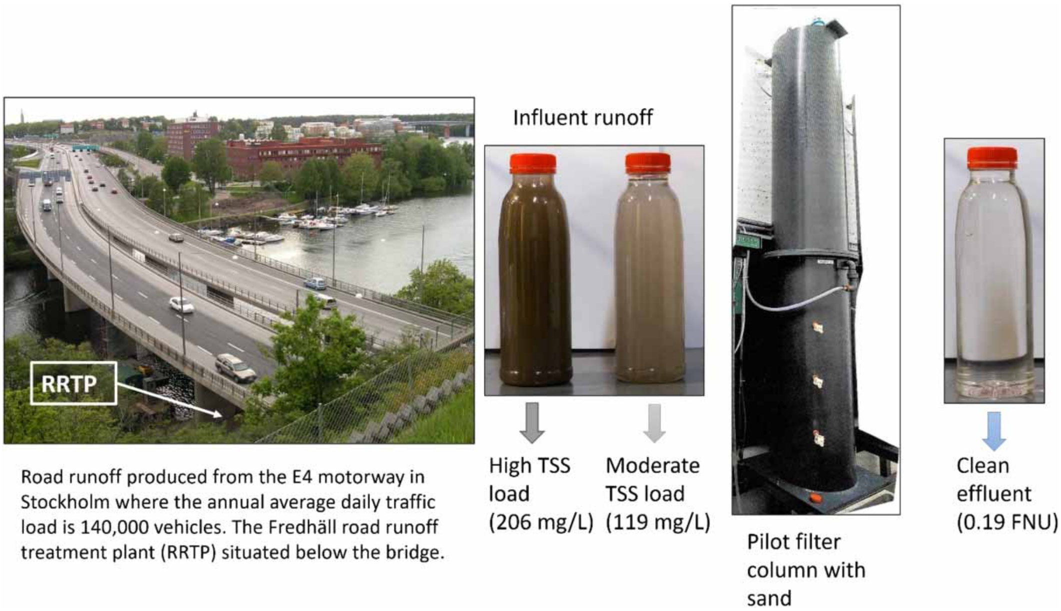
The effects of stormwater discharges on receiving aquatic environments and the need for their purification were highlighted by an EU court in May 2020. The ruling stated the need for removal of dissolved pollutants, which justifies field studies for development of far-reaching methods for runoff treatment. In this study, a standard sand was used as medium for road runoff filtration and removal of dissolved and particle-bound (<0.45 μm) zinc (Zn) and copper (Cu). Data included 24 road runoff events, mimicking the flow variations and pollutant emissions over a seven-month period. The findings showed that sand can be used to remove Zn and Cu from road runoff in a gravity fed treatment system at a surface load ranging from 16.8 to 201 L m^{-2} h^{-1}. The removal of total Zn and Cu was 93 and 67%, respectively. Dissolved Zn was efficiently removed by the sand (87%), however not Cu (19%). The sand efficiently removed total suspended solids (TSS) from the maximum occurring 443 mg L^{-1} to below 5 mg L^{-1}. No head loss due to the TSS loadings was observed. The sand's potential to remove the investigated metals was shown, but in the longer term, effluent concentrations may exceed permitted values.

Key words: clogging, column experiment, dissolved metals, head loss, road runoff treatment plant, stormwater

HIGHLIGHTS

- Removal of Cu and Zn in runoff from the E4 motorway in Stockholm was studied for the first time in a sand filtration system.
- Total Zn, as well as the dissolved phase, was removed to concentrations close to the most stringent effluent values applied in Sweden for stormwater.
- Cu occurs as dissolved species and its removal from road runoff requires more treatment in addition to sand filtration.
- Sand is suggested to be used as a cover layer when reactive filter media are used for road runoff treatment.

GRAPHICAL ABSTRACT



INTRODUCTION

The adverse effects of stormwater discharge on receiving water environments and the need for its treatment was recognized in the 1960s. However, in the EU, the importance of pollutant control measures for stormwater were emphasized for the first time in 2000 through the implementation of the Water Framework Directive (2000/60/EC). During the last years, an increased focus on stormwater and deterioration of receiving waters has been observed, particularly in the ruling in May 2020 of the EU Court in case C-535/18 (Official Journal of the European Union 2020). The ruling made it clear that the chemical status of a water body should not negatively be impacted by discharge or activities in the water body.

In the case of metals occurring in elevated concentrations in runoff, it is known that they have a strong affinity to particulate matter (Huber *et al.* 2016a; Baum *et al.* 2021). Therefore, the general method of treatment of stormwater is sedimentation in ponds. However, dissolved material cannot be removed by sedimentation. The abatement of dissolved metals can be carried out by e.g. filter materials (Reddy *et al.* 2014). The advantages for their use are e.g. high hydraulic conductivity, environmentally benign, adsorptive/reactive, easily available, less costly, and easily replaceable materials (Reddy *et al.* 2014). The time that elapses between the run off events is then important, as it gives extended sedimentation times. Pettersson *et al.* (1999) found that the pond removal efficiency of total suspended solids (TSS) reached up to 84% for stormwater collected from impervious catchment areas. They concluded that the treatment pond surface to catchment area should be at most 250 m^2 ha^{-1}, because larger ponds would not yield higher TSS removal efficiency. Hallberg (2007) performed an *in situ* study on road runoff and presented an empirical model for calculating sedimentation time based on inlet and outlet concentrations of TSS. The study showed that several days between runoff events were needed to obtain a TSS concentration below 50 mg L^{-1}. Larm & Hallberg (2008) compared several studies of stormwater detention ponds and found that a minimum outlet concentration of TSS from a pond would be in the range of 5–32 mg L^{-1}.

Zinc (Zn) and copper (Cu) are metals generally appearing in the highest concentrations in road runoff. Corrosion of materials, driven mainly by low water pH, causes the release of Zn and Cu to the environment (Galster & Helmreich 2022). In this paper, we decided to choose these two metals in our study of sand as filter medium, because of their occurrence in high concentrations found in a pre-study (Hallberg *et al.* 2021) as well as their harmfulness in the water environment. A review by Huber *et al.* (2016a) showed that Zn, Cu, nickel (Ni) and cadmium (Cd) occur at a higher fraction in the dissolved phase, i.e., the metals passing through a 0.45-μm-pore-diameter membrane filter. In the study by Baum *et al.* (2021), the highest metal concentrations in urban stormwater were found in the particle size fractions below 63 μm. Hallberg *et al.* (2007) showed that dissolved concentrations of zinc during summer and winter conditions were 100 μg L^{-1} and 96.5 μg L^{-1}, respectively. The corresponding concentrations for copper were 20.31 μg L^{-1} and 18.90 μg L^{-1}, respectively. The implication of these observations is that treatment of road runoff must go beyond sedimentation, particularly in light of increased demands for low pollutant concentrations in the effluent. According to the Federal Soil Protection Law in Germany, the maximum general allowable pollutant concentrations for Cu and Zn discharged after a treatment device (Sustainable Urban Drainage System, SUDS) should not exceed 50 and 500 μg L^{-1}, respectively (Dierkes *et al.* 2015). National limit values have not yet been determined in Sweden for emissions of metals via stormwater. However, local guideline values are established and often based on background metal concentrations in surface water or those of the receiving water body (Järfälla kommun 2016). These values can for Cu and Zn be set as low as 4.5 and 7.5 μg L^{-1}, respectively.

Hence, the increasing stringent discharge demands call for removal of colloidal material and dissolved material. Huber *et al.* (2016a) pointed out that the 0.45 μm filtrate might contain fine colloids and particulate material in the nano-meter range. Bevers *et al.* (2020) found that zinc oxide particles, making up 21% of the mean zinc mass, had a uniform mean size of 40.2 nm in urban river water. It is then evident from the mentioned studies that the stormwater treatment must go further than only to sedimentation.

The use of sand filters to improve water quality has attracted interest as a purification measure in recent decades (Jaeel & Abdulkathum 2018; Zarezadeh *et al.* 2018). The primarily design criteria have been the removal of TSS, which was possible to obtain by at least 80% of mass (e.g. Zarezadeh *et al.* 2018). However, other constituents such as Cu and Zn did not reach acceptable stormwater benchmarks, showing a removal of 37.4% and 42.7%, respectively (Zarezadeh *et al.* 2018). Stormwater detention ponds constructed in Denmark with added sorption techniques showed promising treatment results after sand filtration (Istenič *et al.* 2012). However, the ponds were planted with two macrophyte species, i.e., the systems were rather engineered as constructed wetlands than conventional sand filters. The TSS concentration varies significantly during runoff events, particularly in stormwater from motorways where concentrations over 1,000 mg L^{-1} can be observed (e.g., Larm & Hallberg 2008). If a physical filter for gravity flow is used, it is imperative that prior the treatment with any filter media, the reduction of TSS in a sedimentation facility is needed to prevent clogging. However, even with low TSS concentrations and hydraulic loadings to the filter, a continuous head loss will occur in the long-term due to clogging (Kandra *et al.* 2014; Hallberg *et al.* 2021).

The inability of sand filters to eliminate dissolved and colloidal pollutants in stormwater has led to the search for complementary measures, e.g. by enriching the sand with iron or aluminum (Istenič *et al.* 2012; Fairbairn *et al.* 2018). However, the area-demanding construction of detention ponds, wetlands and large-scale sand filters is problematic at traffic routes in metropolitan areas. In the case of Stockholm, the Swedish Transport Administration has recently initiated a project, which aim to investigate reliable and sustainable treatment methods for dissolved contaminants. We present in this paper, a part of an on-going pilot-scale column experiment within that project, where filter sand is tested for its ability to remove TSS and metals. Filter sand is challenged by other materials that may have a much better ability to bind metals (Reddy *et al.* 2014; Huber *et al.* 2016b). However, some of these proposed materials are alkaline, such as Leca Filtralite P, which can cause high pH (>10) in the filtrate (Pla *et al.* 2021). This side effect usually appears during the initial filtration but still can question if the alkaline water can be discharged directly to a water body, or it should be neutralized by appropriate methods (Higgins *et al.* 2018; Retka *et al.* 2020). The availability of materials for the construction of large filter systems is likely to be a much greater problem. Sand, of the quality desired for filter purposes, is not always available in Sweden as this natural resource becomes scarce. Instead, authorities propose an increased use of rock-crushed material, according to the Swedish Environmental Goals (Swedish Geological Survey 2020). Sand is generally an important raw material throughout the world in the construction industry and supply may decrease in the future and become a limited resource (Edwards 2015).

The sand filter is usually a state-of-the-art solution for cleaning raw water to drinking water standard (e.g., Verma *et al.* 2017; Freitas *et al.* 2022). The widely used methods in stormwater treatment is the infiltration basins and wet ponds where sand can be used as an integrated part (Egemose 2018). It is still interesting to assess the use of sand or rock-crushed material as media for treatment of traffic stormwater and we predict an increase of constructions of filter systems with sand alone or sand filters preceding filtration with so-called reactive (Rodríguez-Gómez *et al.* 2021) or sorptive (Huber *et al.* 2016b) filter media. The sand is expected to reduce the TSS load to the subsequent reactive filter, the life span of which can then be increased because of less clogging and better metal sorption capacity. Moreover, this research also opens the door to a recovery and recycling strategy for the spent materials. The sand and the metal-saturated filter media should go to a process where valuable metals, emitted from the road environment, can be extracted. We believe that at least sand can be reused several times after such a recovery process but also reactive media after re activation process.

The objective of this study was to investigate sand as a medium for road runoff filtration, particularly with reference to its capacity to remove Zn and Cu, dissolved or bound to particles <0.45 μm. To the best of our knowledge, this *in situ* study is the first of its kind where a pilot-scale sand filter is tested during real road runoff events, mimicking the flow variations and pollutant emissions over a seven-month period. The road runoff used in this study was collected after the grit chamber of a road runoff treatment plant (RRTP) before addition of flocculent agents. The treatment condition for the overwhelming majority stormwater canalisation systems is that gravity flow prevails, implying large flow variation to the RRTP. With flow variation follows variation in TSS, water pH and turbidity, parameters which are of utmost importance for the design of a filter treatment solution operated in full scale.

MATERIALS AND METHODS

Study site – Fredhäll RRTP

An experimental pilot-scale filter system (Figure 1) was constructed and placed in the control room belonging to the Fredhäll RRTP, Stockholm, Sweden, which is built below a road bridge and tunnel. The plant treats road runoff from the E4 motorway where the annual average daily traffic load is 140,000 vehicles. A detailed description of the Fredhäll RRTP and its catchment area is found in Hallberg *et al.* (2007).

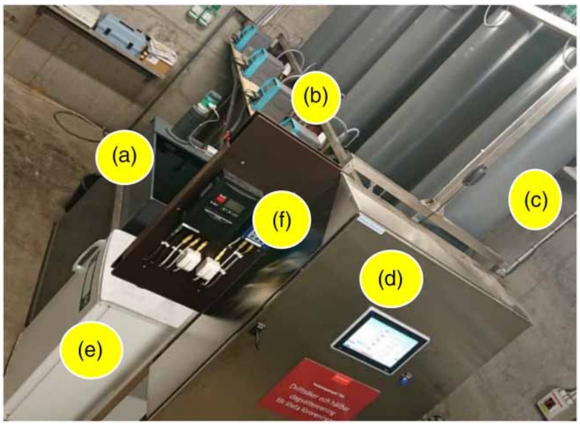


Figure 1 | Photo of the experimental system in the RRTP at Fredhäll. (a) Collection vessel for road runoff; (b) peristaltic pump for pumping to the filter sand column (c); (c) filter column with filter sand; (d) electrical panel with PLC for operation and control of the pilot plant; (e) refrigerator for collection and storage of flow proportional samples from the collection vessel (a); (f) on-line measurement instruments of the road runoff pumped from the collection vessel.

How the pilot-scale filter system was operated

The filter system was constructed to test stormwater purification of filter materials in five columns (Figure 1). Water was pumped during a runoff event from the grit chamber of the RRTP to the collection vessel of the filter system (Figure 1(a)) by a peristaltic pump. The filter column (Figure 1(c)), filled with sand and coarse sand (Figure 2), was made of PVC with an inner diameter of 39 cm. The total height is 246 cm of which 90 cm is employed for the filter section. Influent runoff was pumped and distributed 10 cm over the filter surface via a perforated pipe (Figure 2(c)). The outlet is located 5 cm below the surface of the filter sand (Figure 2(b)), accommodating for saturated conditions during filtration and a constant head of water. An ultrasonic sensor of type SonoiX from MJK Automation AB, Sweden, was placed inside the top of the column to control head loss due to clogging. The washed filter sand and washed coarse sand was received from Rådasand AB, Sweden. The physical properties of the materials are given in Table 1. A programmable logical control (PLC) controlled the flow from the peristaltic pump so that it exactly followed the flow variations to the STP during runoff events. The minimum and maximum surface loadings to the column were 16.8 $L/m^2\,h^{-1}$ and 201 $L/m^2\,h^{-1}$, respectively, generating the hydraulic retention time (HRT) of 1.2 to 16 h depending on the flow during a runoff event.

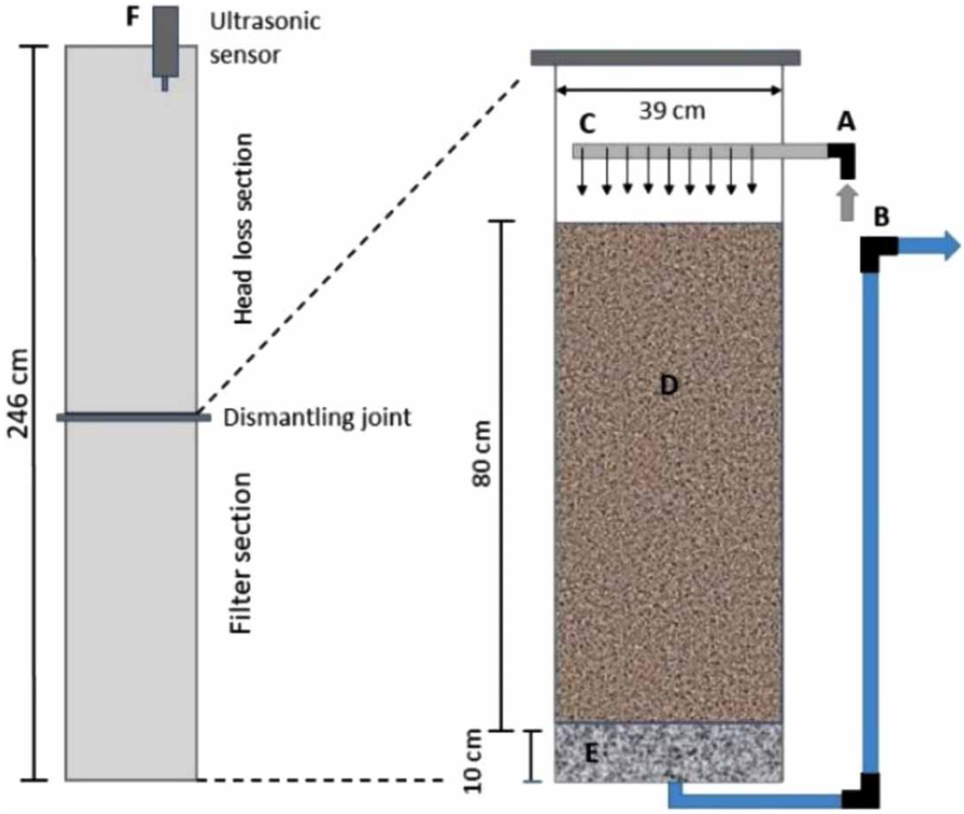


Figure 2 | Column set-up with filter sand. (a) Influent water; (b) Effluent water; (c) Inlet distribution pipe; (d) Filter sand, 80 cm; (e) Drainage layer, 10 cm; Head loss was monitored with an ultrasonic sensor (f).

Table 1 | Properties of the filter sand and the drainage layer with coarse sand

Material	Particle size range (mm)[a]	Particle size (d_{10}/d_{60})[a]	Uniformity Coefficient[a]	Pore vol. (%)	Dry density (g/cm^3)[a]	pH	Hydraulic conductivity ($m\ d^{-1}$)[b]
Filter sand	1.2–3	1.3 (2.4)	1.85	33	1.53	6.00	504
Coarse sand	3–5	n.d.	n.d.	41	1.44	6.21	n.d.

[a]Data obtained from the manufacturer, [b]Saturated hydraulic conductivity was measured using the falling-head method. n.d. = no data.

Water sampling and analyses

Cerlic ITX and Cerlic pHX probes were installed in the collection vessel for on-line measurement of road runoff turbidity and pH. The filter column was fed with a Watson Marlow Qdos30 peristaltic pump. MJK 780 Liquid Vacuum Samplers were used for flow proportional sampling of influent water and effluent filtrated water. Flow proportional sampling was executed every 30 L of the influent water. Sampling of effluent, i.e., the filtrate was done with an interval of every 5 L. The automatic sampler collected a volume of 200 mL each time. The sampled incoming runoff was collected in a polyethylene (PE) plastic container (10 L volume), which was stored in a refrigerator at 4 °C. The effluent samples were collected in PE plastic containers (5 L volume) and stored in the existing room temperature (appr. 12 °C). A dedicated PLC controlled the operation of the pilot plant (Figure 1(d)). Before transport to the laboratory, turbidity and pH of the composite samples were measured using a WTW Turb 430 IR portable turbidity meter and a WTW Portable meter ProfiLine pH 3110, respectively. Samples for metal analyses, TSS, total organic carbon (TOC) and dissolved organic carbon (DOC) were collected in acid washed 60 mL PE plastic bottles, 1,000 and 250 mL plastic bottles, respectively.

The water samples were transported on the same day as the sampling took place to the ALS Scandinavia AB accredited laboratory, Danderyd, Sweden. Analysis of total and dissolved Zn and Cu was carried out using

ICP-SFMS according to SS-EN 17294-2:2016 and US EPA Method 200.8:1994 after digestion with nitric acid. The Sartorius 0.45 μm filter was used prior the analysis of the dissolved fraction. Analysis of TOC and DOC used IR detection method according to CSN EN 1484, CSN EN 16192 and SM 5310. TSS were analysed using SS-EN 872:2005, revision 2, using a 1.6 μm filter.

The low particulate concentration in the column filtrate poses a problem to accurately measure TSS, thus turbidity was used to assess reduction of particulate material in the sand filter effluent. The estimation of TSS from turbidity values can be performed thanks to the good correlation between these two parameters (Hallberg 2007). This indirect measurement of TSS, allows for a high resolution when particularly calculating low TSS concentrations in the treated runoff. In the present study, a good correlation was found ($R^2 = 0.95$) according to the fitted linear expression (1).

$$\text{TSS} = 0.16 * \text{Observed On} - \text{line turbidity} + 17.6 \tag{1}$$

Data analysis

The removal capacity of the filter sand (R_s, %), i.e. the retention or leaching of total or dissolved Zn and Cu during the filtration of stormwater, was calculated according to the equation:

$$R_s = \left(1 - \frac{C_e}{C_i}\right) 100 \tag{2}$$

where C_i is the average influent concentration and C_e is the average effluent concentration for all sampled runoff events.

The sorption of metals, i.e., Zn and Cu, to the filter matrix (S_m, μg g^{-1}) was calculated using the following mass balance equation:

$$S_m = \frac{\sum_{n=1}^{20} (C_i - C_e)\, V_n}{m} \tag{3}$$

where C_i and C_e are metal concentrations in the influent and effluent, respectively, and V (L) is the volume of road runoff treated during a runoff event and m the mass (kg) of the filter sand in the column.

All data obtained through the column trial were statistically treated for analysis of variance using SigmaPlot for Windows software version 13. Only correlations significant at $p<0.05$ are reported in this study.

RESULTS AND DISCUSSION

Runoff events and flow variations

The study was conducted between May 2021 and January 2022. During that period, 24 runoff events occurred for which 82 pore volumes (PVs) were treated in the sand filter (Table 2). In 20 runoff events, flow proportional samples were taken in the runoff and the sand filter effluent. Only the filter effluent was flow proportional sampled during event 7 and for events 10, 16 and 17, flow proportional sampling was not executed (Table 2) due to repair of sampling system. Minimum and maximum flow was achieved to the filter column for most of the runoff events (Table 2). The maximum flow to the filter column was reached for short periods of time, which also would be the case in a full-scale plant when excess flow would be by-passed to avoid overloading of the filter system during extreme runoff events (Figure 3).

Observations of pH, TSS, turbidity and head loss due to clogging

The pH in the runoff varied between 7.1 and 7.7 (Table 3). The measured pH in the filtrate (column effluent) was slightly but not significant higher and varied between 7.4 and 8.2 ($p>0.05$). The TSS concentration in the runoff varied between 12 to 443 mg L^{-1}. After column filtration, the turbidity ranged from 0.3 FNU to 10 FNU (Table 3) corresponding to a TSS concentration of less than 5 mg L^{-1} in all filtrates, showing a good removal of particulate material by the filter sand medium. During the December and January events (events 21–24), an increased TSS concentration was expected due to the use of studded tires on vehicles (e.g., Hallberg *et al.* 2007; Hilliges

Table 2 | Overview of runoff events and sampling

Event no.	Event (Start)	ADP (days)	PVF (n)	AFF (L h^{-1})	MFF (L h^{-1})	FPS (no.)
1	2021-05-05		4.1	16	23	25
2	2021-05-09	4	6.8	10	24	16
3	2021-05-14	4	9.5	10	19	17
4	2021-05-24	7	14.1	4	22	25
5	2021-05-27	1	17.0	4	10	19
6	2021-06-08[a]	11	18.9		n.d.	n.d.
7	2021-06-12	2	22.6	3	24	20[b]
8	2021-06-21	7	23.2	2	7	4
9	2021-06-25	4	25.0	3	22	8[c]
10	2021-07-06	10	28.4	3	24	0
11	2021-07-22	12	32.3	3	24	24
12	2021-08-07	5	39.4	7	24	25
13	2021-08-10	2	41.7	6	24	23
14	2021-08-16	6	48.2	5	24	25
15	2021-08-25	7	53.5	5	24	25
16	2021-08-27	1	59.6	4	24	0
17	2021-09-22	9	60.3	3	24	0
18	2021-10-15	1[d]	63.6	4	24	21
19	2021-11-03	13	65.9	4	9	9
20	2021-11-07	3	72.0	6	22	25
21	2021-12-10	31	75.3	4	12	21
22	2021-12-14	3	76.2	4	9	8
23	2021-12-29	15	78.5	4	9	20
24	2022-01-02	3	82.1	4	14	21

ADP = antecedent Dry period before event; PVF = number of pore volumes filtered; AFF = average flow to filter; MFF = maximum flow to filter; FPS = number of flow proportional samples taken on effluent filtrate during event.
[a]Logged data lost for event six. Data calculated from monitored precipitation in Stockholm by SMHI, the Swedish Meteorological and Hydrological Institute.
[b]Only sampling of filtrate (effluent).
[c]Analysis only of total metals.
[d]The pilot plant was closed from 2021-09-23 to 2021-10-15 for service.

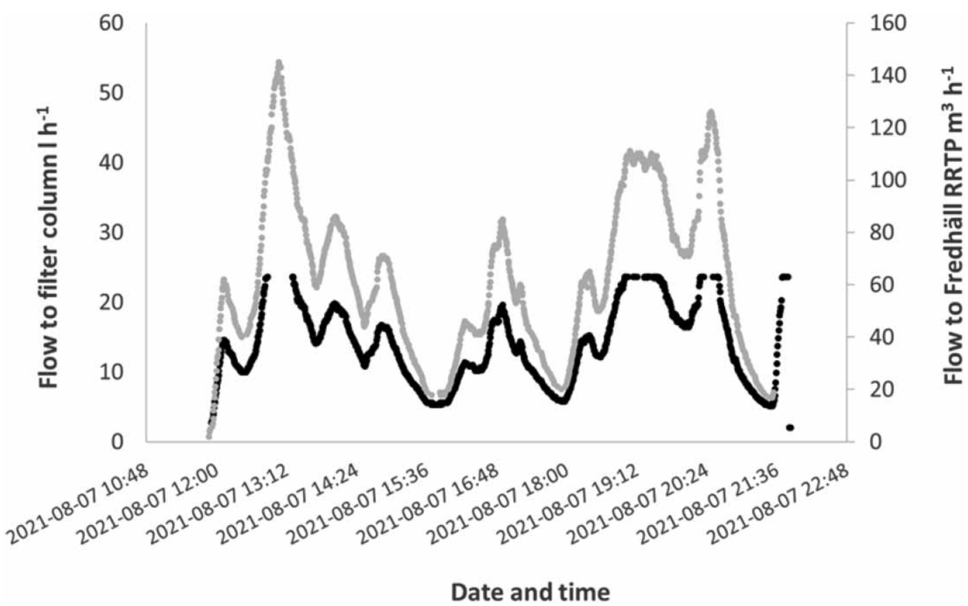


Figure 3 | Runoff event 16, exemplifying the flow variations and peak flows to the filter column during runoff events. Grey line = Flow (m^3 h^{-1}) to the Fredhäll RRTP. Black line = Scaled flow (L h^{-1}) to the filter column.

Table 3 | Turbidity, TSS and pH in filter influent (variable$_{in}$) and effluent (variable$_{eff}$)

Event (n)	TSS_{in} (mg L^{-1})	pH_{in}	$Turbidity_{eff}$ (FNU)	pH_{eff}
1	143	7.7	n.d.	n.d.
2	167	7.7	4.2	7.6
3	59	7.6	2.0	7.6
4	88	7.3	2.2	7.6
5	68	7.4	7.6	7.6
6	n.d.	n.d.	10	7.7
7	35	7.2	1.1	7.4
8	12	7.5	0.6	8.1
9	25	7.4	0.3	8.2
10	74	7.1	n.d.	n.d.
11	14	7.2	0.9	7.9
12	46	n.d.	3.3	8.2
13	65	n.d.	4.9	7.9
14	74	n.d.	1.5	7.8
15	143	7.7	1.8	7.5
16	142	n.d.	n.d.	n.d.
17	7	7.2	n.d.	n.d.
18	44	7.0	n.d.	n.d.
19	22	7.1	2.5	7.4
20	157	7.0	2.5	7.5
21	443	7.4	8.0	7.6
22	172	7.2	8.0	7.6
23	131	7.4	2.2	7.8
24	204	7.2	2.2	7.8

n.d. = no data.

et al. 2017). The runoff events 21 and 22 caused an elevated turbidity (8.0 FNU) in the filtrate. However, the turbidity was low again in the subsequent events (Table 3). These four runoff events were characterized by snowmelt and small waterflow (Table 2) to the filter column.

The head loss was below 10 cm and did not change during the experiment. Our study was carried out *in situ* and in real conditions including natural flow variations. This would suggest that keeping a constant water level during filtration is of importance for the even distribution of water over the sand surface, to avoid propagation of preferential flow as well as local clogging. This is of particular interest with emphasised flow variations during runoff events. A potable water filter is typically a sand with d_{10} and d_{60} of 320 and 520 μm respectively, and with a uniformity constant of 1.63, while a rapid sand filter has a sand with d_{10} and d_{60} of 870 μm and 1,112 μm respectively, and with a uniformity constant of 1.29 (Swedish Water 2010).

Siriwardene *et al.* (2007) studied clogging in a filter consisting of an upper layer of gravel (0.9 m) and beneath a soil layer (0.7 m), corresponding to a very fine sand. The gravel in their study had a d_{10} of 6.4 mm, d_{50} of 10.5 mm and d_{90} of 13 mm. In one trial, they used semi-artificial stormwater with a TSS concentration of 148 mg L^{-1} and kept the water level constant. The median particle size (d_{50}) in the inflow dropped from 40 to 3 μm after the gravel demonstrating good removal of fine particles. After more than 500 PVs treated, no drop in hydraulic head was observed for the layer with gravel, i.e., no clogging occurred. However, the hydraulic pressure in the sand layer gradually declined to zero indicating a development of a clogging layer at the interface between gravel and sand filters. The filter sand in our study had a d_{10}, d_{50} and d_{90} of 1.35, 2.05 and 2.8 mm respectively, thus consisting of a much finer material than the gravel used in the experiment by Siriwardene *et al.* (2007). Our experiment terminated after treatment of 82 PVs where varying high and low influent TSS concentrations occurred. The sand column was fixed to have a constant water level (Figure 2); however, it increased temporarily depending on the discharge of road runoff. Clogging did not occur during the study period and within the head loss of 10 cm mentioned before. We expected a development of a filter cake just below the sand filter surface i.e., accumulation and formation of a so-called 'Schmutzdecke' (Verma *et al.* 2017), also called a colmation layer (Vollertsen *et al.* 2008). This layer will have a much lower hydraulic conductivity than the filter sand, which will limit its hydraulic loading capacity especially if the colmation layer grow to deeper parts of the sand filter (Vollertsen *et al.* 2008). Siriwardene *et al.* (2007) showed that water level fluctuations had impact on the distribution of sediment where clogging at the filter/soil interface will be rapid in systems that regularly empty.

Egemose (2018) showed that organic material was associated to smaller particle size fractions (<63 μm) in the upper layer of a slow sand filter for runoff treatment from an urban catchment area. Particles in stormwater also have mineral origin, which was shown by Bevers *et al.* (2020). Siriwardene *et al.* (2007) concluded that the main driver in the development of the clogging layer is sediment particles less than 6 μm in diameter.

Presence and removal and ratio between total and dissolved fractions of Cu and Zn and causes of changes during filtration

The total and dissolved concentrations of Zn and Cu in the influent un-treated runoff water and effluent filtrate are presented in Table 4. The average removal of total concentrations of Zn and Cu by the sand was 93 and 67%, respectively, while corresponding values for dissolved fractions were 67 and 19%. This high removal efficiency was not shown for Zn in event 5 where the Zn mass loading to the column was 11,115 μg when the mass in the filtrate was 10,925 μg, i.e., corresponding to a removal of only 1.7%. During runoff event 5 an overall release of 7,737 μg of dissolved Cu was observed. The mass calculation showed that 3.53 μg Zn g^{-1} sand, and 0.65 μg Cu g^{-1} sand was retained after treatment of 82 PV. The maximum sorption capacity of Zn and Cu (q_{max}) by the sand used in this study was not determined. Few data exist on sand's q_{max} or retained mass in literature, probably because they are considered as inert media (e.g. Haynes 2015). The starting point is mostly that the sand must be coated with iron-oxides or other compounds aiming at enhanced metal sorption during filtration (e.g. Okaikue-Woodi *et al.* 2020; Zhong *et al.* 2021). However, Zhong *et al.* (2021) performed batch experiments with Zn and Cu solutions and determined the adsorption capacity of a raw sand (0.5–0.6 mm) of unknown origin to 10.4 and 15.1 μg g^{-1}, respectively. It is likely to assume that the sand used in our experiment was far from reaching q_{max} and that metals were trapped in the superficial layers as shown by Hermawan *et al.* (2021).

Table 4 | Total and dissolved concentrations of Zn and Cu in influent runoff and treated effluent and, removal efficiency

	Influent, un-treated runoff (μg L^{-1})		Effluent, treated runoff (μg L^{-1})		Removal (%)	
Element	**Total**	**Dissolved**	**Total**	**Dissolved**	**Total**	**Dissolved**
Zn	223 ± 132	75 ± 62	14 ± 26	10 ± 25	93	87
Cu	64 ± 32	26 ± 13	21 ± 30	21 ± 31	67	19

Data given as rounded mean ± standard deviation values of flow-proportional samples. ($n = 20$)

Figures 4 and 5 shows the ratio between dissolved and total fractions i.e., ratio 1 shows that the metals are completely dissolved while ratio closer to zero shows the opposite, metals mainly bound to particles (<0.45 μm). The ratio for Zn varied around 0.5 from event 8 to event 24 (Figure 4). Copper occurred mainly as dissolved species in the effluent, while the particulate bound fraction dominated in the influent (Figure 5). However, during the two initial events, the sand filter seemed to be unable to trap particulate bound Cu. Thereafter followed an increase and dominance of dissolved Cu in the effluent.

The initial period of 1 to 7 events seems to be a built-up phase with the virgin sand where filtration conditions are unstable. The removal efficiency of Zn is also the significant lowest in this period compared to the 8 to 24 events ($P<0.05$). A period over the 4 to 11 events is characterized by increased loading of dissolved Cu and release during event 5. However, after event 5, total Cu was removed but with an increased concentration of dissolved Cu in the sand filter effluent.

Kandasamy *et al.* (2008) used two filter sand media for studying removal of stormwater pollutants. One finer filter sand with a d_{10} and d_{90} of 150 and 300 μm, and with a uniformity constant of <3 and a coarser filter sand d_{10} and d_{90} of 500 μm and 1,000 μm, also with a uniformity constant of <3. They found no difference between the fine and coarse sand in removal efficiency of the studied pollutants. The inlet concentration of total Zn in the

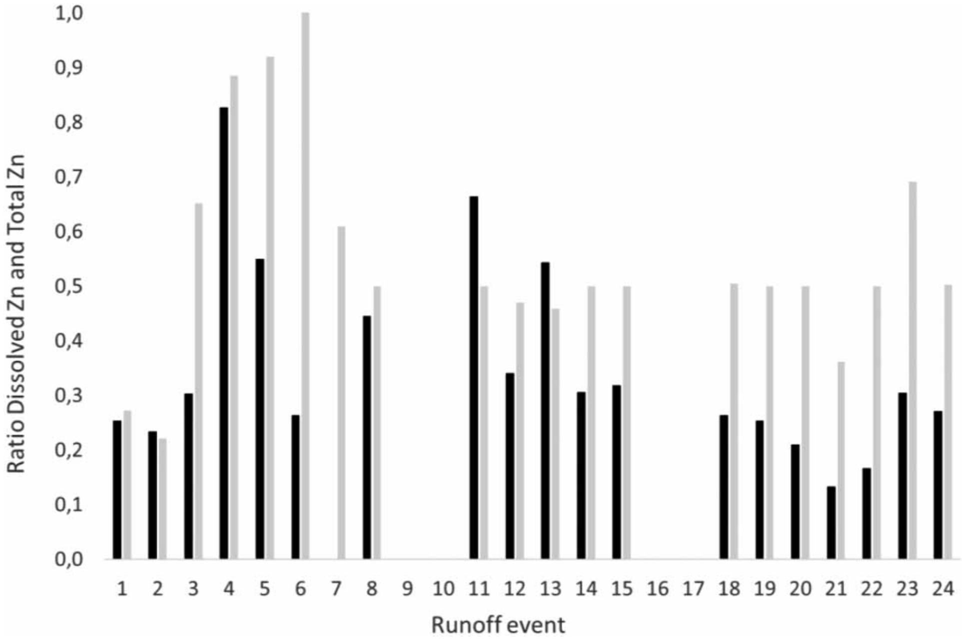


Figure 4 | Ratio for dissolved Zn and total Zn in influent and effluent. Black bars = ratio for dissolved Zn/total Zn in influent; Grey bars = ratio for dissolved Zn/total Zn in effluent filtrate. Data missing for rain events 7 (influent), 9–10 and 16–17.

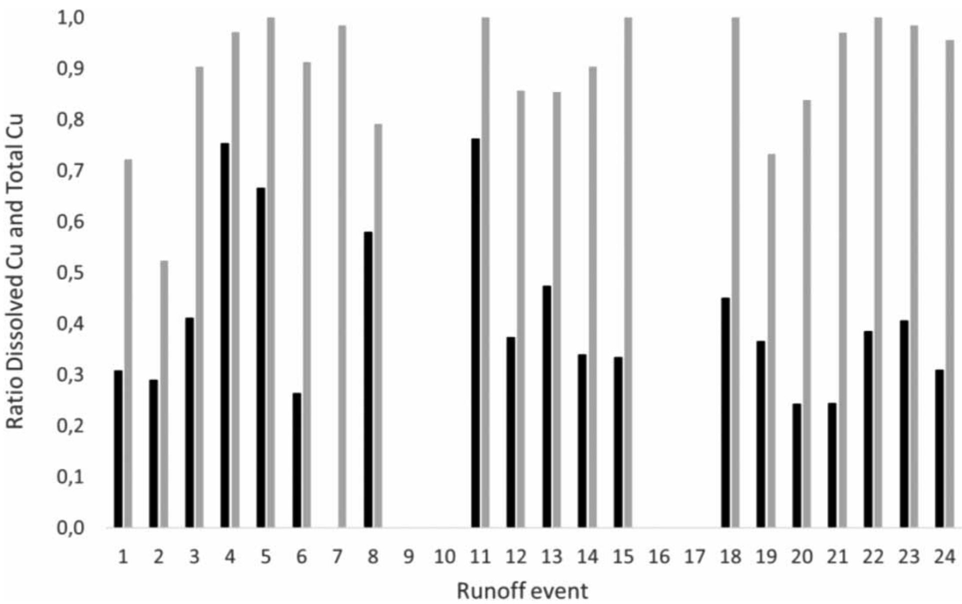


Figure 5 | Ratio for dissolved Cu and total Cu in influent and effluent. Black bars = ratio for dissolved Cu/total Cu in influent. Grey bars = ratio for Cu/total Cu in effluent filtrate. Data missing for rain events 7 (influent), 9–10 and 16–17.

Kandasamy *et al.* (2008) study was 280 μg L^{-1} and in the finer and coarse sand filter effluent, the Zn concentration was 60 μg L^{-1} and 50 μg L^{-1}, respectively. The filter sand used in our study had a particle size between the filter sands used in the Kandasamy *et al.* (2008) and Siriwardene *et al.* (2007) studies and in both cases, we found higher removal efficiency of dissolved Zn (<0.45 μm) during the studied runoff events. In general, studies of stormwater treatment had focus on slow sand filters e.g., Muhammad *et al.* (2005), Egemose (2018) and Siriwardene *et al.* (2007).

McDowell-Boyer *et al.* (1986) discussed particle straining where very small particles relative to the porous media grain size can be removed from solution by physical and chemical forces between particles and the media. They also observed the importance of organic material acting as adsorbents and forming aggregates, depending on the chemistry of the water, e.g. Cu adsorbing fulvic acids. If much smaller pollutants adsorbs to colloidal organic material, they suggested that the ratio between removed particles diameter (d_p) and sand media diameter (d_m) will exceed 1,000. The filter sand in this study had a d_{10} of 1.35 mm (Table 1), which corresponds to a minimum d_p/d_m of 3,000 in relation to material less than 0.45 μm. In the study by Siriwardene *et al.* (2007), the gravel layer in one trial reduced particles sizes down to 3 μm with a d_{10} of 6.4 mm yielding an elevated ratio of 2,100 compared to McDowell-Boyer *et al.* (1986). Bevers *et al.* (2020) sampled stormwater recipient water during runoff events and found that on average, 21% of Zn mass was present as Zn-only particles, with a rather uniform mean size of 40.2 nm. Zn that was detected with one or more other elements, primarily Al, Fe and Si, was likely to be present as hetero-agglomerates or within mineral colloids.

We performed randomly five measurements of TOC and DOC in the influent runoff before the sand filtration (Table 5). The TOC and DOC concentrations varied from 11.4 to 65.9 mg L^{-1} and 9.69 to 40.3 mg L^{-1}, respectively.

Table 5 | Analysed concentration of TOC and DOC in the influent runoff to the filter column

Event	1	4	5	6	21
TOC (mg L^{-1})	14.0	52.2	39.0	11.4	65.9
DOC (mg L^{-1})	10.9	40.3	18.3	9.69	12.8

The TOC concentrations in our study (Table 5) are elevated compared to the TOC concentration (12 mg L^{-1}) found in the study by Muhammad *et al.* (2005). They showed that an increased TOC concentration improved metal removal, while increases in flow rates decreased the removal of metals in a slow sand filter. Moreover, their slow sand filtration trials showed that adsorption via organic ligands was the predominant mechanism for metal removal at the surface of the filter, but chemical adsorption was more important deeper in the filter. Faisal *et al.* (2021) showed that plantation of humic acid nanoparticles on inert sand increased the sorption capacity of Cu. The minerogenic composition differs between different types of sand, which can affect the interaction of humic substances with the mineral surface (Mal'tseva & Yudina 2014). The implication of this relationship is that selected sand for filtration can affect the outcome of the removal of the metals.

It is suggested that the excess of organic material could contribute to the overall removal for the studied runoff events, which also in the long-term can contribute to the formation of a colmation layer. Adsorption of metals to sand and organic matter are the two likely mechanisms and the efficacy depends on filter bed depth and filtration rate (Faisal *et al.* 2021). However, the chemical adsorption is unclear in natural, un-manipulated sand, and should be further investigated by testing different commercial filter sands.

The risk for desorption of metals from full-scale RRTPs using sand as filter medium, should be considered because the use of de-icing road salt has been pointed out as a driver for metal release (Flanagan *et al.* 2018). They found the highest concentrations of pollutants during winter along a road in Paris and explained this observation as related to NaCl-rich runoff water. A batch experiment performed by Zhong *et al.* (2021) revealed that the adsorption efficiency by sand decreased as the concentrations of Cl^- and SO_4^{2-} increased. Huber *et al.* (2016a) investigated NaCl alone and mixed with $CaCl_2$ or with $MgCl_2$ and found that Cu was released the least of all metals. However, sand was not included in their experiment. In our study, the salt concentration increased in the runoff during events 21–24, exceeding 1,000 mg L^{-1}. Desorption from accumulated particles in the sand and increased mobility of particulate matter could then be possible. However, no significant increase in Zn and Cu concentrations was observed in the sand filter effluent during these events in December and January, compared to the previous months ($p > 0.05$). The de-icing salt used on the E4 road in Stockholm consisted of NaCl mixed with little addition of $CaCl_2$.

Aspects for full-scale application using sand as filter medium

The findings of this field study show that the filter sand can remove Zn and Cu from road runoff in a gravity fed treatment system. However, long-term studies are needed in full-scale operated RRTPs where conditions are not as controlled as in a column experiment. We observed the risk for Zn and Cu release in the beginning of the operation period but for the whole nine-month period, a good reduction of Zn and Cu was obtained. The observed release has not been reported in other studies and further investigation is needed, using real road runoff and filter loads appropriate to water volumes to be treated. The fractionation of metals play an important role in the performance of a stormwater treatment system (e.g., Maniquiz-Redillas & Kim 2014; Maniquiz-Redillas *et al.* 2019). Hence, applying low-impact development practices to remove dissolved metal species (Maniquiz-Redillas & Kim 2016) where sand is used, requires accuracy in the systems design and their management.

The Swedish city Göteborg has guidelines for stormwater (Göteborgs Stad 2021), which recommend a general effluent value for Zn and Cu of 60 μg L^{-1} and 22 μg L^{-1}, respectively, thus this type of sand used in our study could be an alternative to meet the requirements for these two metals. However, the stringent guideline values applied in the Swedish municipality Järfälla (2016) (7.5 and 4.5 μg L^{-1} for Zn and Cu, respectively) discredits the filter sand based on the findings from this study. As described by other studies, the removal of particles is likely to occur in the top layers, thus scraping and replacing the top layer (10–15 cm) could be a feasible method for extending the operation time over several years with this type of sand filter. Moreover, we suggest that sand can be used as a cover layer on reactive filter materials such as those studied by Huber *et al.* (2016b), which are designed for pollutant control of road runoff. Since backwashing is excluded as a cleaning process in full-scale RRTPs, scraping and backfilling of new sand would be a feasible method to extend the performance and lifetime of downflow reactive filter systems (cf. de Souza *et al.* 2021).

CONCLUSIONS

We reported results from a pilot-scale plant treating polluted runoff from the busiest motorway in Sweden. The trial was performed on-site, housed in a full-scale treatment plant using sedimentation and flocculation as primary treatment steps. A commercial filter sand was tested during nine months, covering 24 runoff events. The water flow to the full-scale plant varied from 20 to 145 $m^3 h^{-1}$ of which the proportional surface loading to the sand filter ranged from 16.8 to 201 L $m^{-2} h^{-1}$. The following conclusions were drawn from this study:

- The sand filter was able to efficiently remove Zn and Cu total concentrations (93 and 67%, respectively) after only grit chamber sedimentation.
- Dissolved Zn was efficiently removed by the sand to 87% while Cu showed only 19% removal.
- Head loss and clogging were not observed during the study period; however, particle accumulation in the colmation layer was indirectly assumed due to the high removal of Zn, which is known for its particle-bound affinity.
- Future research will include detailed studies of the top sand layer and mechanisms behind the metal removal with regard to the accumulation of organic matter. Other types of sand will moreover be tested in batch and column experiments as their different geological origin and chemical composition can determine their metal sorption capacity.

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DATA AVAILABILITY STATEMENT

Data cannot be made publicly available; readers should contact the corresponding author for details.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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