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# MICROPROOF
## Micropollutants in Road RunOff

# Efficiency of treatment systems
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## Efficiency of treatment systems

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# 1 Introduction

This report presents measured efficiencies of road runoff management systems with respect to specific organic micropollutants (OMP) and microplastic (MP). However, for many OMP there is precious little of such information, and for microplastic the knowledge is even scarcer. For a selection of pollutants where no or insufficient data is available, the effect of the implemented unit operations will be assessed based on theoretical understanding of pollutant properties. The expected effect of treatment technologies will then be extrapolated here from.

The report addresses OMP and MP, and does not target the more conventional pollutants such as suspended solids, heavy metals, and nutrients. It furthermore solely uses data from existing systems, and does not discuss the detailed sizing of solutions to obtain better treatment. For such the reader is referred to the specialized literature on this topic, such as national or regional manuals and guidelines, text books and reports (e.g. Hvitved-Jacobsen et al., 2010; Pazwash, 2011; Torres et al., 2015, etcetera).

# 2 Measured treatment efficiencies of road runoff management systems

Most data on treatment systems for stormwater runoff cover a combination of urban, road and highway drainage areas. However, urban and highway runoff does not differ in terms of hydrology and flow characteristics. The overall chemical composition of the runoff is also quite similar with respect to most of the pollutants of interest for road runoff. Consequently, the implemented technologies are similar and often even identical when it comes to treating urban versus highway runoff. Experience on urban runoff treatment systems can hence generally be extended to also cover highway systems.

The pollutants that have received the most attention for runoff treatment systems are suspended solids, heavy metals and nutrients (Clary and Jones, 2017). In a number of cases, PAH (polycyclic aromatic hydrocarbons) has also been studied due to a high toxicity of some of the individual PAHs. However, stating the treatment efficiency of a certain category of treatment facilities, e.g. retention ponds, is not straight forward even for substances that have been studied in great detail. Treatment systems differ on a wide range of parameters such as their specific treatment volume (the ratio of impervious catchment area to permanently wet treatment volume), detention volume and time, size, geometry, geographic location, and they differ in terms of the land use of the connected drainage area and hence in terms of the pollutant loadings. The sampling approaches applied to obtain the data also differ, as do sometimes also the analytical methods applied for analysing the samples. All in all, asking for example the question “how well does a retention pond treat?” is equivalent to asking “how expensive is a car?”. An example of this issue is shown in Figure **1** where the relation between specific treatment volume and treatment efficiency (left hand figure), and treatment volume and outlet concentrations (right hand figure) is shown for a number of retention ponds (data after Vollertsen et al. (2012), based mainly on the USA and Canadian database www.bmpdatabase.com, and extended with some Danish and Norwegian data).

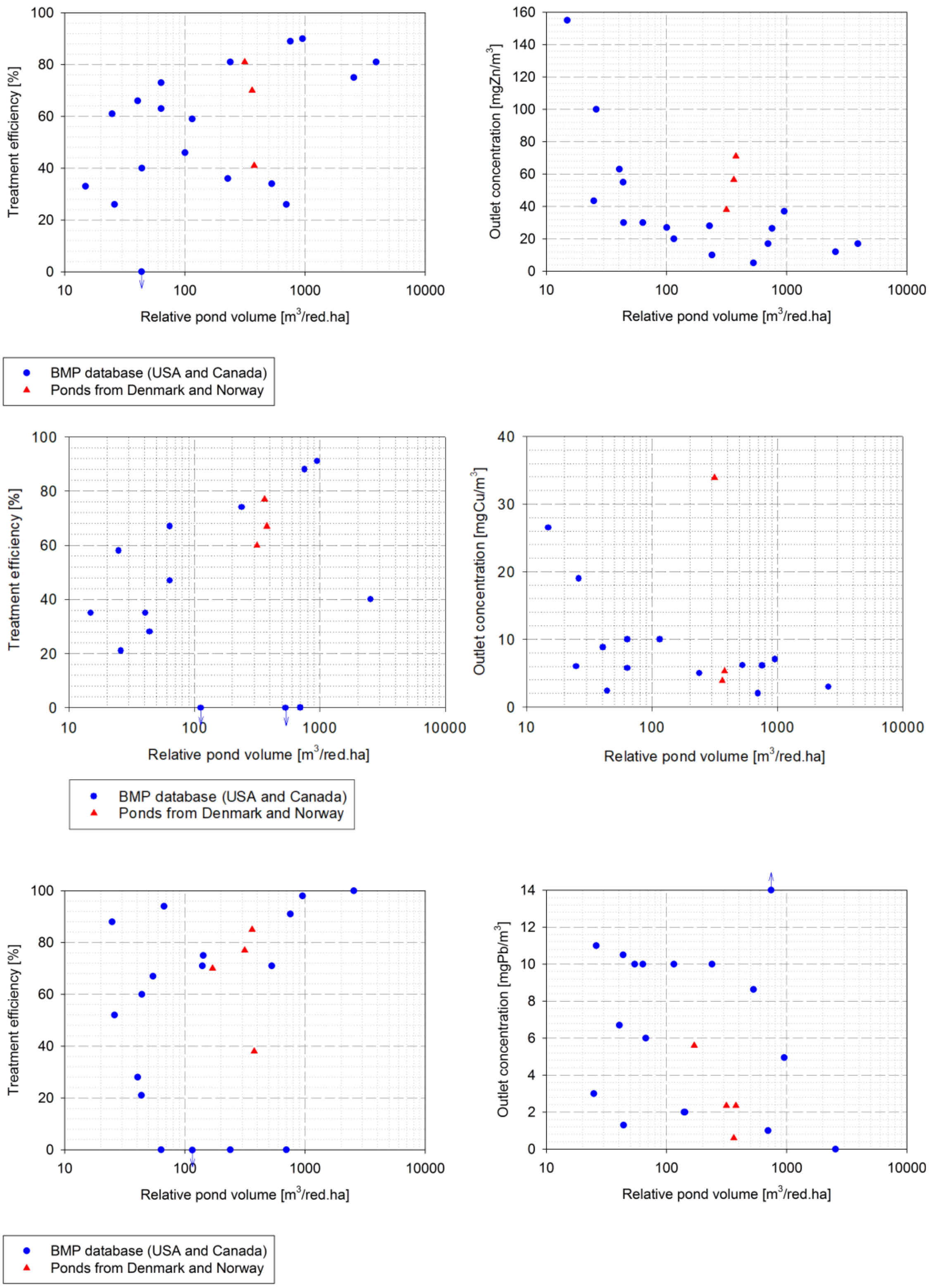

**Figure 1** **Treatment efficiency and outlet concentrations for zinc, copper and lead of stormwater ponds serving a mix of urban and highway runoff. The blue points are from the BMP database data ([www.bmpdatabase.org](http://www.bmpdatabase.org)) and cover retention ponds up till 2012. The figure is after Vollertsen et al. (2012)**

Another issue which must be considered when addressing the efficiency of a treatment solution is that such efficiency is often stated as “percent of the pollutant load removed by the facility”. This approach is inherently problematic for facilities that do not receive similar loads. Generally speaking, it is easier to treat heavily polluted water by a certain percentage, compared to reducing the pollutant loads associated with less contaminated waters. In other words, even though treatment systems are often assumed to follow some simple pollution removal function, like a first order removal process, this is in reality only a rough first approximation of how they work in reality. This is illustrated by real data for retention ponds from the American bmp database including some Danish and Norwegian ones (Figure 2), where it is seen that treatment efficiency depends strongly on the inlet concentrations of four of the five pollutants addressed.

Interpreting reported treatment data can be challenging because sampling methods often have been poorly documented, especially when it comes to an evaluation of how representative the obtained samples actually are for the inflow to the treatment facility. The issue here is amongst others that it is inherently difficult to collect representative samples of stormwater runoff entering a treatment system. Sampling the outlet is somewhat easier, but still not a simple task. Sampling is also commonly done by different approaches. Some studies have applied grab samples taken during a rain event. Other studies have used automated samplers triggered by flow or water level. Some of the studies have analysed single events, comparing inflow and outflow during a runoff event. While this for some systems might be appropriate, it is not so for others. For example when sampling retention ponds, this way has little meaning for quantifying treatment efficiencies, as the outflow is not representative of how well that specific inflow was treated (due to mixing and time delays, and because treatment in such ponds mainly occurs during the inter-event dry periods).

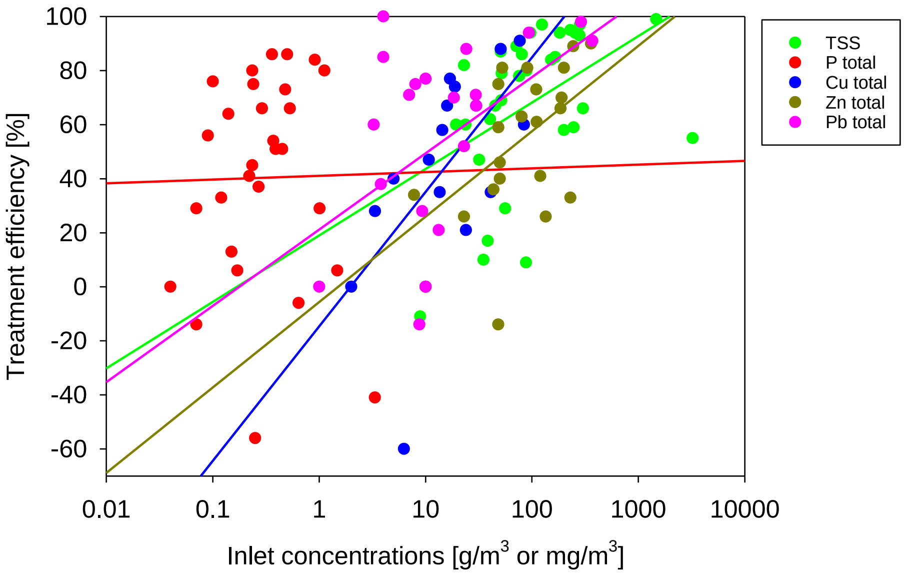

**Figure 2 Stormwater inlet concentration versus treatment efficiency of wet retention ponds using data from the BMP database data ([www.bmpdatabase.org](http://www.bmpdatabase.org)). The figure covers retention ponds up till 2012 and is after Vollertsen et al. (2012)**

Overall, it is consequently difficult to state exactly how efficient a certain treatment technology is to remove a certain pollutant – even when there is abundant data on the substance in question. For organic micropollutants and microplastics, the issue of lack of relevant data further complicates drawing conclusions on the efficiencies of specific treatment systems. The only way around this conundrum when assessing the treatment efficiency towards a substance which has not been studied, is to assume that such substances behaves somewhat similar to a substance where data do exist. For example to assume that the removal of microplastics is similar to the removal of TSS because both substances are particulates.

## 2.1 Retention ponds and wetlands

Stormwater retention ponds and wetlands are probably the best described technology for managing stormwater runoff. Their efficiency in retaining a wide range of particulate pollutants is well-documented, and some data also exists showing that they can reduce part of the dissolved pollutants.

Sébastian et al. (2014) investigated a stormwater retention pond for a number of heavy metals, PAH, and pesticides. They reported that event based removal rates for PAHs (sum of 15 individual PAHs, namely Acy, Ace, Flu, Phe, A, Flh, Pyr, BaA, Chr, BbF, BkF, BaP, IP, D(a,h)A, Bper) were between 67% and 24%. The efficiency towards other OMP was lower, for example did removal rates for alkylphenols vary between 2% and 31% with a median value of 14% for 4-nonylphenol. Polybrominated diphenyl ethers (PBDEs) were measured in one event, showing retentions ranging from 20% to 60% depending on the compound. The pesticides in the stormwaters seemed not to be trapped in the pond. However, when interpreting these data, the previously mentioned caveat for applying a sampling scheme where inlet and outlet of a retention pond is sampled for the same event must be called to mind. Such sampling strategy will not result in correct estimates of treatment efficiencies, as it is not the same water volume which becomes sampled. What can, however, be used are the outlet concentrations from these ponds. For PAH (sum of 15) the outlet concentrations were in the range of 165-600 ng/L. For 4-tert-octylphenol it was around 40 ng/L, 4-nonylphenol ranged from 122 to 1332 ng/L, diuron from 3 to 1400 ng/L, and isoproturon from 0.8 to 65 ng/L. However, care must be taken when interpreting these data for assessment of what is discharged from a retention pond as these concentrations simply might reflect previous events inlet concentrations.

Vollertsen et al. (2009) reports data on a highway pond in Norway and found that treatment efficiencies for total PAHs (USEPA 16 PAH) was 85% while it was 89% for the sum of 4 selected priority PAHs. In that study, the treatment efficiency was calculated as an average over 1 year of continuous, flow proportional sampling. For comparison, the treatment efficiency for PAHs was slightly higher than that for suspended solids. The median of the outlet concentrations were only slightly above the limits of quantification (100 and 10 ng/L, respectively). They also report that two large snowmelt events impacted negatively on the overall removal rates.

Walaszek et al. (2018a) reported findings from an urban constructed wetland in France with 44% of low-traffic roads. The wetland was built as a sedimentation pond followed by a vertical subsurface flow constructed wetland. The water was sampled taking the hydraulic residence time into account, hereby allowing a reasonable estimation of actual removal efficiencies. PAH (USEPA 16 PAHs) were measured. The efficiency of the pond depended on the actual PAH, but was most often close to 100%. The filter was able to reduce the PAHs further, leading to outlet concentrations that nearly always was below the limit of quantification for the individual PAHs (10 ng/L).

Maillard and Imfeld (2014) addressed the mass balance of a stormwater wetland with respect to pesticides. The wetland treated runoff from a vineyard where various pesticides were used. They analyzed for glyphosate, AMPA, dithiocarbamates, kresoxim-methy, pyrimethanil, metalaxyl, tetraconazole, difenoconazole, fludioxonil, spiroxamine, cyprodinil, and cyazofamid. They found that the pesticides were nearly all dissolved, but underwent sorption to sediments and also degradation. Plants enhanced the degradation during the growth season, but were found to release some during plant senescence. The retention on a weekly basis of dissolved pesticides was on average 96% while particle bound pesticides were retained by 98%. Variations were though substantial.

Ivanovsky et al. (2018) investigated a lake receiving a mix of stormwater and raw urban wastewater. In this context they addressed PAHs, PCBs, caffeine and carbamazepine. They found that carbamazepine (log $K_{OW}$ of 2.45) was reduced by 28% while caffeine (log $K_{OW}$ of -0.07) was reduced by 74%. This difference in removal was explained by degradation processes in the lake. The reduction of PAH in the lake was 64% and also the PCBs were reduced. The observed removal rates were though biased by the lake having several inputs and types of inputs, and the rates shall hence not be seen as true removal efficiencies of the system.

A new study on the efficiency of stormwater ponds to retain microplastics has yielded a very first and rough estimate on microplastics retainment efficiencies. Excluding car tire rubber, Olesen et al. (in press) showed that roughly 85% of microplastics could be retained. In other words, the retainment of microplastics was of the same order as the general treatment efficiency for particulate matter in retention ponds. It seems likely that tire particles and tire wear and road particles will be retained to similar degrees.

## 2.2 Filtration and infiltration technologies

Generally speaking, filtration and infiltration technologies can achieve higher pollutant removal than wet ponds and wetlands. The reason is partly that also rather small particles can be filtered out of the flow. For example, Egemose (2018) investigated the accumulation of particles in a sand filter polishing the outlet from at retention pond. The study found that fine particles and also pollutants like phosphorous were trapped in the upper few centimetres of the filter. Another reason is that sorption mechanisms work towards retaining dissolved substances. For example did Walaszek et al. (2018a) address sorption of dissolved metals in a constructed wetland in France (a soil filter) and performed batch experiments. They saw that zinc, copper and lead was substantially reduced by such filter. They also addressed that the pH of the water is important as metals tend to sorb better at higher pH, and that mineral composition of the filter material also plays an important role.

Fronczyk (2017) addressed treatment of artificial road runoff in a soil filter and found removal efficiencies for most heavy metals and PAH close to 100%. Hatt et al. (2009) investigated the pollutant removal in three stormwater biofiltration systems (soil filters) and found that metals and TSS was reliably removed, for example with median removal of zinc from 84 to 99%. Shrestha et al. (2018) also studied bioretention systems (soil filters) and found removal of TSS to be in the range of 89-96%. While none of these studies addressed organic micropollutants, the retainment of TSS and particle bound pollutants does indicate that such systems are efficient towards pollutants associated with particles.

Data from Switzerland on the monitoring of highway runoff filtration give a similar picture, namely that these systems are highly efficient in retaining pollutants. For example did a sand filter of 0.7 m thickness reduce zinc to below 10 µg/L and copper to around 10 µg/L (ilu AG, 2016a). Walaszek et al. (2018b) investigated a system for treatment of stormwater from a small residential catchment in France which comprised a retention pond and a constructed soil filter. They found rather good removal efficiencies in the combined system, albeit incoming water tended to re-suspend particles already settled in the pond. The overall removal was generally high, ranging from 50% for naphthalene to 100% for particulate zinc. Vollertsen et al. (2018) investigated two systems for highway runoff treatment, also comprising of a retention pond combined with a filter. The filters contained artificial soil and sorption materials (limestone and olivine granulates). The systems were able to remove the measured parameters (heavy metals and phosphorous) to levels corresponding to the background in the receiving waters. E.g. was zinc reduced to approx. 2 µg/L. Two planted soil filters in Switzerland treating highway runoff (ilu AG, 2016b) showed similarly that zinc and copper was reduced to or below 10 µg/L during a 3-year monitoring campaign. Misteli (2017) reports the efficiency of 4 other highway runoff filtration systems in Switzerland, where one consisted of a vegetated soil under which there was a layer of gravel/split. The other systems contained a mix of different filter and sorption materials. The system with gravel/split achieved 65% and 77% efficiency for copper and zinc, respectively. The other 3 systems achieved higher efficiencies. For copper and zinc: 71% and 84%; 90% and 93% and 86% and 93%. Steiner and Gosse (2009) reported data on pollutant retention for two highway runoff filtration systems, one with a zeolite filter material and one with an iron hydroxide filter material. Before the water reached the filters, it was pre-treated through at lamella separator. The iron hydroxide filter generally brought copper and zinc below 10 µg/L and total PAH down to 0.5 µg/L. For copper the treatment efficiency corresponded to approx. 80% and for zinc to approx. 96%. The zeolite filter was also efficient, albeit a bit less so than the iron hydroxide filter. Again, these data indicate that these types of systems are efficient at retaining pollutants in general, and it seems reasonable to assume that they also retain organic micropollutants and microplastics – even though little or no data is available to document this.

Pazeller et al. (2017) summarized extensive experimental data from 12 highway runoff (natural) soil filters and 8 highway runoff sand filters. They concluded that the sand filters were significantly more efficient than the soil filters, and explained that this counter-intuitive result is due to soil filters developing preferential flow paths, while this does not occur in sand filters. Hence, even though the soil filters in principle should be able to retain finer particles than the sand filters, and also sorb dissolved substances to for example clay minerals, this effect was overshadowed by the issue of part of the water flowing through the filter via differential flow paths.

Runoff infiltration through porous pavements can also reduce the pollutant content. For example did Berbee et al. (1996) find that runoff from pervious asphalt contained significantly lower levels of copper, zinc, lead, PAH, mineral oils, and organic matter than did runoff from impervious asphalt. Holmes et al. (2017) conduct lab tests on the sorption efficiency of porous concrete to retain heavy metals and found that with the “right” concrete mix, the sorption could be very efficient and of high capacity. Similar results were reported by for example Haselbach et al. (2014) who in lab experiments found zinc and copper was retainment at around 90%. Liu and Borst (2018) compared amongst others porous concrete and porous asphalt and found that the two pavement types had similar efficiencies in retaining metals. Again this leads to an expectation that porous pavements would also be effective towards a range of organic micropollutants and microplastics.

Flanagan et al. (2019) investigated the retention of dissolved and particulate pollutants in two vegetated biofilters treating road runoff from a highway in France. One system was a vegetated filter strip and the other a biofiltration swale. They analysed for trace metals, polycyclic aromatic hydrocarbons, bisphenolA, alkylphenols and phthalates. In general, the efficiency of the biofilters towards particulate bound pollutants was quite good. For example was benzo(a)pyrene – a substance with a rather high log $K_{OW}$ value (6.13) – quite efficiently retained from on average 200 ng/L down to around 25 ng/L. However, during some winter runoff events the outlet of the filters held quite high particle concentrations which was attributed to poor filtration performance during these events. Dissolved pollutants were generally less well retained by the biofilters.

Schmitt et al. (2015) investigated a combination of wet retention pond followed by a vertical-flow constructed wetland. The catchment of the system was a suburban area. The treatment train was quite effective in terms of retaining particulates and metals. For organic micropollutants, however, the documentation of the efficiency of the system was limited by the fact that the addressed PAHs generally were below detection limits in the outlet, and often also so in the inlet.

Fairbairn et al. (2018) investigated a wide range of organic micropollutants in a drainage area in Minnesota, USA and investigated their reduction in iron enriched sand filters. They concluded that those filters effectively reduced the concentration of 14 out of the 48 most detected substances of concern with median removal efficiencies of 26-100%. Especially the hydrophobic (high log $K_{OW}$ value) compounds like PAHs and bisphenol A, and polar-hydrophilic substances like caffeine and nicotine were removed efficiently, indicating that particle retention was important. For some dissolved substances sorption also caused efficient retention.

## *2.3 Technical solutions for stormwater management*

When simple solutions like infiltration systems or retention ponds cannot be established, and the receiving waters require treating the runoff before discharge, more technical solutions can be chosen, for example based on flocculation and/or filtration. While such systems tend to be more costly in terms of investment, operation and maintenance than retention systems, filtration systems and infiltration systems, they can be the only viable solution when space is limited or other conditions require such installations. Properly designed and maintained they can achieve good efficiencies. For example did Bundesamt für Strassen (ASTRA) (2014) report the efficiency of a technical filter for road runoff treatment in Switzerland, applying a sedimentation basin followed by a rotating disk filter. This setup achieved retention for copper and zinc of 66% and 59%, respectively, i.e. in corresponding to the lower range of what can be expected for a retention basin. Bundesamt für Strassen (ASTRA) (2017) reported the efficiency of another system of similar concept which achieved 62% and 59% for copper and zinc, respectively. The achieved mean discharge concentrations were 23 and 140 µg/L, which also is in the range of concentrations commonly achievable by retention basins (Hvitved-Jacobsen et al., 2010). Also other systems like ballasted sedimentation (for example the Actiflo® Process marketed by Veolia) can achieve high removal efficiencies, comparable to or even better than what retention basins can yield.

# 3 Predicting the treatment performance for pollutants

The only organic micropollutant for which there is some data on treatment efficiencies is PAH. Other data are scarce and either not measured at all or to a degree where no general conclusion can be drawn. The same is the case for microplastics, where no data what so ever exists on the efficiency of stormwater treatment systems. Several studies have addressed this issue by establishing process-based models. Zhang et al. (2016) proposed such model for stormwater biofilters and validated it up against measured data from two field biofilters. They combined a pollutant transport and transformation model with a hydrodynamic model including sorption to the filter media and biodegradation. They were able to calibrate the model and validate it against a sand filter media with a submerged zone, but not against a loamy sand without submerged zone. Nevertheless, such detailed tools seem promising when predicting the behaviour of a specific pollutant in a specific system holding a specific filter material.

Vezzaro et al. (2014) attempted modelling of stormwater treatment on a city scale. They also build a model including relevant processes like sorption, filtration, sedimentation, resuspension, and biological degradation covering the various stormwater treatment systems of city. They then applied it on a theoretical catchment and predicted concentrations of various organic micropollutants. The model was, however, not validated on real systems.

For stormwater ponds, Vollertsen et al. (2007; 2009; 2012) suggested to use a simple first order pollutant removal process in a water body simplified as a completely mixed reactor. The system was fed by long measured rain series and simulated the relevant hydrodynamic processes in the pond. The model was calibrated up against one year of measurements at a Norwegian stormwater pond treating highway runoff. A model based on this concept and this calibration is today used in Denmark to predict the efficiency of various stormwater treatment pond configurations (WDP, 2019).

While these attempts to model the treatment performance all have their uses, they fall short when it comes to predicting the behaviour of a novel pollutant. For example microplastics. While it is quite obvious that systems which are good at retaining organic particles are likely also good at retaining microplastics (because the density and properties of the two particle types have similarities), such conclusion is only valid in qualitative terms. Similarly, it is quite obvious that an unknown substance with a high log $K_{OW}$ value will be retained in systems that retain other substances with high log $K_{OW}$ values, as the sorption processes leading to the removal are similar. However, this is again a statement that can only be made in qualitative terms.

A simple qualitative overview of the fate of organic micropollutants that have not yet been measured is given in Table 1-4. For example, if a substance is easily degradable and sorbs easily to solids, it has a high probability of being retained/removed in all types of treatment systems. If it, on the other hand, is slowly degradable and sorbs poorly to solids, it most likely will not be much affected by any system, and the least affected by systems with a short hydraulic residence time. Comparing the approaches laid out below, one can in very general terms furthermore say that systems based on slow soil filtration will tend to achieve the higher removal rates. However, this is only true as a general trend, and the actual efficiencies will depend on the actual layout and design of the systems.

**Table 1 A qualitative assessment of the fate of substances in a stormwater management facility applying a wet retention volume**

| | Particulate | Sorbs well to solids | Sorbs poorly to solids |
|---|---|---|---|
| Easily degradable | 🟩 | 🟩 | 🟨 |
| Slowly degradable or inert | 🟩 | 🟩 | 🟥 |

**Table 2 A qualitative assessment of the fate of substances in a stormwater management facility applying (slow) soil filtration**

| | Particulate | Sorbs well to solids | Sorbs poorly to solids |
|---|---|---|---|
| Easily degradable | 🟩 | 🟩 | 🟨 |
| Slowly degradable or inert | 🟩 | 🟩 | 🟥 |

**Table 3 A qualitative assessment of the fate of substances in a stormwater management facility applying (rapid) soil filtration**

| | Particulate | Sorbs well to solids | Sorbs poorly to solids |
|---|---|---|---|
| Easily degradable | 🟩 | 🟩 | 🟥 |
| Slowly degradable or inert | 🟩 | 🟩 | 🟥 |

**Table 4 A qualitative assessment of the fate of substances in a stormwater management facility applying technical (rapid) filtration or ballasted sedimentation**

| | Particulate | Sorbs well to solids | Sorbs poorly to solids |
|---|---|---|---|
| Easily degradable | 🟩 | 🟩 | 🟥 |
| Slowly degradable or inert | 🟩 | 🟩 | 🟥 |

An example of degradation of an unknown substance could be caffeine (from coffee, tea, soft drinks). It falls in the category of substances that sorb poorly, and it will hence not be retained in a rapid soil filter, a technical filter, or other solution with short hydraulic residence time. It will, on the other hand, be degraded in a stormwater retention pond as it is easily degradable. On the other hand, it will be removed to a lesser degree than a substance that sorbs well but is similar degradable.

Another example of an unknown substance could be car tyre debris and tyre wear and road particles, on which there exists no measurement data what so ever. These are particles which probably behave similar to other organic particles of similar size, and hence likely are removed by processes and to degrees similar to these. Further indication of this is the observation that microplastics in general seem to be retained similar to particulates (Olesen et al., in press), and it seems likely that tyre particles would behave similar.

Summing up, our knowledge on the efficiency of specific treatment solutions towards the wide range of organic micropollutants and microplastics that can be found in road and highway runoff is very limited. The state of knowledge does not allow a detailed assessment of treatment efficiencies and discharge concentrations, but only a qualitative assessment based on treatment system and pollutant characteristics. Only a substantial number of intensive and systematic monitoring campaigns targeted on the substances in question can resolve this issue.

# 4 Acknowledgement

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