# MICROPROOF
## Micropollutants in Road RunOff

# Processes and unit operations for road runoff management

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# CEDR Call 2016: Environmentally Sustainable Roads: Surface- and Groundwater Quality MICROPROOF Micropollutants in Road RunOff

# Processes and unit operations for road runoff management

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# 1   Background

A variety of measures and treatment systems are presently applied to treat road runoff with the purpose of reducing the possible risk of pollutants conveyed herein. Measures like filtration (infiltration into road shoulders, banks, and ditches; filter units discharging to surface waters) or sedimentation (retention ponds or artificial wetlands) are used to reduce the amount of pollutants that will reach a water body. The efficiency of these treatment systems depends not only on the treatment system characteristics but also on a wide range of pollutant characteristics.

With respect to organic micropollutants (OMP), it is important whether the substance is entrapped in a particle or free to sorb and desorb from solid phases (whether it is dissolvable). In this context an important aspect is its ability to sorb to organic materials and inorganic minerals. These parameters determine whether filtration and sedimentation can remove the substance in question. A further aspect that is required to assess the efficiency of treatment systems to retain OMP, is the degree to which these can be degraded, being it biologically or chemically.

Microplastics (MP), on the other hand, behave somewhat differently as they always are particulate. Filtration will hence be an efficient way to retain MP. In an aquatic system like a pond or a wetland, some in principle will sink while other in principle will float as they have a density ranging from approx. 0.8 to 2.2 g/cm$^3$. MP may also be subject to coagulation, sorption to surfaces, biofilm growth, and predation with subsequent excretion of the MP as part of the faecal pellet.

# 2   Processes and unit operations that retain OMPs and microplastics

## 2.1   Particles and their trapping

Particles are important vectors for OMP, and MP are themselves particles. Hence the process which in general leads to particles being retained do also lead to OMP and MP being retained. With respect to OMP, it is furthermore important to which degree they are associated with particles.

### 2.1.1   Particulate pollutants and pollutants associated to particles

Particulate pollutants like MP, OMP entrapped in particles, OMP sorbed to suspended particles, and OMP precipitated by technical or natural processes, are generally removed from the water phase by becoming part of the system's sediments or by filtration. Particulates range from colloidal particles (colloids) to macroscopic particles. Colloids are so small that their movement in a liquid is completely governed by thermal energy. They consist of a number of associated molecules and are significantly larger than most single molecules. Particles are termed colloids as long as they stay suspended in a liquid and do not settle out, even though their density differs from the density of the liquid. There is no clear boundary between what is a dissolved substance, a colloid, and a macroscopic particle. As the particle size increases, particle movement becomes governed by gravitational forces and the particles settle or float in a liquid. In terms of treatment, the settling behaviour of particles is

of major interest as much of the pollutants in road runoff are either particulate or associated with particles.

Fine particles and colloids can coagulate due to attractive forces e.g. van der Waals attraction forces. These forces result from the non-permanent induced dipoles within a colloidal particle or molecule. Various external conditions affect the stability of the coagulated particles, especially the water's pH, ionic strength, temperature, and turbulence. Generally, colloids in road runoff are negatively charged (Hvitved-Jacobsen et al., 2010) and coagulation hence requires an initial step where a positively charged coagulant, for example naturally present iron, aluminium or natural polymers, initiate the formation of larger particles. Upon coagulation, the formed particles tend to flocculate – a process affected by the turbulence of the liquid and its viscosity. In summary, coagulation and flocculation can allow colloids to come together so that the agglomerated material ultimately can settle in the liquid or be filtered out by a porous media.

## 2.1.2 Precipitation

Truly dissolved substances might be directly precipitated as compounds of low water solubility. Or they might be precipitated by complex formation with compounds of low water solubility. The latter is probably the most common phenomenon. An example is iron(III)hydroxide (ferric hydroxide), which also can lead to complex formation with various ligands, including some of the dissolved substances. Formation of metal complexes is for example a mechanism for getting heavy metals out of solution. The degree of complexation and hence removal of dissolved substances will depend strongly on the speciation of the metal. Detailed information on this can for example be gained from speciation and phase diagrams that are based on thermodynamic data or from computer programs which predict speciation. Speciation of metals depend strongly on pH and redox potential. For example, iron(III)-complexes often bind a wide range of substances. When conditions favour reduction of iron(III) to iron(II), previously bound substances may be released back into solution. This phenomenon is, for example, seen during eutrophication-induced oxygen depletion where anaerobic sediments release previously bound metals and phosphorous (e.g. Vink et al., 2017).

The precipitation-products are colloids and will behave as such, meaning they can coagulate and flocculate and ultimately settle out of the water column. They will do so together with naturally occurring colloids. Addition of, for example, iron salts will hence lead to removal of not only substances that react directly with the iron but also of substances associated with other colloids, which then are removed by coagulation/flocculation.

## 2.1.3 Sorption

A number of substances exhibit partitioning between solid and liquid phases. Partitioning between phases is a complex phenomenon, depending on the substance itself (the sorbate) and the solid phase to which it is sorbed (the sorbent). It does, though, also depend on environmental conditions such as pH, ionic strength, and competing sorbates. For stormwater pollutants, the most important sorbents belong to the groups of organic particles and clay minerals (Arias-Estevez et al., 2008). Technical sorption materials such as limestone, zeolite, activated carbon, olivine, and so on are also effective to sorb a wide range of substances.

A vast number of studies have addressed the sorption of various pollutants to a diversity of matrices under a range of conditions, see for example Delle Site (2002). For many practical applications it has been assumed that there is a correlation between a substance's hydrophobicity measured by its $K_{OW}$ value (octanol-water partitioning coefficient) and its ability to sorb in natural systems such as stormwater treatment facilities. This assumption gives, however, only an indication of the relative degree of magnitude of sorption and not an exact value (e.g. Bollmann et al., 2015; Styszko et al., 2014).

Sorption can be divided into two phenomena, namely adsorption and absorption. Adsorption is the adherence of substances onto the surface of a particle. The interactions between a substance in solution and a solid surface are governed by forces across the liquid–solid interface. The substance partitions according to the intermolecular forces, creating either electron-pair bonds or permanent and mutually induced dipoles, binding the substance to the solid. Details on sorption mechanisms can be found in many textbooks on physicochemistry, for example Atkins and de Paula (2014).

Absorption covers the phenomenon where a substance penetrates into a different phase such as a liquid or solid – contrary to adsorption where it is bound to its surface. One can pragmatically view absorption as an uptake of a substance into a different phase, e.g. from the water phase into a solid phase (dissolved substances can to some degree move in a solid phase, for example a polymeric phase like plastics).

Adsorption is an equilibrium process with a constant ratio between the concentration adsorbed on the solid ($C_{solid}$) and the concentration in the liquid ($C_{water}$):

$$K_{s,w} = \frac{C_{solid}}{C_{liquid}}$$

$K_{s,w}$, also often called $K_d$, is termed the partitioning coefficient. Besides depending on the substance and the solid, its actual value will depend on conditions in the water phase like $C_{liquid}$, pH, ionic strength and redox conditions, and on the surface area of the solid phase. For organic substances, the $K_{ow}$ of the substance (its octanol-water partitioning coefficient) is often used as a rough estimate on how well it will sorb to an organic matrix. The higher the value, the better the sorption.

Often sorption is described by sorption isotherms linking water phase and solid phase concentrations. The most common isotherms models applied to describe sorption equilibrium are the Freundlich isotherm, the Langmuir isotherm and the linear isotherm.
Adsorption is not always an instantaneous process and may require minutes to hours to run to completion (e.g. Wium-Andersen et al., 2012). Adsorption kinetics relevant for a process in road runoff treatment are typically well-described by 2nd order rate expressions including both the sorbent and the sorbate.

### 2.1.4 Sedimentation

Sedimentation is commonly described using Stokes law or modifications hereof. While Stokes law for the settling of particles is not physically wrong, it does have severe limitations in terms of fully describing processes within a stormwater management facility. Its assumptions – namely that particles must be spherical and rigid, the viscosity of the fluid is the predominant limitation on acceleration, there is no interaction with other particles or substances in the water, and that the water is not in motion – are in practice seldom valid. In

practice Stokes law tends to only give reasonable estimates for inorganic particles (sand) in the range of probably some 40 – 100 μm and hence only targets a very limited part of the relevant size and density range (Hvitved-Jacobsen et al., 2010). Other theories do exist for the settling of particles within turbulent conditions. However, due to the wide diversity of particle size, shape, and density, the coagulation, flocculation and stability of aggregated particles, as well as the complex macroscopic hydraulics, these models also are challenged when it comes to real systems. In addition hereto, the exact boundary conditions in terms of particle sizes, particle shapes, particle density, inflow patterns, wind induced mixing, and so on, are unknown at the detailed level (Andradottir, 2017). Hence with the need to describe sedimentation using empirical expressions emerges. This typically boils down to surface loadings or volumetric loadings combined with guidelines on how to design the specifics of a certain sedimentation system, for example a retention pond.

Recent studies (unpublished) have shown that MP of density less than that of water are abundant in the sediments of stormwater ponds. How this can be is not well understood, but it seems obvious that sedimentation cannot be the mechanism causing this (as the particles would float). Some simple calculations using Stokes law illustrates this. A rather heavy particle, for example of PET (polyethylene terephthalate) of 100 μm will in 10°C water settle with 5.7 m/h and hence easily be able to settle out in a pond of, say, 1 m water depth. A smaller particle, for example of 10 μm, will only settle with 0.057 m/h. A lighter particle, say of PS (polystyrene) of 1.04 g/cm$^3$ would still be able to settle if it is 100 μm, as this particle would sink with 0.60 m/h. However, a 10 μm particle would only sink with 0.006 m/h. Particles of PP (polypropylene) or PE (polyethylene) are lighter than water and should float. Large tire particles (TP) and tire road and wear particles (TRWP) should sink as they are heavier than water.

### 2.1.5 Other mechanisms causing particles to become part of pond sediments

Recent and still unpublished studies (Liu et al., in prep; Olesen et al., in prep) indicate that particles which do not settle by gravity (for example because their density is lower than that of the water) can still be detained by urban stormwater ponds and become incorporated in their sediments. Very little or no published information is available on this. However, it is well-known from marine science that the so-called "marine snow" can lead to the settling of otherwise buoyant particles, for example buoyant microplastics (Porter et al., 2018; Summers et al., 2018; Linders et al., 2018). Another possible pathway is the uptake and excretion of microplastics. Microplastics are excreted in pellets together with natural substances (Straub et al., 2017). These pellets will have a larger size and possibly also a higher overall density than the microplastics, and can hence be a transport mechanism to convey the microplastics from the water column to the sediment phase. Which processes actually lead to the buoyant microplastics particles to be found in the sediments of water bodies like retention ponds is though unknown. The processes might have to some degree be alike to the processes that cause carbon sequestration in the oceans (Giering et al, 2017; Stukel et al, 2018; Wekerle et al., 2018; Richardson, 2019), but these processes have not yet been studied in stormwater treatment systems.

For particles with low settling velocities like some of the fine microplastics discussed above, convection will probably be a more important transport mechanism than sedimentation. The exact amount of convective transport in a stormwater pond will depend on a number of parameters related to the pond itself (its geometry, depth, wind shadow, and so on), temperature, and external conditions like wind and inflow. So did for example Madsen et al. (2007) observe that an inert tracer added to the highway pond they tested was completely mixed into the water phase within 24 hours. This leads to the hypothesis that fine particles

can be trapped on surfaces like the pond sediments while on their convection-driven random walk in the pond water column – a process that is commonly acknowledged when modelling particle transport in water bodies (e.g. Zhurbas et al., 2010).

### 2.1.6  Erosion and resuspension

Particles once deposited should stay deposited. However, this is not always the case as changes in hydraulic conditions may lead to erosion and resuspension of previously settled material. The erosive conditions might be caused by large inflows or by wind creating internal flows in the water column (Bentzen et al., 2009). Resuspension can also occur at the outlet of a facility if the local velocity causes critical sheer stresses to be exceeded. As with sedimentation, these processes are in practice difficult to describe deterministically as there are too many unknowns and unknowables. For example the degree to which the sediments are cohesive, where sediments actually deposit, plant growths protect sediments from resuspension, and so on. Also here design in practice therefore boils down to empirical guidelines.

### 2.1.7  Filtration

Filtration is an effective approach to retain particles. Its efficiency depends on the media through which it occurs as well as the characteristics of the particles contained in the water that is being filtered. Whenever liquids containing fine particles pass through a porous media, the particles contained in the liquids will clog the upper layer of the media. The layer formed is often referred to as the colmation layer (e.g. Hiscocka and Grischek, 2002; Veličković, 2005). This phenomenon is illustrated in Figure 1, where water containing fine particles are filtered through a coarser medium.

Time Zero, a new leakage is formed. Particles in the liquid start to move with the liquid flow towards the backfill material surface. The liquid still exfiltrates unhindered.
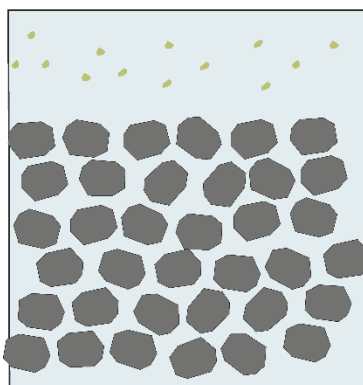
A few minutes or hours after a new leak is formed, a bridging layer develops on top of the backfill material. The exfiltration rate is governed by the permeability of this layer.
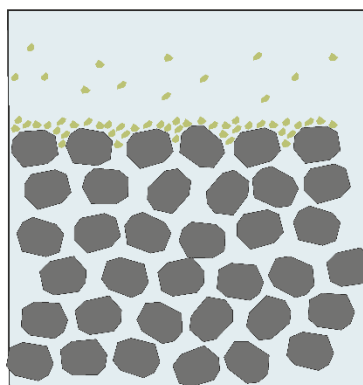
After a new leak is formed, the particles from the liquid slowly migrate deep into the pores of the soil and form a colmation layer. This process takes days or weeks.
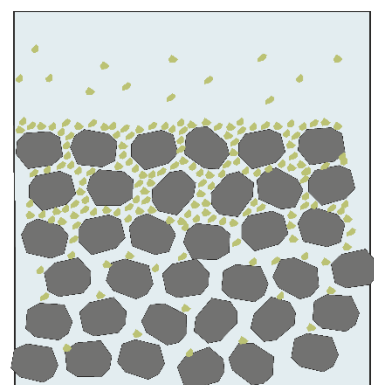


**Figure 1 Colmation of a porous media**

The clogging causes the formation of a colmation layer, which subsequently will govern the hydraulic capacity of the filter surface, and which also will define the active pore size of the filter material. Ultimately this leads to filter surfaces being able to retain particles significantly finer than the pore size of the virgin filter material. This phenomenon is well-known from various contexts where it has been shown to significantly enhance the ability of a soil to

retain pollutants (Vollertsen and Hvitved-Jacobsen, 2003, Dizer et al., 2004; Levy et all, 2011; Harvey et al., 2015). However, this gain does come at the cost of a reduced hydraulic capacity.

Very fine particles and colloids may remain mobile within soil pore spaces. Whilst the exact size of particles that can do this is not well-defined, they are typically within the colloid range. The understanding of this process is though still rather limited (Grayling et al., 2018).

All in all, and conceptually speaking, it is clear that OMP attached to or incorporated in particles will be retained if the particles are retained. Similarly will microplastics be retained if they are larger than what can be mobile in the soil matrix. A concrete quantification of what can pass through the soil and what cannot is though not possible with today's knowledge. From other areas of research it is known that for example silver nanoparticles can be retained efficiently in various soils (Degenkolb et al., 2018; He et al., 2019). It seems likely that microplastics – or at least some of the materials that constitute the microplastics – can also be retained quite efficiently even at the nanometric scale.

## 2.2 Degradation of OMP and MP

Some of the OMP will be degraded in the water phase, and some in the sediment phase. How fast MP is degraded is debated, but it is a well-established fact that plastics are not as durable as popular believes make them. Macroplastics are often degraded within years do decades, even when applying substantial efforts to conserve them, for example as museum exhibits (Shashoua, 2006). How fast microplastics are degraded is basically unknown (Li et al., 2018).

With respect to OMP, some substances are readily degradable by microorganisms while others are not. Most OMP are degraded by co-metabolism, where the degradation of the pollutant depends on the presence of a primary substrate (Lolas et al., 2012; Fischer and Majewsky et al, 2014; Gonzalez-Gil et al., 2017). In co-metabolism, an enzyme produced to catalyse the degradation of a metabolic substrate is also capable of degrading an OMP. In some cases, simultaneous catabolism can also play a role, where an organism derives energy from the degradation of both substrates. Similar to primary substrates, the degradation of many OMP will depend on the redox conditions, especially on the availability of oxygen. Many substances are degraded to varying degrees under both aerobic and anaerobic conditions, albeit typically by different species of micro-organisms. An example is the group of PAHs (polycyclic aromatic hydrocarbons) where the preferred degradation route is by molecular oxygen, but in anaerobic conditions degradation can also take place by organisms reducing sulphate, nitrate and metal compounds as well as by methanogens (Nzila, 2018).

Some substances are readily degraded without the intervention of microorganisms. Oxidative processes and hydrolysis can occur, for example, through light (photodegradation) (e.g. Minelgaite et al., 2017; Wang et al., 2017). Other forms of chemical degradation do also occur, for example by heat, acids, alkalis, salts, galvanic forces, etcetera. Which specific degradation process is actually responsible is seldom known specifically, but photodegradation is often expected to be a major mechanism responsible for chemical degradation of OMP.

Both biological and chemical degradation can cause the formation of breakdown products which also may be environmentally problematic, an issue which is well-known for, amongst

others, pesticides. For example the formation of aminomethylphosphonic acid (AMPA) during glyphosate degradation (e.g. Imfeld et al., 2013).

### 2.2.1 Plant uptake

Plants (macrophytes) do take up some pollutants from the surroundings, but plant uptake is in general not a viewed as an important process for the overall removal of pollutants from stormwater (Istenic et al., 2012; Stephansen et al., 2014). It is commonly assumed that the primary role of macrophytes, being they emergent or submerged, for treatment in a road runoff treatment system is to stabilize sediment beds and hereby reduce the risk of resuspension, and to reduce turbulence in the water phase and hereby enhance sedimentation (Brix, 1999).

### 2.2.2 Dilution and equalization of OMP and MP discharges

The time-scale of the impacts caused by an OMP or MP is important for how treatment should address the mitigation of these pollutants. If for example a pollutant has an acute toxic impact on the receiving environment but only occurs infrequently in the runoff, for example pesticides, attenuation and dilution is an appropriate approach to manage such event. The same is the case for managing of accidental spills, which can cause quite severe localized damage. Accidents are, however, infrequent and do seldom contribute substantially to the annual pollution load of a region. Attenuation and dilution can hence be an appropriate mitigation method. However, if the impact is cumulative, for example that of most heavy metals, it is important to bring down the annual load, while peak concentrations are of lesser or no importance.

Most of the pollutants in road runoff cause accumulate impacts and a main goal of road runoff treatment solutions is hence to reduce the annual load to the environment. But for accidental spills, peak concentrations commonly are of little or no consequence in this respect. Looking at treatment efficiencies, it is hence of limited value to study the efficiency of a system to mitigate a single runoff event. Instead the long-term load-reduction to the environment should be addressed.

## 3 Final remarks

The processes that are involved in retaining OMP and MP in road runoff treatment solutions, or for that matter in urban stormwater ponds, are still only understood at a rather generic level. Detailed knowledge of what exactly goes on in terms of physical, chemical, and biological processes in these systems is generally lacking. This fact has also been emphasized by several researchers attempting to model these systems, either as single systems, or by integrated approaches (Griffiths et al., 2017; Scott and Frost, 2017; Guan et al, 2018; Yang et al, 2018). As a consequence, the prediction of which substances and materials can be detained to what extend by which treatment system configuration will be similar generic. In other words, we will be able to put forward statements like "fluoranthene is retained rather efficiently in a soil filtration system because it has a high $K_{OW}$-value" and also come with statements like "fluoranthene will likely be contained more efficiently in a soil filtration system than acenaphthylene, because the latter has a lower log $K_{OW}$ value (5.16 and 3.93, respectively)". However, we will not be able to deduct the actual retention rate in a concrete system from theoretical knowledge only. To gain that information we will need to

conduct experimental studies and investigations on the substances and systems in question. We might then even dare some guestimates on retainment efficiency for similar substances, extrapolating our knowledge from the obtained database.

# 4  Acknowledgement

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