# PARTICLE FACILITATED TRANSPORT (COLLOIDS, DOM, MICELLES ETC.)

1.   PRINCIPLES

1.1   EQUILIBRIUM DISTRIBUTION

Contaminants may occur in aqueous solution ("truly dissolved") and associated to particulate matter or colloids suspended in water ("suspended solids"). Thus, the total concentration of a compound in water is elevated, its solubility seems to increase and its sorption and retardation during transport seems to decrease. This is relevant for risk assessment, remediation and transport of contaminants in general. Particles in rivers typically comprise suspended sediments or eroded soils during floods as well as urban "road dust" (deposited from the atmosphere) which become mobilized by storm water (see Fig. 1.1). Colloids are defined as particulate matter smaller than 0.45 µm in diameter and thus include dissolved organic matter (DOM), micelles formed by surfactants (above the critical micelle concentration) and lipid droplets in oil/water (micro-)emulsions. Colloids are much more difficult to separate from water than particles which can be removed by centrifugation or filtration. All these "particles" may become loaded with pollutants, e.g., in urban space and potentially become vectors for pollutant transport as discussed in Seidensticker et al. (2017, 2019) for microplastic particles.
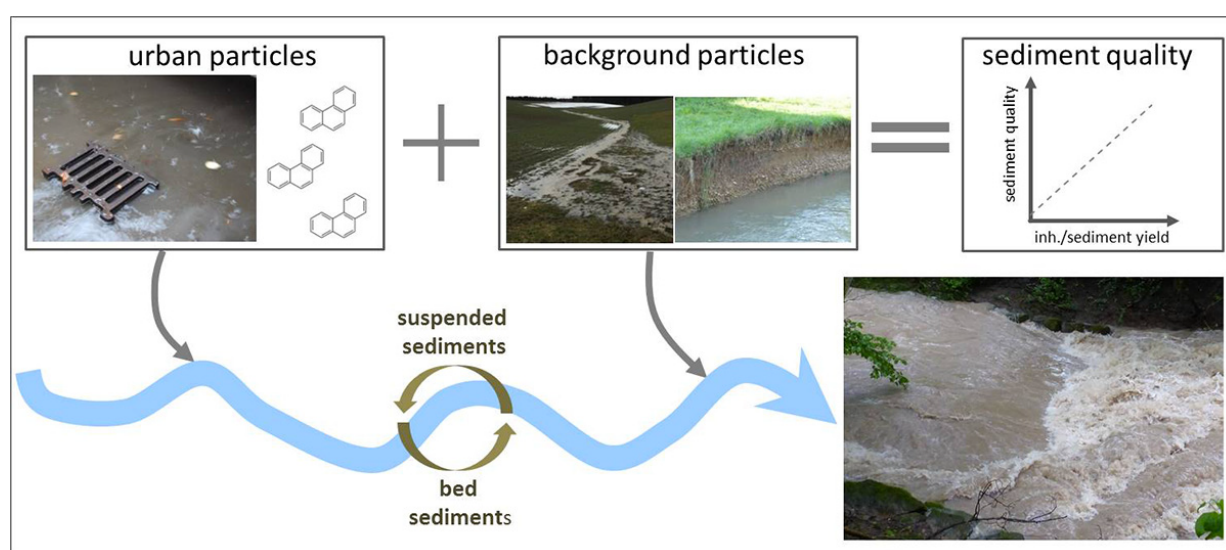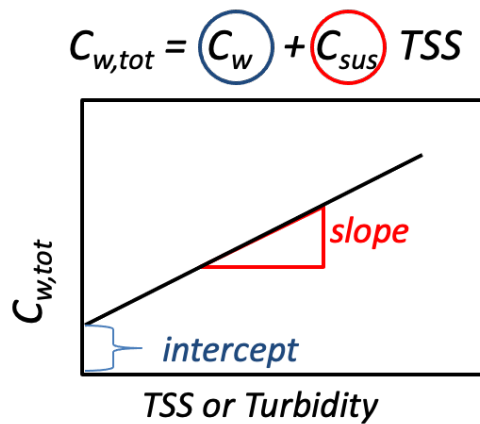

Fig. 1.1: Transport of particles in the environment (from Schwientek, 2017)

The total concentration in water in presence of suspended solids (particles and colloids) is:

$$C_{w,tot} = \frac{C_w V_w + C_{sus}\, m_d}{V_w} = C_w + C_{sus}\frac{m_d}{V_w} = C_w + C_{sus}\, TSS \tag{1.1}$$

where C_w denotes the aqueous concentration of the truly dissolved compound (e.g., in µg l⁻¹), C_{sus} is the concentration of the compound in suspended matter (in µg kg⁻¹) and TSS denotes the suspended solid concentration in the suspension (= m_d/V_w in kg l⁻¹). As shown in Fig. 1.2, eq. 1.1 represents a

linear relationship of $C_{w,tot}$ and TSS with the slope C_{sus} and the intercept C_w. For C_{sus} TSS >> C_w, $C_{w,tot}$ equals $C_{sus} \times TSS$ or C_{sus} is $C_{w,tot}/TSS$.

$$C_{w,tot} = \boxed{C_w} + \boxed{C_{sus}} \ TSS$$

Fig. 1.2: Transport of particles in the environment (from Schwientek, 2017)

In most cases C_{sus} may be expressed as a function of the aqueous concentration C_w ($C_s = C_w K_d$) and eq. 1.1 becomes:

$$C_{w,tot} = C_w + C_w K_{d,sus} \ TSS$$

or

$$\frac{C_{w,tot}}{C_w} = 1 + K_{d,sus} TSS$$

(1.2)

The right hand term ($1 + K_{d,sus} \ f_{sus}$) is also known as solubility enhancement factor (e.g. used in surfactant enhanced solubilization of NAPLs) which also reduces sorption and thus facilitates pollutant transport. Very efficient particle facilitated transport occurs with particulate organic matter (POM) in rivers but also in soils (Ngueleu et al., 2013).

Eq. 1.2 is also valid for facilitated transport of hydrophobic organic compounds in the presence suspended organic particles (or colloidal organic matter, e.g. humic acids, fulvic acid etc.); here the K_{oc} concept may be applied:

$$\frac{C_{w,tot}}{C_w} = 1 + K_{oc,sus} \ f_{oc,sus}$$

or for DOC

(1.3)

$$\frac{C_{w,tot}}{C_w} = 1 + K_{DOC} \ f_{DOC}$$

where the subscripts *oc, sus* and *DOC* denote the fraction of organic carbon content of the suspended solids and the dissolved organic carbon in water. K_{oc} and K_{DOC} are in the same order of magnitude with K_{oc} approximately a factor of 2 higher than K_{DOC}. Of course, transport facilitated by dissolved organic matter (DOM) and suspended particles (or particulate organic matter) can occur in parallel and mathematically is treated the same way. Fig. 1.3 shows how the fraction of particles and DOC affects total concentrations in water.

All this boils down (again) to the question how much is dissolved compared to the total which is the inverse of eqs. 1.2 and 1.3 (see Fig. 1.4):

$$f_{dissolved} = \frac{C_w}{C_{w,tot}} = \frac{C_w V_w}{C_w V_w + C_s m_d} = \frac{C_w}{C_w + C_s \frac{m_d}{V_w}} = \frac{C_w}{C_w + K_d C_w \frac{m_d}{V_w}} = \frac{1}{1 + K_d \frac{m_d}{V_w}} \qquad 1.4$$

The fraction on the solids relative to the total capacity is (the inverse curve shown in Fig. 1.3):

$$\frac{C_s}{C_{w,tot}} = \frac{C_s m_d}{C_w V_w + C_s m_d} = \frac{C_s m_d}{C_w V_w + C_s m_d} = \frac{1}{\frac{V_w}{K_d m_d} + 1} \qquad 1.5$$

Note, that the principles explained here also apply to the atmosphere in terms of fractions of pollutants in the gas phase compared to the fraction in particulate atmospheric matter (e.g. $PM_{2.5}$, PM_{10}, soot, black carbon, microplastics, etc.).
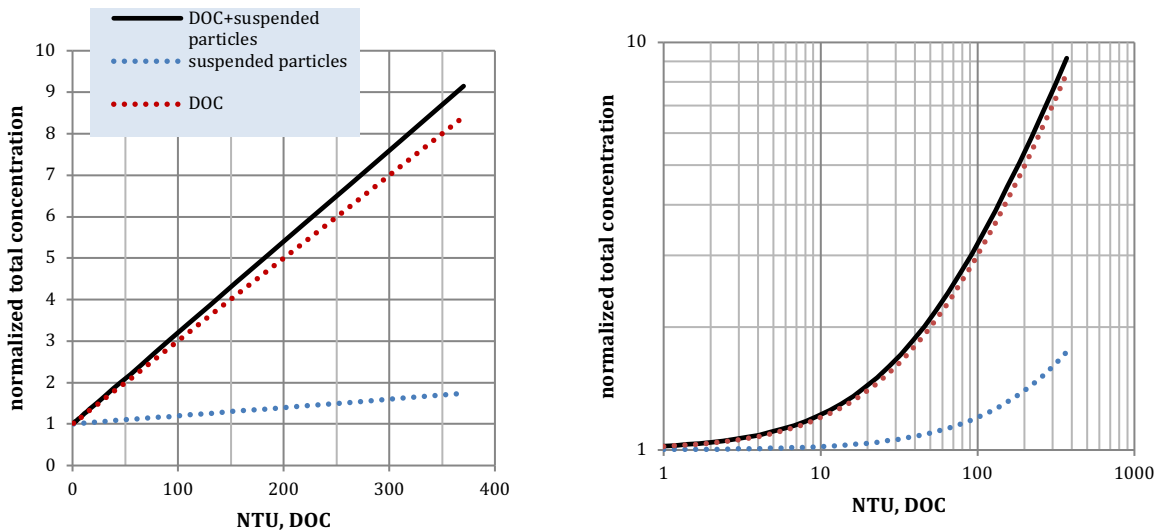

Fig. 1.3: Normalized concentrations ($C_{w,tot}/C_w$) *vs.* DOC (mg l^{-1}) and turbidity units (NTU) which corresponds to approximately 1 mg l^{-1} suspended solids; f_{oc} of suspended solids = 10%; $K_{oc,sus}$ and K_{DOC} = 20 000. DOC dominates the solubility enhancement and facilitated transport at similar values of NTU or DOC, respectively; note, however, during floods NTU is over 100 while DOC gets diluted to e.g. less than 10 mg l^{-1}.
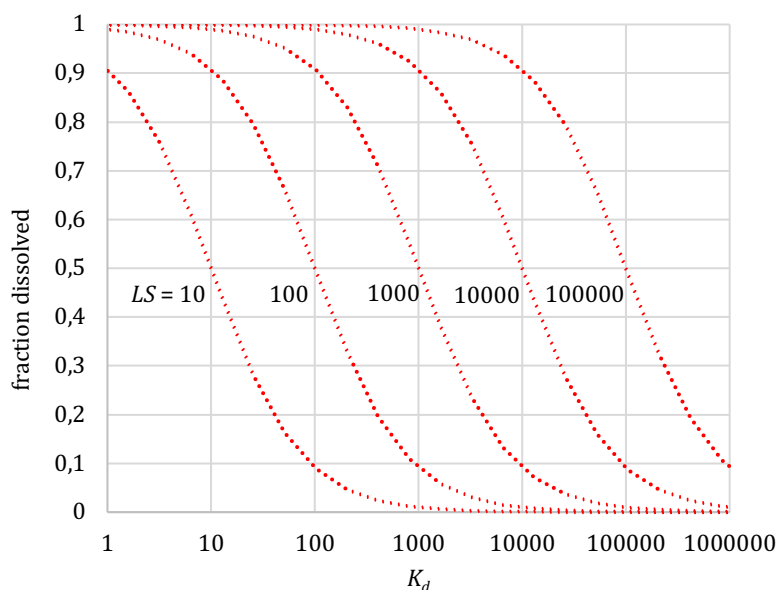
Fig. 1.4: Fraction dissolved (C_w / $C_{w,tot}$ = 1/(1+ K_d m_d/V_w)) *vs.* K_d of suspended solids; number on lines denote liquid to solid ratios – if LS equals K_d then 50% of the compound is freely dissolved, if K_d decreases (increases) one order of magnitude then already 90% (only 10%) are dissolved; in rivers the total suspended solid concentrations are about 10 mg l[-1] (V_w/m_d: 100 000 l kg[-1]) but during floods 100 – 1000 mg l[-1] (V_w/m_d: 10 000 – 1000) are reached.

1.2   KINETICS

How fast do contaminants move from a particle into water and/or to other clean (background) particles? Box 2 shows an example calculation on this (based on first order kinetics, diffusion through an aqueous boundary layer) and Fig. 1.5 characteristic times as a function of K_d. For infinite bath conditions, mass transfer kinetics slows down with increasing K_d. For small particles (here 0.1 mm diameter) equilibrium is reached relatively fast, time scales depend on Sherwood numbers (Sh) e.g., in rivers or lakes (or stirred reactors in laboratory experiments). Fig. 1.6 gives an overview on Sherwood number relationships for different flow/turbulence regimes. If intraparticle diffusion becomes relevant (e.g., in hard plastic particles) time scales may be much longer. In order to evaluate whether diffusion in the particle or though the external aqueous boundary layer limits mass transfer the dual film diffusion model can be evaluated (see section D1, chap. 2). Note, particles are only vectors for pollutant transport if desorption kinetic is slow enough and pollutants do not desorb completely before other environmental compartments are reached. Particles which equilibrate rapidly rather mirror their environment and function as passive samplers (Seidensticker et al., 2019), e.g., the concentration on particles reflects the pollution of the water in oceans and lakes.
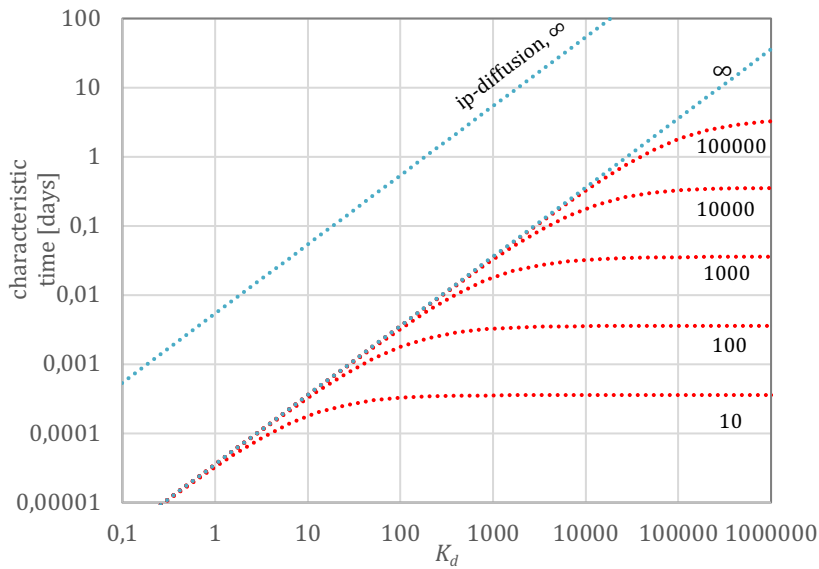
Fig. 1.5: Characteristic time for sorption/desorption (63.2%) vs. K_d for different liquid/solid ratios (, dottet red lines, numbers denote V_w/m_d – the invers is the suspended solids concentration); 100 µm sized suspended particles with a minimum Sherwood number of 2 (low Reynolds numbers). If K_d values get larger than V_w/m_d, (more than 50% dissolved), then kinetics become independent on K_d. Diagonal blue lines denote infinite bath conditions; for intraparticle diffusion (ip) an internal porosity of 0.1 is assumed, thus the effective diffusion coefficient is 2 orders of magnitude smaller than in water (slows kinetics down by two orders of magnitude).
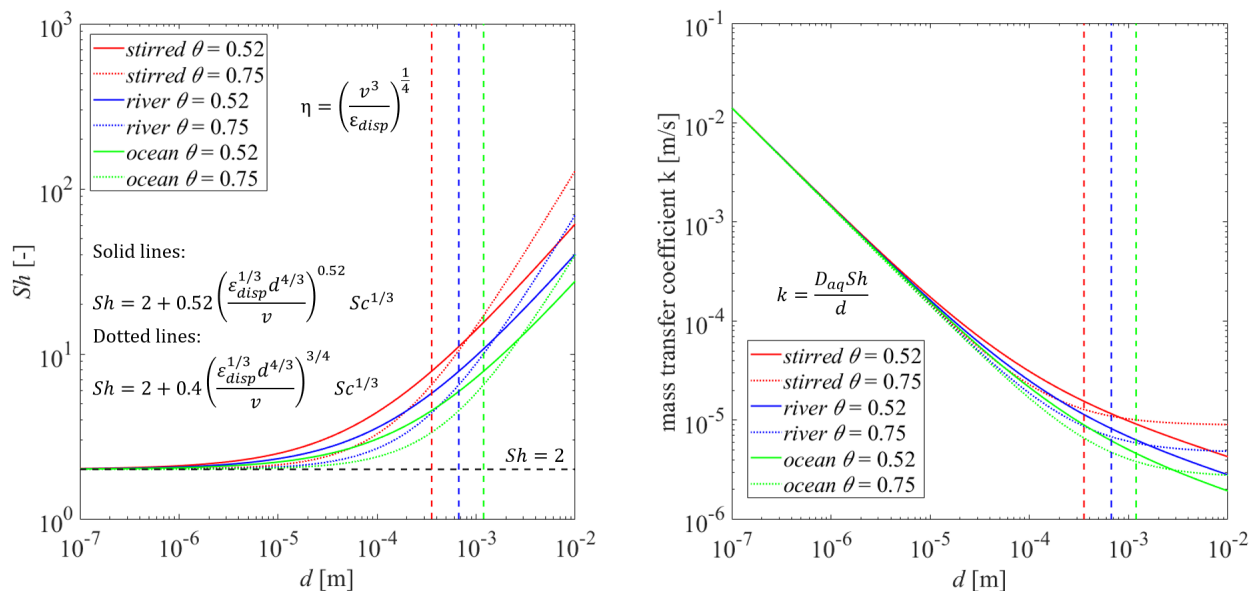
Fig. 1.6. Influence of particle size (d) on Sherwood numbers (left) and mass transfer coefficients (right) in different flow systems (stirred laboratory experiments, rivers, ocean with energy dissipation rates (ε_{disp}) of $10^{-4.2}$ m² s⁻³, 5×10⁻⁶ m² s⁻³, and 5×10⁻⁷ m² s⁻³). The horizontal and vertical dashed lines denote the minimum Sherwood number ($Sh = 2$) and Kolmogorov length scales $\left(\eta = v^3/\varepsilon_{disp}\right)^{1/4}$, repetively.

From: Liu, B., Finkel, M., Grathwohl, P. (2022). First order approximation for coupled film and intraparticle pore diffusion to model sorption/desorption batch experiments. J. Haz. Mat., 429, 128314
https://doi.org/10.1016/j.jhazmat.2022.128314

Box 1.1: Kinetics – how fast does the water equilibrate with suspended particles (e.g. desorption from "microplastics")?
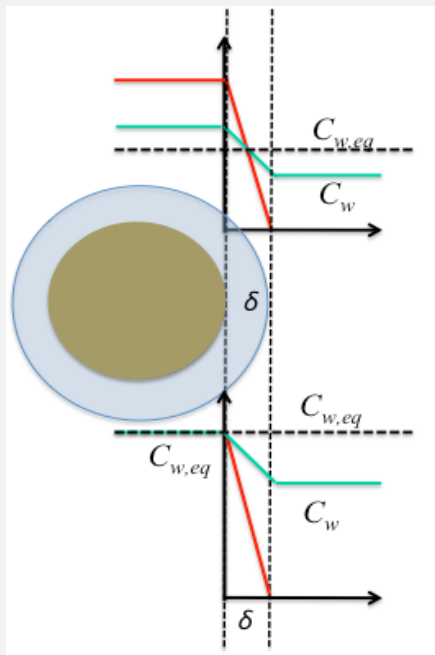
$$\frac{C_w}{C_{w,eq}} = 1 - \exp\left(-k\,\frac{6}{\rho\,d}\left(\frac{1}{K_d} + \frac{m_d}{V_w}\right)t\right)$$

or

$$\frac{C_w}{C_{w,eq}} = 1 - \exp\left(-k\,\frac{6}{K_d\,\rho\,d}\left(1 + K_d\,\frac{m_d}{V_w}\right)t\right)$$

For very large K_d values (> V_w/m_d i.e. low fraction dissolved) we find that desorption kinetics is independent on K_d:

$$\frac{C_w}{C_{w,eq}} = 1 - \exp\left(-k\,\frac{6}{\rho\,d}\,\frac{m_d}{V_w}\,t\right)$$

k is the mass transfer coefficient (= D_{aq}/δ).

The film thickness δ may be estimated based on the Sherwood number ($Sh = \frac{k\,d}{D_{aq}} = \frac{d}{\delta} \implies \delta = \frac{d}{Sh}$)

For low Reynolds numbers or small particles, e.g., with radius of 10 μm (d = 20 μm) the Sherwood number is small, e.g., 2 and δ equals the radius. The mass transfer coefficient for small particles then is approximately: $k = D_{aq}\,Sh/d = D_{aq}/\delta = 5 \times 10^{-10}\ \mathrm{m^2 s^{-1}}/1 \times 10^{-5}\ \mathrm{m} = 5 \times 10^{-5}\ \mathrm{m/s}$

The specific surface area (A^o) available for diffusion in the infinite bath corresponds to the surface to mass ratio of the particle (= $6/(d\,\rho)$ [m² kg-1]; in the finite bath (batch system) the solid to liquid ratio ($m_d\,/\,V_w$) leads to specific surface area per volume of water in m² per m³ (= $6/(d\,\rho)\,m_d\,/\,V_w$ [m² m-3 = m-1]. ρ is the density of the solid often around 2700 kg m-3. For a turbidity of 100 mg l-1 and 20 μm-sized particles A^o is: $0.1\ \mathrm{[kg\ m^{-3}]} \times 6/2650\ \mathrm{[kg\ m^{-3}]}\ /\ 20 \times 10^{-6}\mathrm{[m]} = 11.3\ \mathrm{[m^2\ m^{-3} = m^{-1}]}$. The smallest (at Sh = 2) overall rate constant (at Sh = 2) for large K_d-values then is 5.65×10^{-4} s-1. The characteristic time (63% of equilibrium reached is then 1800 s (or 30 min), if the grains size was 10 times larger, it could take 100 times longer (ca. 2 days). Note, that mass transfer in such systems (e.g., a river) is not well known.

For desorption in the infinite bath (the contaminant finally desorbs completely and the particle becomes clean) the relative change of the concentration in the particle is:

$$\frac{C_s}{C_{s,eq}} = \exp\left(-\frac{k}{K_d}\,\frac{6}{\rho\,d}\,t\right)$$

Now the desorption time scale increases with increasing sorption and for a K_d of 10 000 l kg-1 the overall rate constant is: $5 \times 10^{-5}\ \mathrm{[m\ s^{-1}]} \times 6/10\,000\ \mathrm{[l\ kg^{-1}]}/2.65\ \mathrm{[kg\ l^{-1}]}\ /\ 20 \times 10^{-6}\mathrm{[m]} = 5.66 \times 10^{-4}\ \mathrm{[s^{-1}]}$. The characteristic time is again 1800 s (30 min or 0.5h). Note, that with increasing desorption (> 50% desorbed) the mass transfer resistance may shift into the particle and slows down significantly, the characteristic time for 90% desorption with intraparticle diffusion is:

$$t_{90} = 0.183\,\frac{r^2}{D_a} \approx 0.183\,\frac{r^2\,K_d\,\rho}{D_e}$$

Assuming an effective diffusion coefficient two order of magnitude smaller than in water ($D_e = 5 \times 10^{-12}$ m² s-1 e.g., at an internal porosity of 0.1) the desorption time scale would now amount to 27 hours.

## 2.   TRANSPORT BY SUSPENDED PARTICLES AND DOC

2.1   ANALYSIS OF PARTICLES AND DOC

Suspended particles in water cause scattering of light leading and lead to turbidity (cloudiness) of the water. Turbidity can be quantified by measuring the amount of light scattered in a water sample sideways from a light beam using a "nephelometer". Turbidity is reported in Nephelometric Turbidity Units (**NTU**). Another widely used units for turbidity is FTU ([Formazin](#) Turbidity Unit; formazin is an aqueous suspension of 5 g/L [hydrazine sulfate](#) and 50 g/L [hexamethylenetetramine](#)). [ISO](#) (International Standardization Organization) uses FNU (Formazin Nephelometric Units). The amount of light scattered depends on particle size, shape, color and reflectivity. Therefore, correlations between the total amount of suspended solids and NTU maybe specific for each location or situation (e.g., Thackston and Palermo, 2000). However, as Fig. 2.1 shows, differences are only within a factor of 2 and 1 NTU corresponds roughly to 1 mg l^{-1} suspended solids (Rügner et al., 2013).
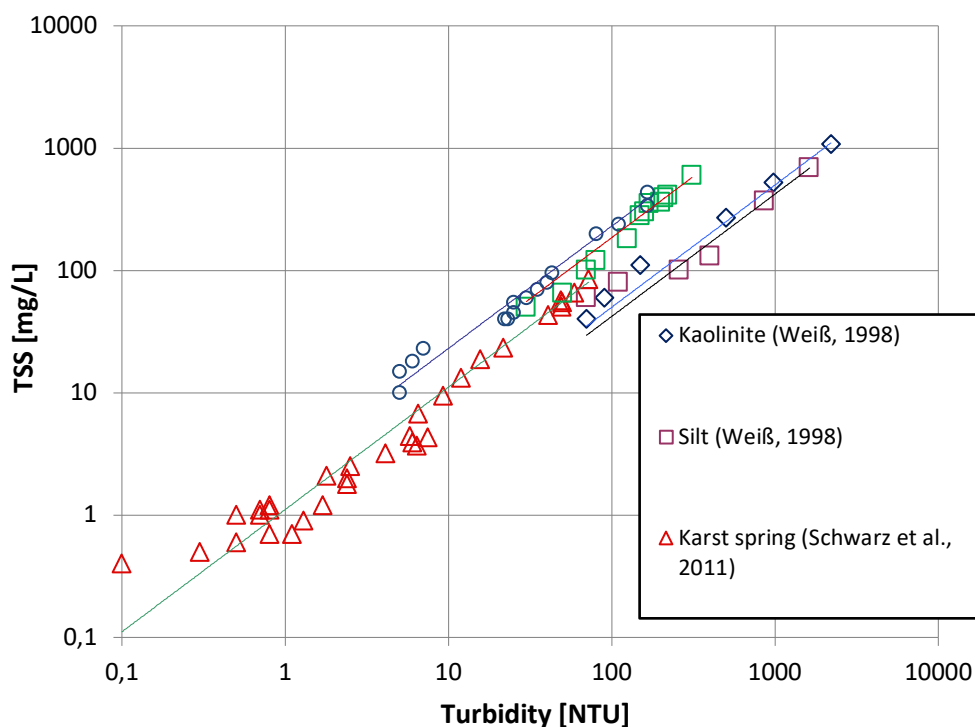
Fig. 2.1: Correlations of total amount of suspended solids (particles > 0.45 μm) to turbidity measured with light scattering (NTU); from Schwientek et al. (2013).

Dissolved organic matter is hard to separate from water and thus is measured via the organic carbon content of a water sample. The most common method is based of infrared detection of CO_2 which is obtained by combustion of the sample at around 1000°C under oxygen (oxidation of organic carbon). Before the measurement inorganic carbon and CO_2 is removed by acidification and N_2 stripping of the sample. DOC is usually given in mg l^{-1}. At concentration above 50 mg l^{-1} the water starts to show distinct yellow colors, at several 100 mg l^{-1} it looks dark to black (Fig. 2.2).
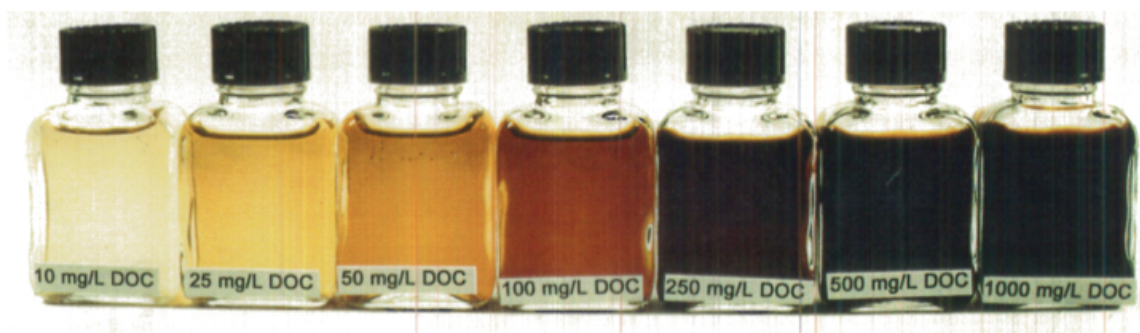
Fig. 2.2: Coloring of water samples with increasing DOC (mg l[-1]); Aldrich Humic Acid (Pyka, 1994).

2.2   SOLUBILIZATION BY DOC (OR MICELLES)

As the amount organic colloidal matter increases, its pollutant load may dominate over the "truly" dissolved concentration (C_w) and in compound mixtures, e.g. PAHs the distribution pattern shifts from a predominance of low molecular weight compounds which have a relatively high solubility towards low solubility compounds as shown in Fig. 2.3 as expected:

$$C_{w,tot} = (C_w +)\, K_{DOC} C_w f_{DOC} \qquad\qquad 2.1$$

$K_{DOC}\, C_w\, f_{DOC}$ represents the concentration in the suspended organic colloid. At high f_{DOC} and high K_{DOC} the concentration in water is governed by the amount DOC. As known for particulate carbonaceous materials in soils and sediments, dissolved organic matter may exhibit different affinities for organic solutes depending on the molecular properties (Pyka, 1994, Gauthier et al., 1987, Neale et al., 2011). Pyka (1994), e.g., found increasing K_{DOC} values with increasing absorption of light at a wavelength of around 436 nm. E.g., for fluoroanthene K_{DOC} values increase with: $K_{DOC} = 5013\, Abs_{436}\, 1000$ and amount to 25000 for humic acids and while for fulvic acids only 6300 are reached.
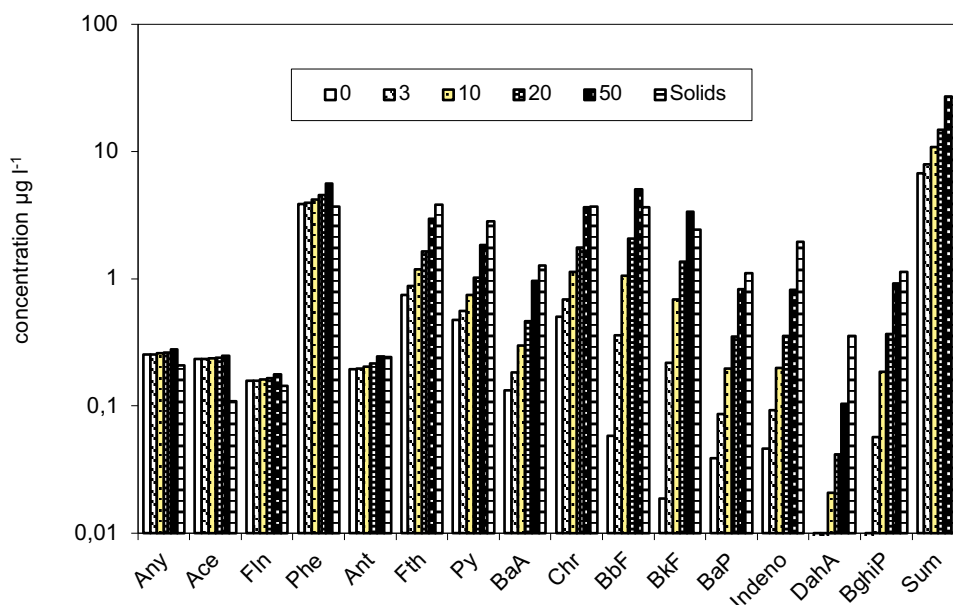

Fig. 2.3: Distribution pattern of 16 EPA PAHs in aqueous leachate concentrations of soils. With increasing DOC (0 - 50 mg l[-1]) the aqueous phase concentrations shift towards the distribution pattern of the solids (right bar for each PAH). Low molecular weight PAHs up to anthracene (Ant) are not much affected and a DOC of more than 20 mg l[-1] is needed to cause a significant increase in the sum of the 16 EPA PAHs.
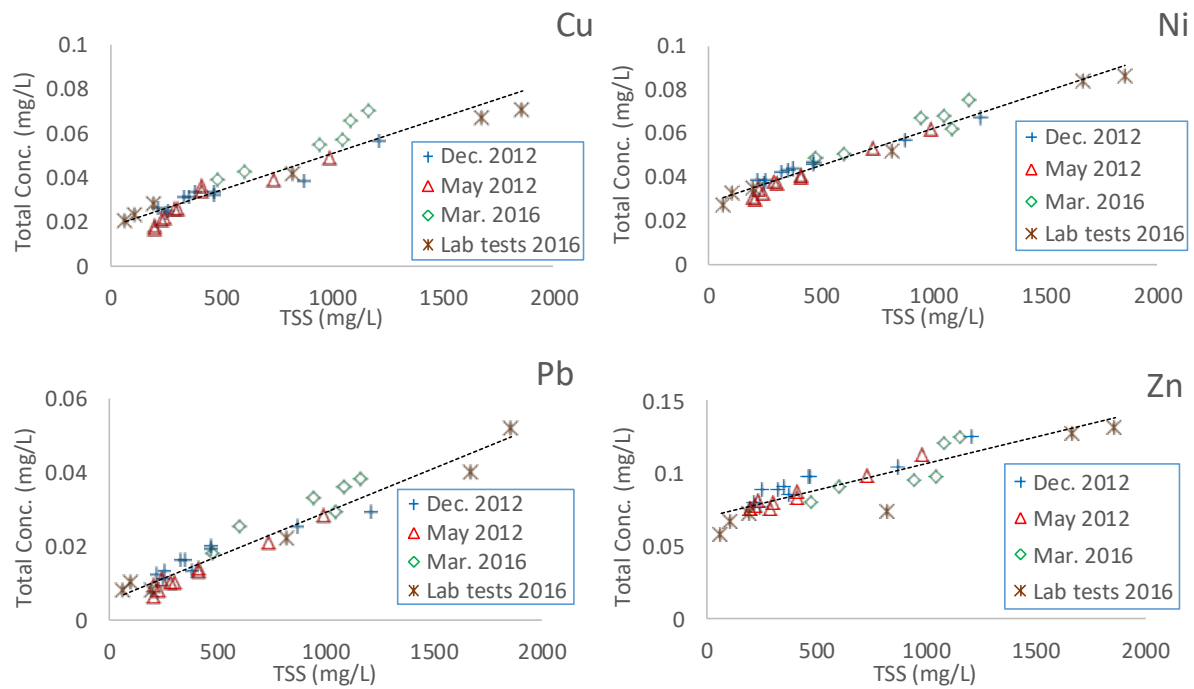
## 2.3 PARTICLE FACILITATED TRANSPORT OF PAHS AND METALS IN RIVERS

Figs. 2.4 and 2.5 illustrate particle facilitate transport in rivers. In general, data from different seasons and years as well as lab tests show very good reproducibility (Rügner et al., 2014, Nasrabadi et al., 2018) and follow nicely eq. 1.1 as to be expected ($C_{w,tot} = C_w + C_{sus} \times TSS$). While PAHs come almost entirely from anthropogenic emissions into the environment, heavy metals and other elements often reflect geogenic background concentrations in soils or more generally in the earth crust (Nasrabadi et al., 2018). Particle facilitated transport in the environment can be tracked from the atmosphere over atmospheric deposition to road dust and finally suspended sediments in rivers (Nasrabadi et al., 2021). As shown in Fig. 2.6 the concentration of anthropogenic pollutants on total suspended particles (C_{sus}) depends on the urban impact (or urban particle input) on the catchments with respect to "clean" background particle abundance (Schwientek et al., 2013, 2017); alpine or remote rivers typically have lower pollutant loadings on particles compared to urban settings (Rügner et al., 2019).
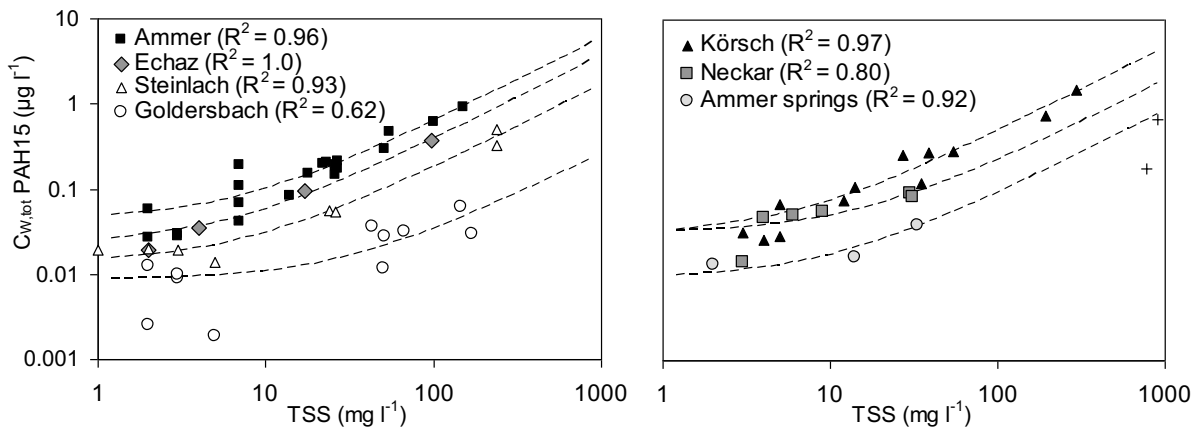
Fig. 2.5: Linear regressions of total heavy metal concentrations vs. TSS during different sampling campaigns in the lower Haraz catchment (March 2016 plus lab tests, 2012 data adapted from Nasrabadi et al., 2016); from Nasrabadi et al., 2018.
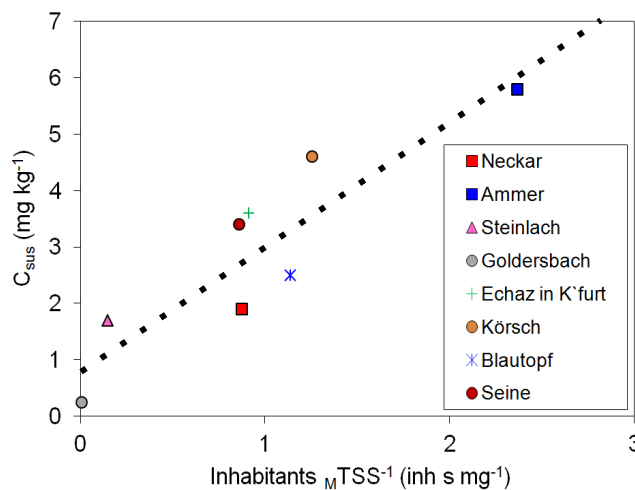
Fig. 2.6: PAH concentrations on suspended particles (C_{sus}) versus inhabitants normalized by the mean sediment mass yield ($_M$TSS); adapted from Schwientek et al., 2013 (see also Schwientek 2017).

3.   COSORPTION AND MICELLE (OR COLLOID) FACILITATED TRANSPORT

If particles, colloids, micelles, etc. act as pollutant carriers in the environment, their interaction with environmental surfaces becomes also relevant with respect to pollutant transport. This is obvious for transport in porous media (soils and aquifers) as illustrated on Fig. 3.1. Particles and colloids may be retained in porous media, organic substances such as humic and fulvic acids undergo adsorption on surfaces and with them pollutants are trapped. In the following, this is discussed for cotransport surfactant micelles and pollutants which applies also to natural organic substances.
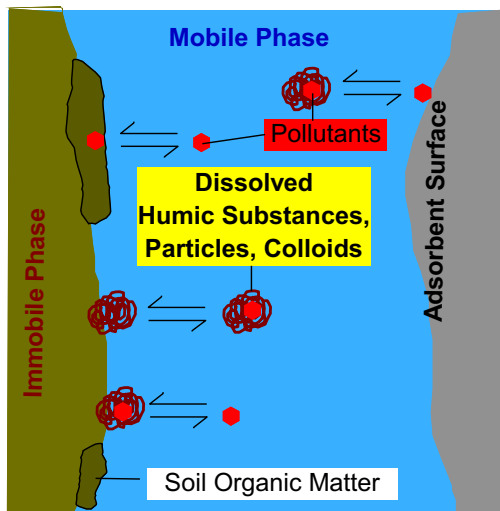

Fig. 3.1: Facilitated transport in a porous medium

Surfactants are able to form micelles above the critical micelle concentration and at the same time admicelles may form on mineral surfaces as shown in Fig. 3.2. Usually a maximum occupation ("loading") of the mineral surface is reached (q_{max}) as shown in Fig. 3.3 – surfaces are then coated with a "organic film". Since the available surface area per mass of solids is highest for finer grained solids the amount of adsorbed surfactants increases with decreasing grain size as shown in Fig. 3.4.
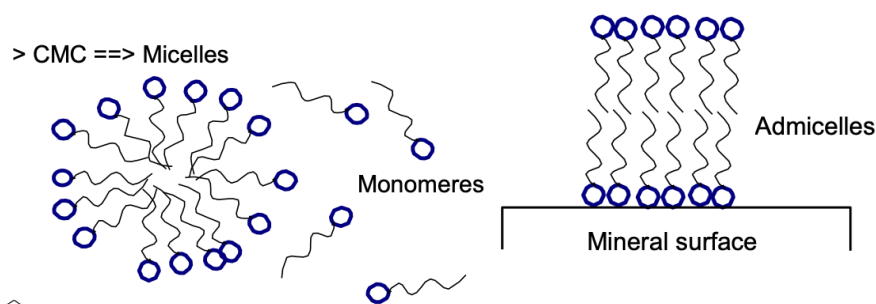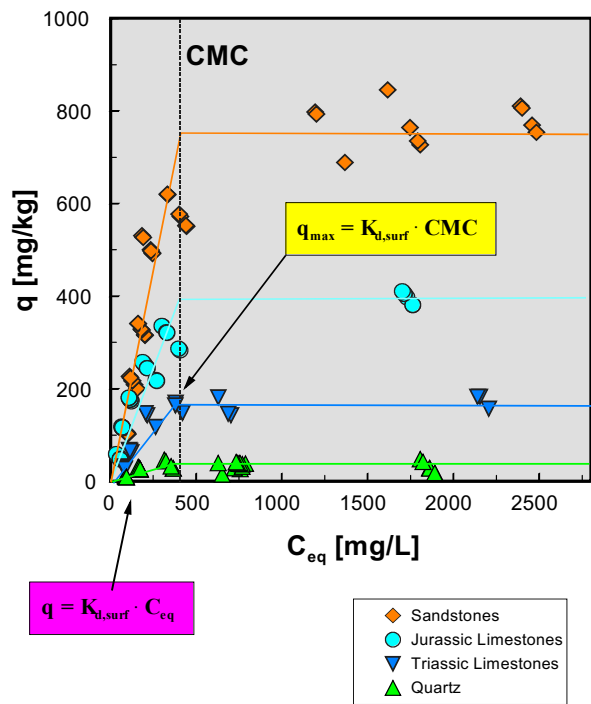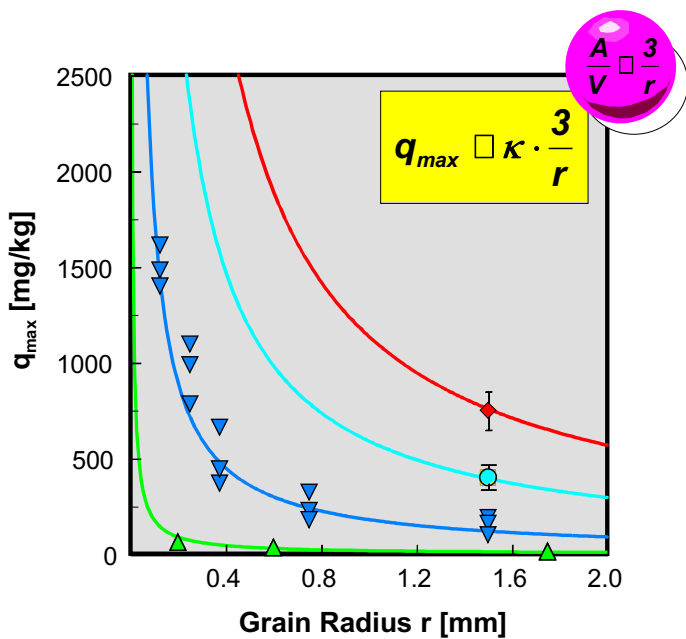

Fig. 3.2: Surfactants form micelles above the critical micelle concentration (CMC) and admicelles by adsorption on mineral surfaces

Fig. 3.3: Adsorption of surfactants at different rocks and minerals (Example: Terrasurf G50,DOW Chemicals), see Danzer and Grathwohl (1998)

In the first figure: CMC, $q_{max} = K_{d,surf} \cdot CMC$, $q = K_{d,surf} \cdot C_{eq}$

Legend:
- Sandstones
- Jurassic Limestones
- Triassic Limestones
- Quartz

$$\frac{A}{V} = \frac{3}{r}$$

$$q_{max} = \kappa \cdot \frac{3}{r}$$

Fig. 3.3: Increasing adsorbed concentrations with decreasing grain size (Example: Terrasurf G50,DOW Chemicals), (see Danzer and Grathwohl, 1998)

Like mobile micelles, adsorbed surfactants ("admicelles") are capable of taking up hydrophobic contaminants (= "cooperative" sorption) leading to an increased retardation of contaminants as long as the CMC is not exceeded. If the surfactant concentration is just below the CMC, a maximum retardation in contaminant transport is observed (in situ sorption barriers) and above the CMC facilitated transport of contaminants starts. The concept and the derivation of retardation factors for such a system is depicted in Box. 2.1. It should be noted that humic substances (DOC) can be treated analogously to surfactants.

The retardation factor for micelle (or DOC) affected transport of organic contaminants can be simply calculated based on the **"total to mobile ratio"**:

$$R_d = \frac{total\ amount\ of\ contaminant\ (\text{"capacity"})}{mobile\ fraction}$$

$$= \frac{C_w\, n + C_w\, K_{oc}\, f_{oc}\, \rho_b + C_w\, K_{mic}\, f_{mic} + C_w\, K_{admic}\, f_{admic}}{C_w\, n + C_w\, K_{mic}\, f_{mic}}$$

$$= \frac{n + K_{oc}\, f_{oc}\, \rho_b + K_{mic}\, f_{mic} + K_{admic}\, f_{admic}}{n + K_{mic}\, f_{mic}}$$

if for strongly sorbing compound n is negligible and if $K_{mic} \approx K_{oc} \approx K_{admic}$ we get an approximation: 　　　　(2.2)

$$R_d = \frac{f_{oc}\, \rho_b + f_{mic} + f_{admic}}{f_{mic}}$$

and if sorption is negligible ($f_{oc}\, \rho_b$ very small) R_d becomes.

$$R_d = \frac{f_{mic} + f_{admic}}{f_{mic}} = 1 + \frac{f_{admic}}{f_{mic}}$$

and finally for large f_{mic} R_d decreases to 1.

The sorption of admicelles (same applies for DOM) causes retarded transport unless the micelle (or dissolved organic colloids, DOC) concentration becomes really high (i.e., g l^{-1}). This is because of the strong sorption of surfactants (or DOC).

The various partitioning coefficients needed may be estimated using databases such as the one shown at the end of this section (see also Valsaraj and Thibodeaux, 1989).

Box: 2.1: Coupled transport of contaminants and surfactants (see Danzer and Grathwohl, 1998)

$\underline{K_d}$ for cosorption without free micelles, i.e. only admicelles $\underline{(C_{surfactant} \le CMC)}$:

$K_{d,admic} = f_{oc}K_{oc} + f_{admic}K_{admic}$ with: $K_{admic} \approx K_{mic} \approx 0.35\ K_{OW}$; $f_{admic} = q_{max}$ [g/g]
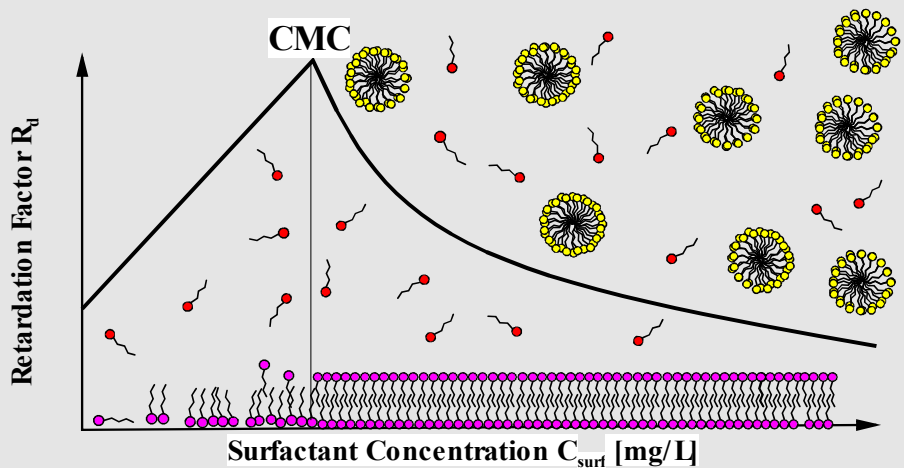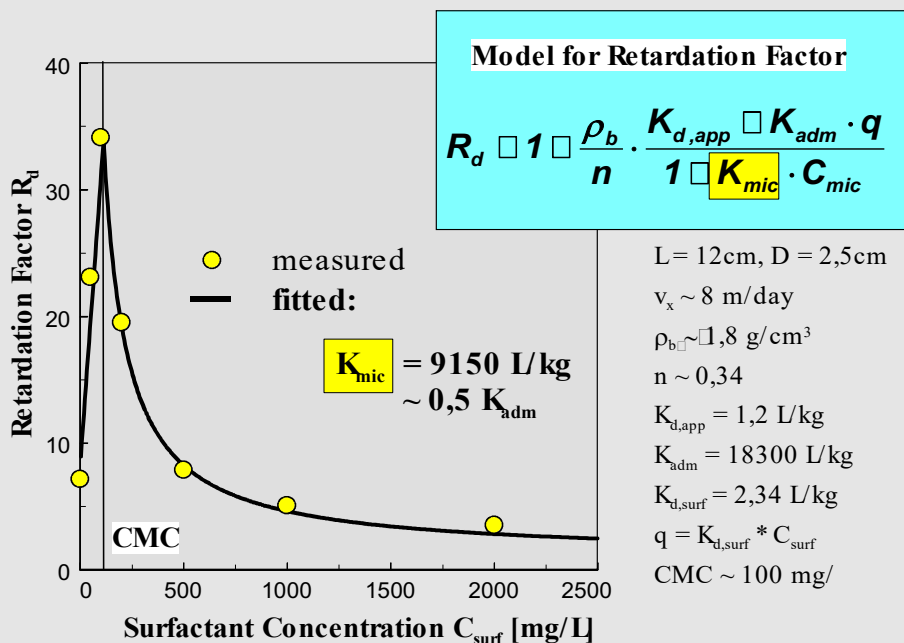
$\underline{K_d}$ for reduction of the sorption in the presence of free micelles, i.e. $\underline{C_{surfactant} >> CMC}$:

$K_{d,mic} = \dfrac{f_{oc}K_{oc}}{1+f_{mic}K_{mic}}$ with: $K_{mic} \approx 0.35\ K_{OW}$; $f_{mic} = C_{surfactant}$ [g/L] for $C_{surfactant} >> CMC$

(note that the factor $1 + f_{mic}\ K_{mic}$ represents also the apparent increase of the water solubility with the application of surfactants).

Phenanthrene Retardation Factors vs. Surfactant Concentrations
Neckar River Alluvial (1-2 mm), Terrasurf G50

Model for Retardation Factor

$$R_d = 1 + \frac{\rho_b}{n} \cdot \frac{K_{d,app} + K_{adm} \cdot q}{1 + K_{mic} \cdot C_{mic}}$$

○ measured

━ **fitted:**

K_{mic} = 9150 L/kg
$\sim 0.5\ K_{adm}$

L = 12cm, D = 2,5cm

$v_x \sim 8$ m/day

$\rho_b \sim 1,8$ g/cm³

n $\sim 0,34$

$K_{d,app}$ = 1,2 L/kg

K_{adm} = 18300 L/kg

$K_{d,surf}$ = 2,34 L/kg

$q = K_{d,surf} * C_{surf}$

CMC $\sim$ 100 mg/

CMC

Retardation Factor R_d (y-axis)

Surfactant Concentration C_{surf} [mg/L] (x-axis)

CMC

Retardation Factor R_d (y-axis)

Surfactant Concentration C_{surf} [mg/L] (x-axis)

https://www.ufz.de/index.php?en=31698&contentonly=1&m=0&lserd_data[mvc]=Public/start&lserd_data[formSubmit]=1

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