

Anthropogenic pollutants affect ecosystem services of freshwater sediments: the need for a “triad plus x” approach

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Abstract

Purpose Freshwater sediments and their attached microbial communities (biofilms) are essential features of rivers and lakes, providing valuable ecosystem services such as nutrient recycling or self-purification which extend beyond the aquatic environment. Anthropogenic pollutants, whether from the industrial era or as a result of our contemporary lifestyles, can negatively affect these functions with hitherto unknown consequences on ecology, the economy and human health.

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Thus far, the singular view of the involved disciplines such as ecotoxicology, environmental microbiology, hydrology and geomorphology has prevented a deeper understanding of this emerging issue.

Main features This paper discusses briefly the progressions and the state-of-the-art methods within the disciplines of concern related to contaminated sediments, ranging from ecotoxicological test systems, microbiological/molecular approaches to unravel changes of microbial ecosystems, up to the modelling of sediment transport and sorption/desorption of associated pollutants. The first bilateral research efforts on contaminated sediments include efforts to assess ecotoxicological sediment risk including sediment mobility (i.e. ecotoxicology and engineering), enhance bioremediation potential (i.e. microbiology and ecotoxicology) or to understand biostabilisation processes of sediments by microbial assemblages (i.e. microbiology and engineering).

Conclusions and perspectives In freshwater habitats, acute, chronic and mechanism-specific toxic effects on organisms, shifts in composition, structure and functionality of benthic microbial communities, as well as the obstruction of important ecosystem services by continuously discharged and long-deposited pollutants, should be related to the in situ sediment dynamics. To achieve an improved understanding of the ecology of freshwater sediments and the impairment of their important ecosystem functions by human-derived pollutants, we suggest a “triad plus x” approach combining advanced methods of ecotoxicology, environmental microbiology and engineering science.

Keywords Biofilm · Freshwater · Interdisciplinary approach · Management of sediments · Pollutants · Risk assessment

1 Addressing pollutants in aquatic habitats: short history and emerging challenges in the involved natural sciences

1.1 Introduction

1.1.1 Freshwater sediments and ecosystem services

Freshwater sediments are complex ecosystems comprising multifaceted structures of geological origin and biological provenance (e.g. matrix of extracellular polymeric substances (EPS), humic substances) as well as a variety of autotrophic and heterotrophic communities. The initial settlement of organisms, their further development, architectural capacity and functionality depends on and interacts with abiotic factors in the environment (e.g. sediments, hydrodynamics, chemistry, nutrient availability; Fig. 1). The surface-associated sediment communities (i.e. biofilms) contribute notably to the essential functionality of lakes and rivers in providing a variety of ecosystem services that reach far beyond the aquatic habitats from which they originate. Ecosystem

functions cover provisioning (e.g. food, clean drinking water, energy generation); regulating (e.g. carbon sequestration, decomposition, detoxification, self-purification); and supporting (e.g. biogeochemical cycles, biostabilisation) services (see Fig. 1), which are important for the river system health and consequently for biodiversity resources, availability of high-quality drinking water and recreation.

1.1.2 The challenge: anthropogenic pollutants

Despite significantly improved water quality in freshwater habitats of the Northern hemisphere over the last few decades, their associated sediments still store large quantities of hazardous substances originated from the past era of almost unlimited industrial production (“Legacy of the past”; Förstner et al. 2004). Whilst it is already difficult to deal properly with this time bomb of sediment-associated heavy metals and organic pollutants, we are being confronted by new challenges. A spate of recent persistent chemicals—nanoparticles, perfluorinated compounds (Giesy and Kannan 2002), pharmaceuticals and personal care products (Kümmerer 2009)—which originate from the modern lifestyles of the industrial nations are increasingly accumulating within the aquatic environments around the globe. Catastrophic events such as the Elbe River floods in 2002 provided clear evidence of the ecological, economical and human health-related consequences of remobilized pollutants from the sediment, affecting freshwater habitats, adjacent areas (agricultural floodplains, residential areas) and finally the marine environment (Kammann et al. 2005; Oetken et al. 2005). Since the Sandoz disaster of 1986 (Giger 2009), great efforts have been undertaken to gain a better understanding of the complex pattern of pollutant effects on sediments and beyond. Yet, the mostly singular view of the involved disciplines has consistently shown that they are inadequate in providing a fundamental understanding of the functionality of freshwater sediments and the potential impact by anthropogenic pollutants. Initial attempts to combine different methodologies were a revelation in realizing the clear limits of the single disciplines (Hollert et al. 2005; Keiter et al. 2008). In the following, the achievements within ecotoxicology, microbiology and engineering science concerning sediment and pollutant dynamics in freshwaters will be described briefly and research needs presented.

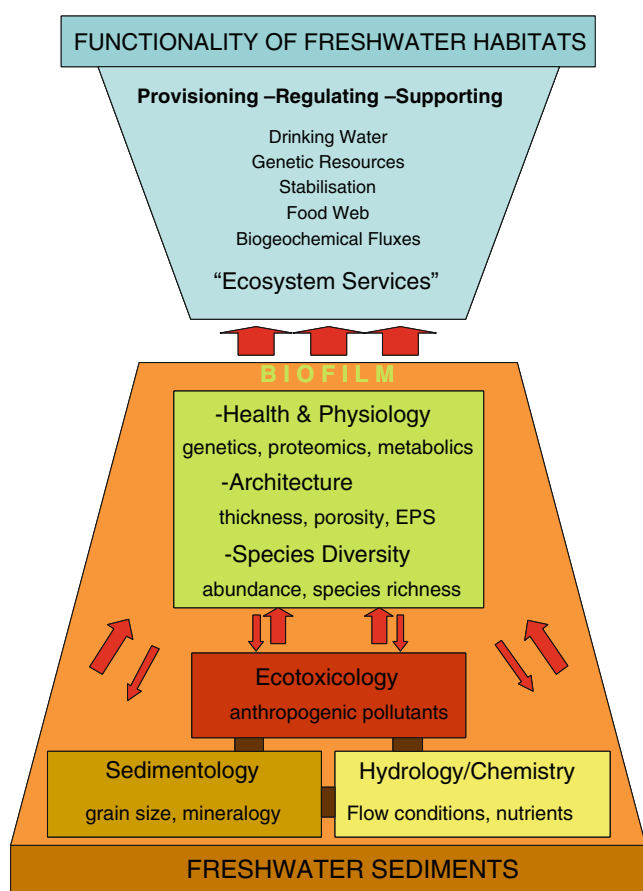


Fig. 1 The characteristics of freshwater sediments are determined by mutual dependent hydrological, sedimentological and abiotic conditions as well as the abundance and type of organisms. Anthropogenic pollutants might affect the inhabiting organisms and thus impair their important ecosystem services within and beyond the sedimentary environment

1.2 Ecotoxicology: the effects of sediment-associated pollutants—is a triad approach sufficient?

1.2.1 Bioassays complementing chemical tests

In the early 1970s, chemical investigations identified and quantified the source substances and their metabolites, but complete screenings of all chemicals are costly and time-

intensive. Moreover, the pure chemical approaches failed to give information on the bioavailability or on the synergistic and antagonistic effects of the pollutants for aquatic organisms (Chapman et al. 2002). Thus, about a decade later, a broad range of standardized bioassays were introduced to assess the possible hazardous effect of particulate matter and elutriate (Burton 1991). The bioassays included *in vivo* and *in vitro* tests at different levels of the aquatic food net (protozoa–metazoa). Various microbiological toxicity tests have been developed and validated for use in sediment risk assessment during the past 20 years (Ahlf et al. 1989; van Beelen 2003). The combination of such test systems on multiple levels of biological organization in biotest batteries allowed for the assessment of the different functional entities of biocoenoses. However, to sufficiently consider the bioavailability of sediment-bound contaminants, sediment contact bioassays were developed and applied (Hollert et al. 2003; Feiler et al. 2005). In addition, to account for more than the general mechanisms of acute and chronic toxicity, specific mechanism-based bioassays on mutagenicity, genotoxicity, as well as dioxin-like and oestrogen-like effects, were used (for a review, see Hallare et al. 2011).

The shortcomings of studies employing solely chemical target analyses were impressively demonstrated by parallel investigation of ecotoxicological effects. For instance, Keiter et al. (2008) demonstrated that the well-known and frequently measured priority dioxin-like target analytes (e.g. polycyclic aromatic hydrocarbons, polychlorinated biphenyls and polychlorinated dioxins and furans) could not explain 43–100% of the dioxin-like activity of sediments from the Danube River. The same holds true for endocrine disruptors where known endocrine active compounds, such as bisphenol A or alkylphenols, accounted for only 10–15% of the entire endocrine-disrupting potential measured in aquatic systems (Hollert et al. 2005).

1.2.2 Combining lines of evidence

These initial bilateral approaches verified that neither instrumental chemical trace analyses nor ecological test systems alone can comprehensively predict and explain the adverse effects of pollution on ecosystems. Combining chemistry and ecotoxicology has already led to the strategy of the “effect-directed analysis” that has a great potential for identifying novel substances of ecotoxicological concern and implementing them in regulatory frameworks and analytical monitoring programmes (Hecker and Hollert 2009). Applying this approach within the joint EU project Modelkey, numerous sediment-bound contaminants with strong toxicological potencies and occurrence in several catchment areas could be identified (Brack et al. 2007).

The need for the development and application of integrated approaches was also expressed earlier by Chapman (2000).

The Sediment Quality Triad, as proposed by Chapman (1990), is such an approach that simultaneously investigates chemistry, sediment toxicity and macrobenthic community structure. With these three “lines of evidence”, it was possible to draw conclusions based on the risk indicated by each endpoint and the confidence of the measurement. The resident benthic fauna is undoubtedly the most exposed to sediment contaminants, especially the microorganisms that are living in close contact with their microenvironment and are capable of reacting immediately to any disturbance. Thus, alterations in microbial communities have been used as an additional “line of evidence” in weight-of-evidence approaches (Kostanjsek et al. 2005). Chapman (2000) explicitly encouraged evolution of the triad concept and its components to account for site-specific features or to distinguish between chemical and non-chemical stressors. Thus, the design and evaluation of new risk assessment strategies should consider that anthropogenic influence cannot only be expressed in the structure but also in the functional potential of microbial sediment communities. For instance, the substrate utilisation pattern of indigenous microorganisms can support expensive chemical analyses as long as one regards the adaptations of the community to specific nutrient conditions. Moreover, the microbial assemblages provide, via their metabolic potential, a wide range of ecosystem services (e.g. self-purification, decomposition, mineralisation) that have not been linked so far to ecotoxicological investigations or triad risk assessment. Despite the growing numbers of publications in ecotoxicology, there are only few publications that are related to microbiology (Fig. 2). The same is true for hydrology, although it was proposed to include hydrodynamics as an additional “line of

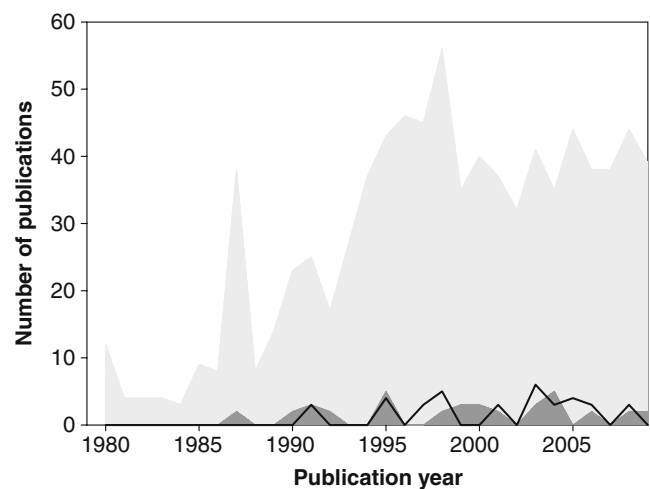


Fig. 2 Number of publications on contaminated sediments within the last three decades in the research area “ecotoxicology” (light grey), “ecotoxicology + microbiology” (black line) and “ecotoxicology + hydrology” (dark grey). Relative estimates based on search queries in ISI Web of Knowledge

evidence” to be able to estimate the environmental impact of the resuspension, transport and relocation of contaminated sediments (Chapman and Hollert 2006). However, field studies accounting for real aquatic exposure scenarios are rare—predominantly because the experimental link between physical resuspension of sediments and adverse ecotoxicological effects on aquatic organisms is not yet satisfactorily established (Wölz et al. 2009). However, regarding newly emerging challenges that are expected with climate change, knowledge of the possible implications of increasingly frequent and intense flood events is urgently required.

1.3 Microbiology: on its way to functional capabilities of microbial communities

1.3.1 *The interactions between pollutants and microbes*

Interactions with environmental pollution are mutual both on the level of individual bacterial cells and microbial populations as a whole. Reactions can occur passively, e.g. adsorption of metal ions by negative surface charge of the bacterial cell wall or the biofilm matrix that leads to a temporary immobilization of the pollutants. In contrast, chemical elements being actively (co-)transported through the bacterial cell envelop will subsequently interact with bacterial metabolism with ambiguous effects on bacterial physiology. Due to the vast diversity of metabolic pathways within the high numbers of bacterial cells in sediments, the exposure of pollutants does not inevitably lead to metabolic inhibition to harm cell growth or induce cell lysis. Instead, some species might be tolerant or even able to degrade and solubilize the pollutants, producing either intermediates or products ideally less toxic compared with the original substance.

Consequently, one important focus of microbiology is on decomposition, mineralization and bioremediation performed by bacteria and fungi, mainly in soils (about 3600 articles to date), but increasingly in the field of sediment science (about 1,100 articles to date), with continuously increasing topicality concerning human-made pollutants. The enhanced application of microbial remediation approaches is due to their efficiency and economic advantages (e.g. Pal and Paul 2008), but has some drawbacks regarding degradation times and predictability (e.g. Perelo 2010). In the context of this work, modes of actions and regulatory pathways were identified, and the global and specific expression patterns of genes as a response to the surrounding substrates were investigated (e.g. Hassanshahian et al. 2010). Biodegradation is an excellent example of the metabolic versatility of microbes since it can be rapidly performed by the induction and subsequent expression of existing genes. Moreover, bacterial communities have an excellent ability to adapt to new conditions, which

mainly relies on mutations and/or recombination of genetic information. Gaining a more in-depth understanding of the overall capabilities of microbial pathways, adaptation and degradation potential is crucial in order to develop methods and improved strategies to attenuate environmental pollution.

1.3.2 *Diversity and functionality of microbial ecosystems*

Environmental pollution might favour microbial species which are able to degrade the anthropogenic compounds by profiting twofold: from the additional nutrient sources and the suppression of potentially competitive species within the biocoenosis. Consequently, the presence of pollutants can act as a strong evolutionary factor within the microbial biocoenosis. Due to the horizontal gene transfer among a broad range of phylogenetic levels (which means bacterial gene transfer is not restricted to a certain species), and additionally fastened by mobile DNA elements, microbial biocoenosis is able to adapt quickly and flexibly to the input of pollutants entering the ecosystem. Possible loss or significant shifts in biodiversity are discussed with regards to the functional capabilities of the ecosystems (Solan et al. 2006). Although there has been evidence that some species (drivers or keystone species) are more in control than others (passengers/minor species; (Walker 1992), diversity as such seems to provide a general robustness in ecological resilience that might also be important for ensuring functionality under toxicant exposure (Solan et al. 2008). On the other hand, the conservation of a given functionality is often warranted by the flexibility of a microbial community with minority community members that may become dominant in a short period following significant perturbation (Marzorati et al. 2008), thus assuring a fast recovery from a stress condition such as the presence of pollutants (Fernandez et al. 2000). To ascertain how anthropogenic disturbance such as contamination input affects the microbial diversity of complex natural aquatic habitats is one of the most challenging objectives in environmental microbiology today (Lachmund et al. 2003). Since the characteristics of a single species and varying consortia of microorganisms will be decisive for several other direct and indirect processes in the environment, it is equally important to investigate the functionality in relation to shifts in species composition.

However, up to now, it was methodologically difficult to examine the diversity of microbial communities since the study of the ubiquitous heterotrophic bacterial component latterly relied on selective culture techniques and biased counting methods. Yet, with the help of molecular techniques such as fluorescence in situ hybridization (FISH), denaturing gradient gel electrophoresis (DGGE), terminal restriction fragment length polymorphism and

other contemporary approaches, the analysis of the taxonomic community structure of attached microbial communities has reached a new level (Manz 1999; Boeckelmann et al. 2000; Nocker et al. 2007; Schutte et al. 2008). For instance, the interpretation of the 16S rRNA gene molecular fingerprinting pattern by DGGE allows inferences to be made about the diversity and the grade of specialization in a community (functional organization), their rates of changes by detachment (dynamic value) and the carrying capacity of an ecosystem (range-weighted richness; Marzorati et al. 2008). Thus, conclusions can be drawn as to whether a particular environment is suitable or unsuitable (e.g. due to chemical exposure) for microbial settlement. Subsequent sequence classification of separated DGGE bands by FISH can reveal the affiliation of the observed pattern to different groups of bacteria (phylum to species level) and hence identify species that are not influenced, more tolerant or more sensitive to pollutants. Using molecular techniques, Näslund et al. (2008) could relate shifts in a microbial community to the presence of the antibiotic ciprofloxacin, along with significantly reduced mineralization rates of the contaminant pyrene within marine sediments. Similarly, microbial community shifts were detectable after nearby dredging of polluted sediments in the Baltic Sea (Edlund and Jansson 2006). There are also hints that pollutants such as heavy metals tend to affect bacterial physiology and their capability to grow on substrates more than their total genetic diversity. Thus, pollutant effects may be better visualized by culture-dependent methods such as plate counts in contrast to culture-independent approaches such as 16S DGGE (Ellis et al. 2003). Recently, metagenomic approaches (monitoring of genes) were introduced in order to identify variations in bacterial genetic diversity and bacterial members on different levels (domain, phylum, subphylum, orders, families, genera) and to unravel species complexity and their interactions within microbial ecosys-

tems (Raes and Bork 2008). Metagenomic sequencing along with metaproteomics (transcripts on protein level) and metabolomics (metabolites) could provide important insights into the community structure and also into the functional role of the microbes, and thus are promising tools to study the effects of toxicant exposure on complete microbial systems.

1.3.3 The significance of EPS

In recent years, it became apparent that the microbial secreted EPS embedding the microorganisms in what is called a “biofilm” (Fig. 3) have a major impact on the dynamics of sediments and their associated pollutants. Firstly, the first attachment of the microbes to a surface, and thus to a potential pollutant, is dominated by electrochemical forces (e.g. London dispersion forces) that are enhanced by, for example, EPS-complexed iron species (Gehrke et al. 1998); hence, EPS essentially mediate the contact between degrader and pollutant. Secondly, the developing EPS matrix consists of a conglomerate of polysaccharides, proteins, lipids, nucleic acids and humic substances that offer various and different binding sites depending on their anionic or cationic properties (Flemming and Wingender 2001). Thus, pollutants can be directly adsorbed and immobilized within the EPS matrix, influencing their bioavailability and degradation (Pal and Paul 2008). Thirdly, EPS are involved in the flocculation of suspended sediment material, as we know from wastewater research (Liu and Fang 2003). Such aggregates have been shown to be hot spots of microbial settlement (enhanced surface) and metabolic activities (mutual advantages for microbes due to cascade-like degradation, co-metabolism; Passow 2002; Wotton 2004). Finally, microbial polymers are known to enhance the binding between sediment grains, leading to an increase of the overall sediment stability

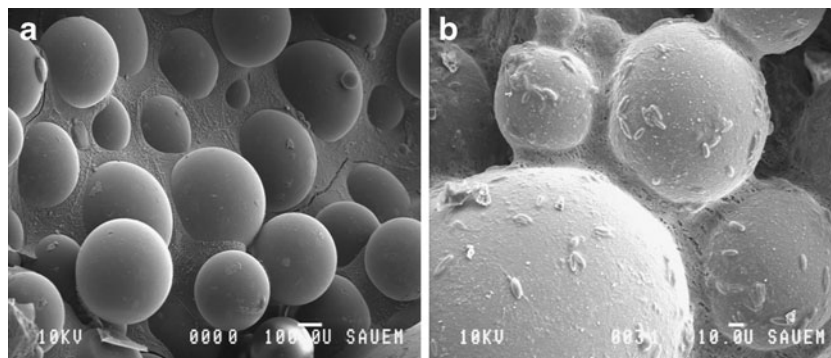


Fig. 3 Microbial benthic communities fulfil important ecosystem services (here biostabilisation) that are often closely coupled to their structure (here biofilm matrix) and metabolic activities (here EPS secretion). **a** Surface image of a glass bead layer without biofilm (control); frozen water forms a solid matrix surrounding the glass

beads. **b** Surface image of a glass bead layer with a mixed biofilm; clearly visible are the EPS matrix covering and connecting the glass beads plus single diatoms moving within the biofilm (source: Lubarsky, Paterson and Gerbersdorf, unpublished)

which can resist hydrodynamic forces (Underwood and Paterson 2003; see Fig. 3). Conglomerations of microbes to form film, mats or flocs consist of over 90% EPS (which in turn consist of more than 90% water; Flemming and Wingender 2010); thus, the extracellular polymers are the structural component in the ubiquitously occurring biofilms in natural or artificial habitats. Consequently, considerable research efforts on EPS highlight its importance for various fields such as ecology, biomedicine, biotechnology and industry (Vu et al. 2009). There is now a consensus that the functionality of EPS is largely influenced by its quantity and quality and that both depend strongly on the secreting species (Gerbersdorf et al. 2008b, 2009a) and their metabolic activities (Gerbersdorf et al. 2008a, 2009b). Anthropogenic pollutants can indirectly impact the EPS secretion pattern by affecting bacterial physiology or by inducing shifts in bacterial species composition. For the antibacterial compound triclosan, it was shown that the pollutant interfered with the quorum sensing signalling of Gram-negative bacteria (direct effect on EPS compounds that are strongly involved in cell communication); as a result, bacterial attachment, growth and the formation of biofilm were significantly hampered (Dobretsov et al. 2007). Thus, altering EPS secretion might significantly impair important functionalities of biofilm for the aquatic habitat such as sediment stabilisation (also see Section 2.3).

1.3.4 Micro-architecture and structure of biofilms

Shedding light on the diversity of micro-consortia, the functions of their encoded genes, and the EPS quantity and composition is an important step, but provides only scarce information about the resulting structural organization of biofilms and the closely linked functional capability of these communities up to the level of ecosystem services. Battin et al. (2003a, b) show that the biofilm structure was largely a result of complex biotic (growth, grazing) and abiotic (e.g. hydrodynamic, sedimentology, nutrients) conditions and influenced, in turn, mass transfer towards and within the biofilm (e.g. transient zone thickness, porosity), as well as the retention of particles (e.g. local and temporal change in hydrodynamic conditions). Moreover, the biofilm has a stabilizing effect on its surroundings which may even be enhanced by increasing particle deposition through filtration and adhesion, a classic self-energizing feedback mechanism (Larson et al. 2009). It is thus likely that the micro-architecture of the biofilm also significantly affects the deposition, incorporation and recycling of pollutants. On the other hand, the exposure of anthropogenic pollutants might impact biofilm structure and related functions, with unknown mutual influence. Nowadays, the development of advanced microscopy and spectroscopy techniques such as confocal laser scanning microscopy, scanning transmission

X-ray spectro-microscopy, atomic force measurements, nuclear magnetic resonance imaging, or Raman spectroscopy allows a highly resolved exploration of biofilm matrices and intermediate spaces as well as compositional mapping of macromolecule distributions (Dynes et al. 2006; Denkhaus et al. 2007; Vu et al. 2009). A review on the advanced imaging techniques has been recently published (Neu et al. 2010). As a first important step, the visualized structural components and spatial arrangements have been linked to microbial diversity (Manz et al. 1999, 2000). The ultimate goal is to relate diversity and structure to ecosystem services of the aquatic habitats and attribute changes to toxicant exposure.

1.3.5 Functional capabilities of microbial ecosystems

The functional responses of indigenous microbial communities in sediments have been assessed for over two decades by different enzymatic reactions (e.g. ammonification, nitrification, sulphate reduction, degradation of contaminants). Advanced test systems have been developed, such as the commercially available BIOLOG system that investigates the utilisation of carbon sources, representing the metabolic capacity of bacteria. Originally developed 20 years ago for pure cultures, BIOLOG plates were introduced by Garland and Mills (1991) for the assessment of bacterial communities in environmental samples. Despite some concerns about the preferential enrichment of (aerobic) culturable bacteria capable of growing in high nutrient environments, it could be shown that this method provides metabolic fingerprints from complex communities, expressed as a community-level physiological profile (Garland and Mills 1991) and allows for sufficient discrimination between different sediment sites (Garland 1999). This assessment of functional capability of microbes provides insights into the important processes occurring in one ecosystem, but has to be linked to the involved community members (Raes and Bork 2008). This has led to the application of combined methods such as in microautoradiography (MAR)-FISH, where the uptake of radioactively labelled substrates (metabolic activity) has been linked to FISH techniques (phylogenetic identification; Lee et al. 1999). Thereby, a specific advantage of rRNA-targeted oligonucleotides is that they can be designed on various phylogenetic levels ranging from domain to genus and species specificity, allowing a “top to bottom” hierarchical approach for the analysis of the microbiocoenosis. In another approach, stable isotope probing has been investigated in parallel with nucleic acid-based fingerprinting techniques (Radajewski et al. 2003), and the metabolic potential of bacteria has been related to microbial diversity (Manz et al. 2001). Ginige et al. (2004) used a combination of stable isotope probing, full-cycle rRNA analysis and

FISH–MAR to study a denitrifying microbial community. Future studies should combine descriptive (species identification), structural (micro-architecture) and functional aspects (e.g. metabolic activities such as secretion, degradation, conversion of substrates).

1.3.6 Ecosystem functions and their alterations

Recognizing functional capabilities of microbial assemblages in their aquatic habitat is an emerging theme in modern ecology (Paterson et al. 2008). Still, microbial studies rarely include services on an ecosystem level (e.g. impact on biogeochemical fluxes, food web, stabilisation of sediments), although this is of ever-increasing importance regarding the growing anthropogenic influence on aquatic habitats and their inhabitants. Thereby, one big challenge will be to recognize potential pollutant effects on ecosystem functions of microbial communities under varying environmental conditions, whether due to natural short-term rhythms (e.g. sporadic–weather, regular–daily, tidal, seasonal cycles) or long-term climate changes (e.g. temperature, CO₂ availability, pH value). To that purpose, the cooperation between microbiology and ecotoxicology needs to be strengthened. It is further necessary to distinguish between pollutant-induced effects on microbial ecosystems and habitat morphology-induced alterations in the field (e.g. artificial barriers, straightened river courses; Chapman 2000). Habitat models (Noak and Wieprecht 2010; Tuhtan et al. 2010) based on physical parameters are able to quantify the influence on habitat quality for, for example, fishes or macrozoobenthos, due to changes in the morphological regime in river systems where the goal is to evaluate and distinguish the superimposition of pollutant-induced effects. In this respect, the hydrodynamic conditions play a crucial role for morphological alterations, pollutant exposure, as well as for the settlement, growth and metabolism of microbes. It is thus important to include knowledge on hydrology and geomorphology as provided by engineering science. The fundamental challenge is to cross interdisciplinary boundaries to come to a conceptual understanding on the dynamics and effects of sediment-associated pollutants.

1.4 Engineering science: recognizing biological influence on the dynamics of sediment and associated pollutants

1.4.1 The importance of sediment dynamics

Knowledge of sediment transport processes has been important in many fields such as sedimentary geology, geomorphology, and civil and environmental engineering for a long time. In rivers, it is crucial to determine where and to what extent erosion and deposition processes may

take place in order to ensure that flood control projects, navigable waterways, hydropower production and nature reserves are properly maintained. The physical functionality of a river depends on its habitat for aquatic organisms as well as its landscape. Thus, sediment stability is one of the central variables in predicting sediment transport and dynamics within a river, along with the prevailing hydrodynamic forces. Whilst it is possible to predict the bed load transport of granular material (sand, gravel) under certain critical bed shear stresses up to a certain order of accuracy, the complexity of physical properties in cohesive sediments reduces the precision of the results gained by standard approaches. Additionally, biological interactions (e.g. bio-turbation, attachment of organism) and chemical reactions (e.g. surface loadings of organisms, organic material, silt as well clay particles) in cohesive sediments still preclude the definition of a general, physically valid theory for the prediction of erosion of cohesive sediments (Black et al. 2002; Haag and Westrich 2002).

1.4.2 Sediment stability

Traditionally, studies attempting to elucidate the mechanisms governing the stability of cohesive sediments concentrated on physicochemical sediment properties such as bulk density, particle sizes, mineralogy and organic carbon (Jepsen et al. 1997; Ravisangar et al. 2001; McNeil and Lick 2004). There is now a consensus that biology, and in particular organic material such as EPS, plays a crucial role in sediment stability since it acts like a “glue” by permeating the pore space between the sediment grains and enhancing their interparticle forces (Underwood and Paterson 2003; Gerbersdorf et al. 2008b, 2009a). Today, we have a much better understanding of the importance of the interactions between physics, chemistry and biology in influencing the stability of cohesive sediments (Gerbersdorf et al. 2005, 2007, 2008a, 2009b). Still, whilst the number of publications on biostabilisation is steadily increasing, academic sediment transport models, as well as commercially available software, consider the process of incipient motion as primarily being governed via equations describing the physical processes; if biostabilisation is considered at all, it is included by empirical relations.

The critical condition for the incipient motion of sediment is normally measured against the critical bed shear stress, τ_c . When non-dimensionalized by fluid and sediment parameters, it is referred to as the critical Shields parameter $\Theta_c = \tau_c / (\rho_s - \rho_w) g d$ (where d is sediment particle diameter, g is gravitational acceleration, and ρ_s and ρ_w are the densities of fluid and sediment, respectively). The Shields ratio is limited to uniform sediments; however, the Shields diagram remains the most widely used criterion at present, even for the calculation of non-uniform sediment

motion (Shields 1936). This is despite the legendary inconsistencies and misconceptions (Buffington 1999) as well as experimental discrepancies (Shvidchenko and Pender 2000). In many models, it is common to introduce a correction factor to account for specific effects, such as for the hiding and exposure mechanisms of non-uniform sediment transport (Belleudy and Sogreah 2000; Wu et al. 2000, 2003). Similarly, attempts have been made to include stabilizing and destabilizing effects of organisms by means of empirical modification of the formulation for the critical bed shear stress and the erosion rate. For instance, terms to describe the effects of microphytobenthos and grazers on sediment erosion in a coastal region (Westerschelde, the Netherlands) have been proposed by, for example, Holzhauser (2003) and Paarlberg et al. (2005) using the data sets of Widdows et al. (2000) and Widdows and Brinsley (2002). Borsie (2006) implemented a parameterization of the influence of biological activity on sediment strength parameters (i.e. critical bed shear stress and erosion rate) in a numerical model applying the process-based model Delft3D. Since the empirical approaches for biological processes are based on measurements in the Wadden Sea, the validity of the results is restricted to the investigated area.

In summary, there are some attempts to describe biological influences on sediment dynamics as forcing variables to be implemented in sediment transport models. This is certainly important to better judge the expected erosion or deposition zones (concerning mass/volume as well as spatial distribution), leading to improved management measures (concerning water quality and rivers). Yet none of the approaches is transferable or universally valid. The identified relations are still dependent on the temporal and hydrological conditions relating to site-specific conditions. Moreover, interactions and feedback mechanisms between physical and biological processes are not yet considered in mathematical sediment transport models (Le Hir et al. 2007).

1.4.3 Floc characteristics

Pioneering research has stated that microbial organisms and EPS also influence post-entrainment flocculation because the organic matrix largely affects the characteristics of the sediment flocs (Liss et al. 1996; Perkins et al. 2004), along with the physical properties of the sediment and the depositional history (Lau and Droppo 2000; Stone et al. 2008). Once eroded, the characteristics of the suspended sediment (i.e. effective particle size, shape, density, strength) alters the path length of lateral transport as well as the downwards flux (Droppo 2001, 2004), influencing sediment dynamics in rivers. However, sediment transport models are mainly based on the “primary” particle size

classes of the sediments, determined by sieving after removing the organic matter. Knowing the particle size distribution, Partheniades (1965) introduced the influence of adhesive and cohesive forces by the parameter M_{ero} , which describes erosion rates related to the critical shear stress. Parchure and Mehta (1985) deduced an approach which is primarily applicable for soft soils and gives an erosion rate which is considerably steeper as compared with Partheniades (1965). These types of functions are widely used in models predicting sediment transport of fines, yet one cannot distinguish between biological and non-biological effects on cohesion. As these represent aggregate parameters derived from very site-specific situations, this stands in sharp contrast to the sophisticated modelling of the hydrodynamics where a highly resolved 2D or even 3D distribution of water depth, flow velocity and shear stress can be calculated. Deriving assumptions of transport behaviour of suspended load or even transport rates remains difficult when including binding effects that are only marginally known or quantifiable since the biological contribution varies significantly both spatially and temporally (Underwood and Paterson 2003). In recognizing this issue, the interactions between sediment particles (e.g. changed adhesion and cohesion) mediated by microbially produced EPS need to be fully understood in order to successfully implement them in sediment transport modelling.

1.4.4 Absorption/desorption of pollutants

Anthropogenic pollutants are mainly associated with the fine sediment fractions (<63 μm) and their flocs, coupling closely the fate of cohesive sediments and pollutants. Due to water treatment in the last few decades, the load and accumulation of heavy metals and organic pollutants has been drastically decreased, but historic sediments still store high quantities of pollutants which can be easily remobilized by flood events that exceed the critical erosion values (Fig. 4). Modern chemicals such as pharmaceuticals or perfluorinated compounds remain largely unknown regarding their accumulation rates in sediments and remobilisation behaviour, but they are associated with younger sediments that may be eroded more easily under relatively low discharges due to a lower degree of compaction. Ultimately, resuspension of polluted cohesive sediments leads to the exposure and elevated toxicity of formerly immobilized pollutants (e.g. Calmano et al. 1993), whilst subsequent sediment deposition can remove them from the oxic water column. These episodic resuspension and deposition cycles of fine sediments or flocs might also stimulate microbial activity and are thus important for the recycling of pollutants in the aquatic environment. The impact of suspended sediments on biogeochemical cycles,

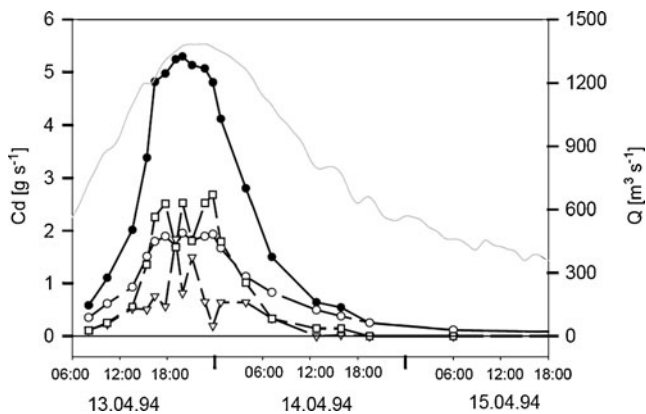


Fig. 4 Particulate cadmium (Cd) load bound to suspended sediments in the afflux (open circles), the efflux (black circles), within eroded new sediments (open triangles) and within eroded aged sediments (open squares) relative to the hydrograph (grey line) of HQ₅ in River Neckar (Germany) with Q (discharge) $> 1100 \text{ m}^3 \text{ s}^{-1}$ (mean discharge $88 \text{ m}^3 \text{ s}^{-1}$). Characteristic is the hysteresis behaviour between discharge and suspended Cd concentration with higher loads on the rising limb as compared with the falling limb of the hydrograph. Still, the sediments contributed more than 50% of the total Cd load, with the highest source due to former consolidated aged sediments. From Westrich et al. (2000)

in particular trace metals and hydrophobic organic micro-pollutants, has been addressed by quantifying and modelling water–particle interactions such as adsorption–desorption and precipitation–dissolution processes using partitioning or distribution coefficients ($K_D = P/C$, where K_D is the relation of chemical sorbed to suspended particle to that chemical dissolved in water; Turner and Millward 2002). It is well known that the distribution coefficients differ with varying conditions of salinity, pH, redox conditions or concentration of organic material, but still the adsorption isotherm of one pollutant is empirically established for one particular condition in the laboratory. Transferring this knowledge into the field is difficult since one cannot account for the expected temporal and spatial variations of the most influential parameters on partitioning coefficients (Karnahl 2009). The ultimate goal would be to predict distribution coefficients from theoretical principles and thermodynamic constants (Turner and Millward 2002).

In order to derive models that are more reliable for sediment dynamics and associated pollutants, the binding force of microorganism and their organic matrix should be determined and implemented. By identifying biology as important for the erosion, transport, deposition, consolidation cycle of sediments, one could predict that pollutants affect sediment stability and post-entrainment flocculation by being influential on the sediment-inhabiting organisms. To unravel these mutual dependencies and feedback mechanisms, a close cooperation between engineering and biological sciences (in particular microbiology), as well as ecotoxicology, is warranted.

2 Addressing pollutants in aquatic habitats: bilateral efforts to bridge the gaps

2.1 Ecotoxicology and microbiology: a strong alliance to address bioavailability of pollutants, induced community shifts and bioremediation

Since the early 1980s, ecotoxicological research on sediment-associated pollutants using standardized tests has been mostly based on benthic macroorganisms (e.g. *Lumbriculus variegatus*; Dermott and Munawar 1992; Chironomid larvae: OECD 218 2004). Yet there are single-species toxicity tests using microorganisms such as *Arthrobacter globiformis* (DIN 38412–48 2002), *Bacillus cereus* (Liss and Ahlf 1997) or *Vibrio fischeri*, bacterial luminescence inhibition test (ISO 11348-1 1998; Ahlf et al. 2002). Two bacterial test systems have been developed for the detection of genotoxic and mutagenic potentials in freshwater: the umu test (Oda et al. 1985), based on DNA damage followed by induction of the DNA repair gene umuC, and the Ames test (Ames et al. 1975), verifying the mutagen's ability to cause a reversion in histidine mutants to grow on a histidine-free medium. Today, this agar plate incorporation assay is the most widely used mutagenicity test system worldwide. However, these test systems have a limited predictive power regarding the complex effect pattern of pollutants in situ on a multitude of organisms and their interactions (for a review, see Burton 1991). To overcome these limitations of single-organism test systems, multispecies assays have been introduced, such as the microbial assay for risk assessment that employs ten relevant bacterial species of taxonomically diverse affiliation and one yeast species (Gabrielson et al. 2003; Wadhia and Dando 2009). This conceptual design was shown to be capable of detecting (a) acute effects of chemicals, (b) can be used for toxicant fingerprinting and (c) was evaluated for its potential use in the investigation of environmental samples such as water, soils, sediments and effluents in the context of the European Water Framework Directive (WFD 2000). Another promising tool to assess community-level alterations as a potential consequence of exposure to pollutants is the measurement of pollution-induced community tolerance. Due to the successive inhibition of sensitive species by pollutants, the remaining community has an elevated tolerance over unaltered sediment-borne communities. In comparisons to controls, the proportion of the autochthonous microflora affected by pollution can be determined to explore the potential environmental hazard (McClellan et al. 2008). Ecotoxicology and microbiology are also strongly coupled in testing new bioremediation techniques for contaminated soils and sediments where ecotoxicological effects have been related to shifts in microbial communities (e.g. Kostanjsek et al. 2005; Jonker

et al. 2009). Another important topic is the microbiological analysis following ecotoxicity studies to isolate the appropriate microorganisms capable of coping with and degrading the prevailing toxicants (Dercova et al. 2008).

The implementation of the European Water Framework Directive (WFD 2000) has stipulated the characterization of microbial communities in river sediments to assess the ecological status for European river basin management. Presumably, future ecotoxicological studies will account for more structural and metabolic functions of microbial communities. Combined ecotoxicology/microbiology laboratory studies will tend to present a potential risk assessment for the aquatic habitats rather than reflecting on real exposure scenarios under consideration of in situ sediment dynamics. Thus, sound investigations of empirically determined sediment stability with biological influence under various hydrodynamic conditions are needed to predict erosion risks and help put the laboratory-observed potential risks into perspective.

2.2 Ecotoxicology and engineering science: from potential to realistic risk assessment of sediment-associated pollutants

To date, the possible ecotoxicological effects of contaminants bound to suspended material have been scarcely related to in situ sediment dynamics. In the last 5 years, several studies have addressed the ecotoxicological impact after major flood events such as the Elbe River floods of 2002 (Grote et al. 2005; Oetken et al. 2005). It became evident that the flood caused elevated short-term and long-term toxic effects in different bioassays (Hsu et al. 2007). Kammann et al. (2005) reported on significantly elevated enzyme 7-ethoxyresorufin-*O*-deethylase activities in the livers of dab (*Limanda limanda*) from the German Bight (North Sea) as compared with monitoring data from 1995 to 2003. Cellular changes in livers from flounder (*Platichthys flesus* L.) and digestive glands of blue mussels (*Mytilus edulis*) could be determined in the Elbe River estuary and the Wadden Sea 5 months after the flood disaster (Einsporn et al. 2005). Hence, these alarming findings support the hypothesis that remobilized sediments can (a) induce severe ecotoxicological effects on organisms in the aquatic habitat and (b) reach beyond the freshwater system to affect adjacent floodplains and wetlands, and also brackish and marine systems (Schwartz et al. 2006). During the Elbe River flood in 2002, this effect could be observed explicitly; in the course of the River Elbe from upstream (Czech Republic border) to downstream (Geesacht, Germany), a decreasing gradient of pollutants in the main river was monitored whilst the majority of suspended particles was deposited on the floodplains. Remobilized sediments originated from the main river could be detected, and an exceedance of critical values for Cd, Hg and Zn was

measured. Additionally, high oxygen attrition caused fish mortality in the tributaries which reached alarming proportions (IKzSdE 2004). Consequently, ecosystem services on different levels are affected such as livestock breeding on wetlands, fishery, recreational activities at the coast and rivers and, last but not least, drinking water supply (Maier et al. 2005; Kuehlers et al. 2009).

Wölz et al. (2008) related the measured ecotoxicological effects (e.g. Aryl hydrocarbon receptor AhR agonist activities of suspended particulate matter) to the flow rate, indicating a flood-dependent increase of toxicity culminating at the peak of flow in the rivers Neckar and Rhine. That confirmed the measurements of Westrich et al. (2000) where the particular load of heavy metals increased significantly during a flood event in the Neckar River, mainly due to the erosion of aged contaminated sediments (see Fig. 4). More recently, effect-directed analysis was used to identify the pollutant source for the activity of the AhR agonists (Wölz et al. 2010). Via calculation of chemical toxicity equivalent concentrations (chem-TEQ values), the EPA-polycyclic aromatic hydrocarbons (PAHs) explained between 5% and 58% of crude extract bio-TEQs from both rivers. In conclusion, these studies gave a first idea of the ecotoxicological hazard of flood events using in vitro approaches and relating the results to the amount of remobilized sediments and to the hydrological conditions.

In order to come to a reliable erosion risk assessment of sediment-bound pollutants, pioneer studies linked empirically derived data on sediment stability, modelled hydrodynamic scenarios (based on cross-section and morphology) and vertical depth profiles of sediment characteristics and pollutant concentrations (Haag et al. 2001). Adding bioassays to test the bioavailability and toxicity of the released pollutants, initial investigations combined ecotoxicology and engineering science (Hollert et al. 2000). Since then, the need to merge the knowledge of both disciplines has been verbalized, but not implemented in science yet (Babut et al. 2007; Hollert et al. 2007).

The pathfinder project “Floodsearch” (RWTH, Aachen, Germany) is based on this idea to relate the mobilization of sediments to the release of associated pollutants and hazardous effects on biota during flood events. Within the first interdisciplinary proof-of-concept study, a suitable methodology was presented to expose rainbow trout (*Oncorhynchus mykiss*) to resuspended artificial sediments spiked with PAH using an annular flume (Brinkmann et al. 2010). The first results are encouraging in demonstrating the uptake and conversion of PAHs by fish, as well as indicating their genotoxic effects on the rainbow trout even after a short exposure of 5 days. The observations within the Floodsearch project confirm the necessity to combine ecotoxicological, hydraulic and sedimentological processes for an actual pollutant exposure risk assessment, preferably

extended by the inclusion of microbiology in a “triad plus x” approach. Detailed investigations of the microbial communities involved will help elucidate feedback mechanisms with the pollutants and how this affects the microbial ecosystem services of freshwater sediments.

In summary, whilst it is valuable to learn more about potential ecotoxicological hazards of anthropogenic pollutants for the aquatic organisms, it is equally important to know about (a) the probability at which the release of sediment-bound pollutants might happen and (b) the direct effects of remobilized pollutants on organisms.

2.3 Microbiology and engineering science: the discovery of mutual dependency

The release and bioavailability of contaminants is strongly coupled to sediment dynamics in aquatic habitats. The growing knowledge of the erosion resistance of sediments in both engineering science and biology originally followed parallel routes, without much consideration of each other. Less than a decade ago, the awareness grew that only multidisciplinary studies will lead to a more realistic and, thus, better understanding of the dynamics of natural cohesive sediments with their inherent physical and biological complexity (Black et al. 2002). There is virtually no fine sediment devoid of biota (Riding and Amrawik 2000), and the inhabiting life forms and their surrounding sedimentary environment will constantly influence each other to render a higher resilience towards external forcing (Stal 2010). The first studies to take this on board considered a selected range of biological and sedimentological properties (de Brouwer et al. 2000; Paterson et al. 2000; Haag et al. 2001; Haag and Westrich 2002), followed by more comprehensive approaches (Gerbersdorf et al. 2005, 2007, 2008a, 2009b). Significant relations between microalgal biomass, bacterial cell numbers, EPS carbohydrates, EPS proteins, total organic matter, cation exchange capacity and grain sizes revealed their interaction in influencing sediment stability (Gerbersdorf et al. 2005, 2007, 2008a).

Fine grain sizes, organism surfaces and the organic matrix (EPS and humic substances) offer various binding sites for pollutants that will eventually lead to their immobilization within the sediments. The situation is very different after an erosion event where the exposure to the oxic environment might result in a release and renewed bioavailability of the pollutants. Undoubtedly, the stability of sediment is a crucial variable in the dynamics of pollutants, but pollutants also affect the binding forces between the sediment particles and thus the erosion resistance of bed material. Numerous studies indicated negative effects of the pollutants on the most prominent sediment stabilizer, the biofilms, by reducing their biomass, growth rate and metabolic activity (Lawrence et al. 2009; Ricart et al. 2010). Whilst some authors claim that the

exposure to pollutants enhances the EPS production of the biofilm to serve as a protection matrix (e.g. under exposure to Cr(VI); Priester et al. 2006), others report on lower EPS concentrations as a result of harmed metabolic processes and growth (e.g. exposure to the herbicide 2,4-dichlorophenoxyacetic acid and to petrol hydrocarbons such as benzene; benzene, toluene, xylene; and gasoline; Onbasli and Aslim 2009). Evidently, the EPS secretion is crucial for the interparticle forces in the sediment and thus influences the dynamics of sediments and associated pollutants. This has inspired our recent work where it could be shown that triclosan, a personal care product ingredient, affects biofilm growth, resulting in significantly lower EPS secretion, reducing sediment stability. Increasing sediment vulnerability to erosion and subsequent enhanced release of sediment-bound pollutants could again result in a negative feedback mechanism on biofilm growth, pollutant bioavailability and pollutant degradation. This is an excellent example of the importance to merge the knowledge of microbiology and engineering, instead of polarizing the debate into “physics versus biology” (Black et al. 2002), and the necessity to consider more than descriptive (here microbial biomass and composition) and functional (here EPS secretion) variables to learn about higher ecosystem functions (here sediment stability). Still, these studies should ideally be combined with investigations on acute and subtle toxicity of the pollutants on microbial communities in order to predict short- and long-term effects on microbial ecosystem functions. Crossing disciplinary boundaries has already led to a better conceptual understanding of sediment dynamics, but the same is now required for the fate of sediment-associated pollutants in the aquatic environment.

3 Conclusions and future directions

Freshwater sediments are highly complex ecosystems where geomorphology and biology are strongly linked in a nonlinear way; this feedback between the sedimentary habitat and the inhabiting organism is a fundamental link in the ecosystem engineering paradigm (Darby 2010). The sediment communities (e.g. biofilm) ensure important functionalities of the freshwater habitats by providing important ecosystem services such as provisioning (e.g. clean drinking water, food); regulating (e.g. detoxification, self-purification); and supporting (e.g. biogeochemical turnover, biostabilisation) functions. The environmental impacts of anthropogenic sediment-associated pollutants—mostly heavy metals and organic pollutants from the past industrial era, and also increasingly modern persistent chemicals such as pharmaceutical and personal care products—became evident in aquatic habitats and beyond (i.e. adjacent floodplains, marine environment) during

catastrophic events such as the Elbe River floods of 2002. Hence, considerable research effort has been undertaken to address this emerging issue.

However, most of the research of the involved disciplines has a singular view of contaminated freshwater sediments. In ecotoxicology, a broad range of standardized bioassays has been developed that were combined on multiple levels of biological organization and accounted for acute, chronic and specific mechanism-based toxicity. In this context, the combination of chemistry, ecology and ecotoxicology (e.g. the triad approach) has been an important step towards a more comprehensive ecological risk assessment. In microbiology, modern techniques increasingly allowed the characterization of the microbial community to reveal the functions of their encoded genes, their structural organization, their metabolic activities and their ecosystem services in order to provide information on the pollutant mode of actions on microbial ecosystems. Modelling of sediment transport and adsorption/desorption of pollutants in river systems has been further advanced in engineering science as an important tool in integrated water and sediment management, but the implementation of biological variables is still missing. There have also been some bilateral efforts concerning the potential ecotoxicological risk for aquatic organisms in relation to in situ sediment dynamics (i.e. ecotoxicology and engineering) to enhance ecotoxicological test systems and bioremediation potential (i.e. microbiology and ecotoxicology) or unravel pollutant effects on microbial communities and their sediment stabilisation potential as one important ecosystem function (i.e. microbiology and engineering science). So far, no study combines the expertise on toxic pollutant effects for the exposed organisms with knowledge on shifts in microbial assemblages, their structure, and functions and relates this to actual sediment and pollutant dynamics. One ultimate goal is to merge the expertise from the natural sciences to get comprehensive insights into the functions of these complex sedimentary ecosystems and their reaction to disturbances such as pollutant exposure. This knowledge is important for improving both the theoretical understanding and for practical environmental management. We thus propose a “triad plus x” approach involving the interdisciplinary skills of ecotoxicology, micro- and molecular biology, as well as engineering science to come to a more realistic risk assessment of contaminated sediments in freshwaters and beyond.

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