

Toxicity and risk assessment of fluvial sediments and suspended matter: a short overview of past and recent developments in sediment toxicity assessment

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Abstract *Background* In recent years, sediments have become a central topic of scientific and public discussion as an important factor for determining water quality. While the quality of surface waters in Germany has significantly improved during the past years, highly contaminated sediments still create a considerable threat to the quality of several European catchment areas.

Main features Here, we summarize different concepts and methods for the assessment of sediment quality and report on some novel integrative test methods for assessing sediment toxicity including contaminant re-mobilization during simulated re-suspension events.

Results and discussion Currently, different approaches for assessment of sediment pollution exist. While instrumental chemical analyses are not suitable to accurately describe sediment toxicity, combinations of biological and chemical test procedures and integrated approaches, for example weight-of-evidence studies and effect-directed analysis (EDA), have the potential to identify key contaminants. Inter-disciplinary studies combining hydrodynamic and

toxicological aspects coupled to real exposure of aquatic organisms to contaminants are currently being developed.

Conclusions Monitoring and assessment of sediment quality are of increasing importance, not only for national legislation but also for the implementation of the European Water Framework Directive (WFD). Integrated approaches for the determination of sediment stability play a key role in the appropriate sediment-monitoring strategies.

Keywords Sediment mobility · Sediments · Suspended particulate matter · Toxicity · Weight-of-evidence approaches

Toxizität und Risk Assessment fluvialer Sedimente und Schwebstoffe: Eine kurze Übersicht bisheriger und neuerer Entwicklungen

Zusammenfassung *Hintergrund* Sedimente sind als wichtiger Faktor der Gewässerqualität in den vergangenen Jahren verstärkt in die wissenschaftliche und öffentliche Diskussion getreten. Während sich die Wasserqualität in jüngster Zeit deutlich verbessert hat, werden die zum Teil hoch kontaminierten Sedimente in vielen europäischen Einzugsgebieten die Gewässerqualität noch für viele Jahrzehnte nachhaltig beeinflussen.

Schwerpunkte Dieser Artikel gibt einen kurzen Einblick in die aktuellen Entwicklungen und konzeptionellen Ansätze zur Bewertung der Toxizität und der Umweltrisiken von Sedimenten und Schwebstoffen. Des Weiteren wird ein neuer Ansatz zur Untersuchung der Sedimenttoxizität unter Berücksichtigung der Schadstoffremobilisierung während simulierter Resuspensionsereignisse vorgestellt.

Ergebnisse und Diskussion Derzeit existieren viele verschiedene Ansätze zur Bewertung der Sedimentbelastung: Während instrumentelle chemische Analytik für sich nicht

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in der Lage ist, die Toxizität von Sedimenten hinreichend genau zu beschreiben, können Kombinationen aus biologischen und chemischen Testverfahren und integrierte Ansätze, z.B. Weight-of-Evidence-Studien und effektdirigierte Analyse (EDA), zur Identifikation der Schlüsselkontaminanten beitragen. Methoden zur Umweltrisikobewertung kontaminierter Sedimente, z.B. die Simulation von Erosions- und Sedimentationsprozessen bei gekoppelter Exposition aquatischer Organismen, werden derzeit entwickelt.

Schlussfolgerungen Überwachung und Bewertung der Sedimentqualität kommen nicht nur im Rahmen nationalen Rechts, sondern auch bei der Umsetzung der europäischen Wasserrahmenrichtlinie (WRRL) große Bedeutung zu. Um die daraus resultierenden Ansprüche an Sedimentuntersuchungen zu erfüllen, sind integrierte Ansätze und die Berücksichtigung der Sedimentstabilität unabdingbar.

Schlüsselwörter Schwebstoffe · Sedimente · Sedimentmobilität · Toxizität · Weight-of-Evidence

1 Sediment quality assessment

In response to the emerging issue of sediment contamination, and in order to protect aquatic life, comprehensive methods and approaches for identifying and assessing sediment contamination have been developed since the early 1970s (Fig. 1). The very first approach was to merely employ chemical analyses. Though being a highly effective analytical tool that is capable of detecting and quantifying both source substances and their metabolites, the chemical

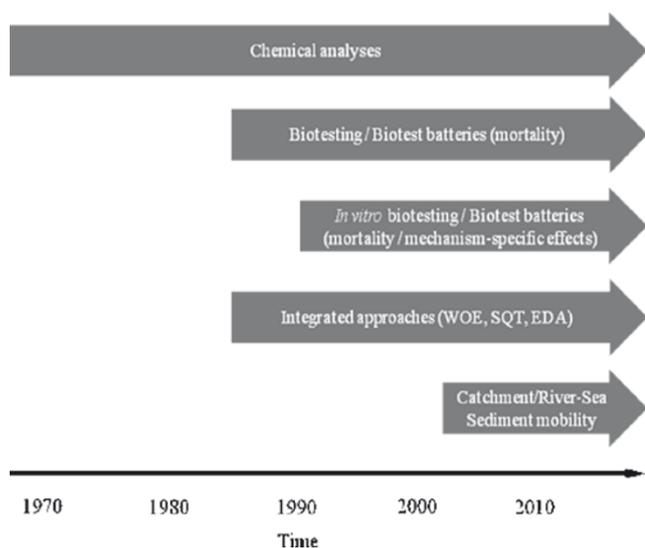


Fig. 1 Historical development of sediment risk assessment by continuous integration of additional approaches, e.g. biotesting, weight-of-evidence approaches (WOE), the sediment quality triad (SQT) or effect-directed analysis (EDA)

analysis approach fails to provide information on the actual bioaccessibility and on the biological activities of toxicants (Chapman et al. 2002). Likewise, chemical data do not provide information concerning synergistic and antagonistic potential in sediments. The presence of numbers of chemicals in water and sediments also obstructs any complete chemical screening for time, effort, and cost considerations. About a decade later, a battery of *in vitro* bioassays was developed and adapted for the evaluation of sediments, soils, and suspended particulate matter. In order to ensure that the actual bioaccessibility of sediment contaminants is sufficiently considered, bioassays using direct sediment contact exposure were applied in different exposure scenarios (Hollert et al. 2003b). However, since sediment contact assays with whole organisms usually provide only acute (mortality) and chronic toxicity data, it has become necessary to utilize more specific mechanism-based bioassays (e.g. mutagenic, genotoxic, teratogenic, dioxin-like, and estrogen-like responses, Hollert et al. 2009). In this way, research on sediment toxicity can gain more comprehensive insights into the ecotoxicological hazard potential of sediments.

For the assessment of aquatic sediments with respect to their adverse effects on ecosystems, however, neither biotests nor chemical-analytical techniques alone are sufficient. Thus, a need for integrated and hierarchical approaches combining chemical, ecotoxicological, and ecological information has been proposed (Ahlf et al. 2002; Burton 1991; Chapman 2000; Heise and Ahlf 2002). As a consequence, Chapman (1990) introduced the Sediment Quality Triad (SQT) approach which simultaneously investigates sediment chemistry, sediment toxicity, and sediment ecology (Fig. 2a, e.g. modifications of benthic community structure). These three lines-of-evidence, considered as the original components of the SQT, are integrated to reach conclusions based on the degree of risk indicated by each measurement and the confidence in each measurement. In the context of ecological risk assessment (ERA) of contaminated sediments, the SQT provides a weight-of-evidence framework (Chapman 2000) comprising a determination related to possible ecological impacts based on multiple lines-of-evidence. Recently, it has been proposed to include hydrodynamics as an additional line-of-evidence to assess the environmental impact of contaminated sediments (Chapman and Hollert 2006). Since then, the combination of hydrodynamics and ecotoxicology, as a novel approach, has become a promising field in environmental research (Hollert et al. 2007; Wölz et al. 2009).

Another example of a powerful blend of biological testing and instrumental chemical analyses is the concept of effect or bioassay-directed fractionation (Fig. 2b). This sequential procedure combines physico-chemical fractionation, biological testing, and instrumental chemical analyses (Brack 2003; Hecker and Hollert 2009) that ultimately leads

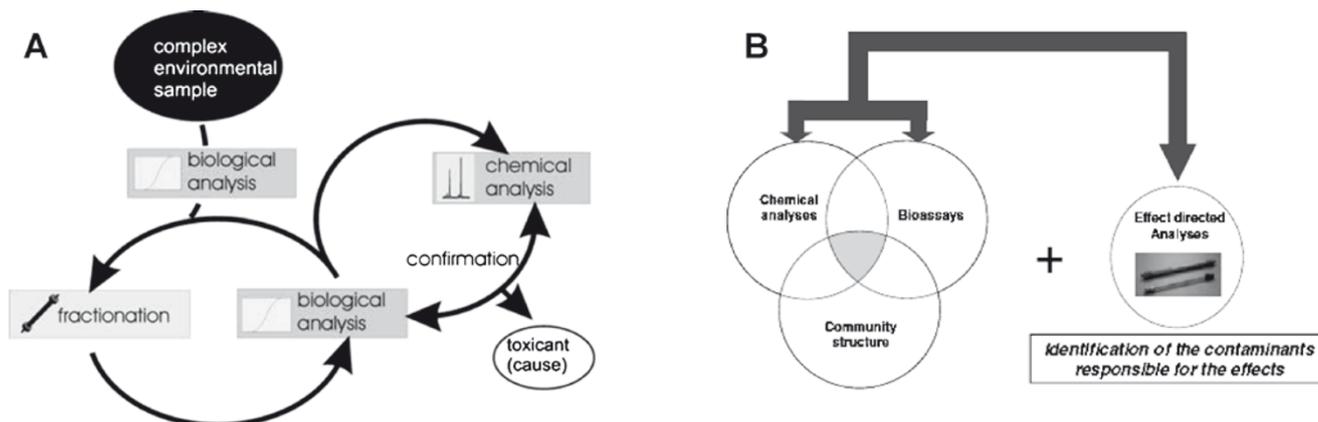


Fig. 2 (a) Methodology of bioassay-directed fractionation of toxicants from complex environmental samples (redrawn from Brack 2003) and (b) conceptual combination of the Sediment Quality Triad (SQT) according to Chapman et al. (1990) and bioassay-directed fractionation techniques (Brack 2003; Hollert et al. 2009) to identify single substances causing effects in the environment (redrawn from Hecker and Hollert 2009)

to the identification of biologically active and, thus, problematic substance classes or even single substances. Bioassay-directed fractionation studies possess high potential for the causal analysis of complex environmental problems, especially those using acute as well as mechanism-specific biological endpoints in combination with in situ investigations in the context of weight-of-evidence studies. This will then provide important inputs and appropriate action plans for managing highly contaminated rivers and streams (Hollert et al. 2009).

2 Sediment mobility and flood risk assessment

Fluvial sediments mainly originate from the erosion of the terrestrial surface or the open channel itself and are transported in a coastal direction, ultimately leading to sedimentation in the estuary and the oceans. The sediment bed can be stable over remarkably long periods. However, increased discharge, for example after heavy rainfall, can lead to increased bed shear stress allowing the re-suspension of sediments and the consequent distribution into adjacent floodplains and flood retention areas. The various inorganic and organic constituents of sediments (e. g. clay minerals, humic substances, and extra-cellular polymeric substances, EPS) provide a large number of binding sites for contaminants (Calmano et al. 1993; Gerbersdorf et al. 2009). Consequently, suspended sediment particles can accumulate pollutants from the water column and, hence, they can constitute secondary sources of contamination during dredging or (extreme) flood events (Ahlf et al. 2002; Hollert et al. 2000, 2003a). Thus, the assessment of sediment stability and the associated uncertainty is now perceived to have an important and emerging role for further implementation of

the European Water Framework Directive (WFD) (Hollert 2007; Hollert et al. 2007). The high relevance of sediment re-mobilization together with the increased bioavailability of particle-bound pollutants has also been viewed as an apparent result of global climate change. In general, globally intensified water cycling is expected, which will also cause a significantly greater flood risk (Hulme et al. 2002; Wilby et al. 2006). It is believed that the frequency and intensity of flood events will increase (Ikeda et al. 2005; Kay et al. 2006), and a higher number of extreme weather events and heavy rains is expected in Central Europe, leading to the occurrence of 500-year floods, such as the River Elbe flood in Germany in 2002 (Schüttrumpf and Bachmann 2008). In order to manage the threat of extreme flood events, many countries currently allocate additional retention areas, such as the “Integrated Rhine Program (IRP)” along the River Rhine in Baden-Württemberg (Disse and Engel 2001). Despite building and construction efforts to minimize damage by the flood wave itself, the transported pollutant load remains a great concern. A comprehensive and integrated flood risk assessment has to consider the consequences of such contaminant loads.

3 The joint research project SEDYMO

As a reaction to these requirements, the joint research project SEDYMO (“Feinsedimentdynamik und Schadstoffmobilität in Fließgewässern”; Westrich and Foerstner 2007), funded by the German Federal Ministry of Education and Research (BMBF) from May 2002 to July 2006, aimed at a profound understanding of sediment dynamics and pollutant mobility. It comprised 13 research projects geared towards an interdisciplinary approach for investigating the redistribution of

contaminated sediments and the re-mobilization of nutrients and pollutants into the water column as a consequence of hydrodynamic processes in rivers and estuaries. The gained data revealed that it is inevitable to consider erosion stability of the sediment bed in the implementation of the WFD and other comprehensive management concepts of other catchment areas. An important contribution was the development of experimental procedures and models for the transfer of laboratory data to in situ conditions. In a recently published book, the uncertainty of chemical, hydraulic and biological sediment data was highlighted (Ahlf and Heise 2007; Westrich et al. 2007). Furthermore, the urgent need for studies combining sediment mobility and toxicology was highlighted (Hollert et al. 2007).

4 The projects RIMAX-HoT and FLOODSEARCH

However, questions concerning the implications of flood events to particle-bound contaminants have not yet been sufficiently addressed by the scientific community. To bridge this gap, a recent project assessed the risk of (extreme) discharge events for drinking water supply at the site of the future flood retention area in Bellenkopf-Rappenwört, near Karlsruhe, Germany (Maier et al. 2005). Furthermore, within the framework of the German Excellence Initiative, the “Pathfinder” project Floodsearch at RWTH Aachen University, Germany, has made the first attempt to experimentally link hydrodynamic questions with ecotoxicology to assess the ecological relevance of re-suspension events to aquatic ecosystems (Wölz et al. 2009). In this new inter-disciplinary approach, rainbow trout (*Oncorhynchus mykiss*) were exposed to unspiked artificial sediment and to sediment that was spiked with polycyclic aromatic hydrocarbons (PAH) under simulated 5-d flood-like conditions in an annular flume which is a device developed for the investigation of erosion and deposition processes (Schweim et al. 2001; Wölz et al. 2009). Following exposure, a set of different biomarkers in the test organisms was investigated (Brinkmann et al. 2010). The set included measurements on the transcript level (quantitative reverse-transcription real-time PCR), on the protein level (Western immunoblot analysis of Cytochrome P450 1A concentrations, 7-ethoxyresorufin-*O*-deethylase activity, glutathione-*S*-transferase activity, catalase activity), as well as biochemical (lipid peroxidation) and cellular alterations (micronucleus formation). Furthermore, concentrations of the PAH metabolites 1-hydroxypyrene, 1-hydroxyphenanthrene, and 3-hydroxybenzo[*a*]pyrene in the bile fluid of exposed animals were measured. Experimental results supported the assumption that re-mobilization of sediments during simulated flood events in the annular flume can lead to uptake and biological effects of sediment-bound con-

taminants. Especially the average micronucleus frequency was significantly elevated after exposure to spiked sediments compared to exposure to unspiked artificial sediment and positively correlated with the biliary concentrations of 3-hydroxybenzo[*a*]pyrene, which is a metabolite of the indirectly genotoxic PAH benzo[*a*]pyrene.

Thus, this new approach has been shown to be promising for successfully conducting hydro-toxicological studies with rainbow trout. However, technical modifications of the annular flume (e. g., monitoring of more environmental variables) and an increase of the dimensions to (1) better simulate environmental conditions, and (2) reduce the influence of the exposed organisms on the physico-chemical processes, would be desirable. Currently, rainbow trout have a significant influence on the re-suspension dynamics of the sediment bed. Furthermore, it appears necessary to systematically control environmental variables such as pH and temperature in future studies, especially with respect to investigations of climate change effects.

The results of the study have shown that relatively short exposure to re-suspended sediments during simulated flood events can lead to – potentially adverse – alterations of biological functions in rainbow trout. Thus, the ecological and toxicological impacts of contaminant re-mobilization during flood events have to be considered highly relevant. Integrated approaches for risk assessment of regularly flooded rivers are urgently required.

5 Conclusions and outlook

Various approaches for assessing the toxicity of fluvial sediments and suspended matter were introduced and proposed in the past. The application of well-designed bioassay-directed fractionation techniques using acute, as well as mechanism-specific biological endpoints in combination with in situ investigations in the context of weight-of-evidence studies possess high potential for the causal analysis of complex environmental problems. In addition, such an approach can also lead to better sustainable action plans for the management of highly contaminated rivers and streams. However, there is also an urgent need for the development of other reliable tools for flood risk assessment integrating sediment stability and hydrodynamics in management strategies for the catchment areas in focus.

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