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Critical Review

ASSESSING CONTAMINATED SEDIMENTS IN THE CONTEXT OF MULTIPLE STRESSORS

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Abstract—Sediments have a major role in ecosystem functioning but can also act as physical or chemical stressors. Anthropogenic activities may change the chemical constituency of sediments and the rate, frequency, and extent of sediment transport, deposition, and resuspension. The importance of sediments as stressors will depend on site ecosystem attributes and the magnitude and preponderance of co-occurring stressors. Contaminants are usually of greater ecological consequence in human-modified, depositional environments, where other anthropogenic stressors often co-occur. Risk assessments and restoration strategies should better consider the role of chemical contamination in the context of multiple stressors. There have been numerous advances in the temporal and spatial characterization of stressor exposures and quantification of biological responses. Contaminated sediments causing biological impairment tend to be patchy, whereas more pervasive anthropogenic stressors, such as alterations to habitat and flow, physical disturbance, and nutrient addition, may drive large-scale ecosystem responses. A systematic assessment of relevant ecosystem attributes and reference conditions can assist in understanding the importance of sediments in the context of other stressors. Experimental manipulations then allow for the controlled study of dominant stressors and the establishment of causal links. This approach will result in more effective management of watersheds and waterways. Environ. Toxicol. Chem. 2010;29:2625–2643. © 2010 SETAC

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INTRODUCTION

The science and approaches used to assess sediment and ecosystem quality have progressed dramatically over the past three decades, but uncertainties that prevent us from establishing stressor causality still exist. Understanding the cause-and-effect relationship between stressors and biota is crucial for the effective management, restoration, and preservation of aquatic systems. It is generally understood that sediments can act as stressors in ecological systems because of both their quantity and their quality. Methods for assessing site-specific ecological impairments resulting from the physical stress of excessive sediments are not highly developed, whereas methods for assessing the links between sediment quality and ecological impairment are more varied and defined [1].

Sediment quality assessments are usually conducted in response to a regulatory driver, and tend to use standardized toxicity and bioaccumulation tests and comparisons with sediment quality guidelines (SQG) [2–4]. Determining the impact of sediments on an ecosystem requires more than asking whether sediment quality guidelines (e.g., probable no-effect concentrations) are exceeded, whether a chemical-specific (or sediment mass) clean-up goal is met, or whether sediment toxicity exists [1,4]. Accurate assessments of ecosystem and sediment quality integrate multiple methods, each with their strengths and limitations [1,5–10]. Even when a weight-of-evidence (WoE)-based environmental quality assessment has been conducted using multiple lines-of-evidence, study designs are often disjointed and do not produce powerful tests of stressor causality [8,11]. For example, sampling for physicochemistry of sediments and water, benthic communities, and toxicity is not done simultaneously, from the same sample or at the same time, nor are high-flow/storm events considered. This confounds exposure-and-effect linkages, and then leads to ineffective decisions and actions on how to improve, restore, and/or remediate the ecosystem [4,12].

Many comprehensive publications and reviews have addressed sediment quality and benthic community health, most of which focus on sediment quality guidelines and WoE-based approaches [1,5,6–8,13]. Research and regulatory activities dealing with contaminated sediments have been primarily directed towards chemical contamination and a single exposure route (bedded sediments), rather than suspended solids, resuspended sediments, food, pulse events, or effects in the context of nonchemical stressors. A great deal of research into sediment processes, movement, and geomorphology has also been done, but less focus has been directed on identifying biological effects of clean sediment (e.g., smothering, clogging, and abrasion), particularly in the presence of other stressors. As a consequence, gaps exist in our understanding of when and why sediments are important anthropogenic stressors in aquatic systems.

Given the large economic cost of remediating and restoring contaminated sediments and stream habitat in human-dominated systems, it is both surprising and disconcerting that very few cases have demonstrated improvement in ecosystem quality [4,14,15]. With the tremendous degree of spatial and temporal heterogeneity of ecosystems and their associated sediments, determining appropriate reference conditions from which to evaluate ecological impairment and recovery is paramount [16]. Sediment remediation and restoration projects often fail to adequately establish baseline conditions, the role of other stressors in ecosystem responses, possible sources of contamination to the system other than the targeted sediments, and follow-up monitoring of important ecological receptors [4].

The extensive state and U.S. Environmental Protection Agency's (U.S. EPA) biannual surveys of U.S. waterways have consistently ranked siltation, loss of habitat, and nutrient addi-

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tion as common and important stressors in human-dominated waterways ([17,18]; http://www.epa.gov/waterscience/criteria/sediment/pdf/sab-discussion-paper.pdf, [19]). A threats analysis for the Great Lakes ecosystems (open lake, coastal shore, coastal marsh, lake plains, tributaries, uplands, and wetlands) ranked lake levels/dynamics, stream flow, nuisance invaders, and habitat destruction as high threat stressors in terms of severity and scope, with toxics, nutrients, and water tables ranking as medium threats ([20]; http://www.epa.gov/ecopage/glbd/issues/). While not all other nation states have gone through a process of ranking waterway stressors, they certainly acknowledge a range of potential changes to environmental conditions that may result in ecological deterioration such as altered pH, physical disturbance, flow, habitat removal, pest species, pathogens, and altered connectivity ([21]; http://www.ozcoasts.org.au/pdf/CRC/69_estuary_assessment_final_screen.pdf). For example, the primary objective of Australia's Great Barrier Reef Water Quality Protection Plan is to reduce clean sediment and contaminant inputs from nonpoint source broadscale land use [22]. Despite the prevalence and importance of a range of stressors, their risk and interactions in relation to contaminated sediments needs to be strengthened to provide for more accurate ecological risk assessments. Indeed, Suedel et al. [23] have recommended the formal consideration of nonchemical stressors in the risk-informed decision framework for dredging projects.

A number of excellent reviews and studies have recently highlighted the importance of ecological responses resulting from interactions between benthic communities and various ecosystem attributes and drivers [24–27]. These drivers of ecological responses must be considered when assessing site conditions in order to determine stressor causality. This critical review will focus on some of the key issues that are important considerations when assessing sediment contamination.

WHAT IS THE EXPOSURE?

A principal source of uncertainty in predicting ecological risk lies in characterizing exposure to a stressor or stressors [11]. Ideally this would consist of measuring and integrating the multiple routes of exposure to multiple stressors, both spatially and temporally. In human-dominated ecosystems, this is a daunting task, given the many stressors and possible interactions. To address this complex issue in the context of sediments as stressors, it is useful to classify waterways and consider the major ecosystem attributes that influence the relative importance of each stressor.

The European Union's Assessment System for the Ecological Quality of Streams and Rivers throughout Europe Using Benthic Macroinvertebrates system for assessing ecological quality in European streams is an example of an ecosystem ''type-specific'' approach [28]. For each stream, a different set of calculations of ecosystem quality is used and comparisons are made with different reference conditions [29]. In locations where an ecosystem classification process has not yet been conducted and suitable reference conditions have not been described, it is still productive to contrast broad ecosystem categories such as low- versus high-energy waters (Table 1, Fig. 1), natural versus human-dominated, marine versus freshwater, tropical versus temperate, polar versus temperate, shallow versus deep, vegetated versus unvegetated, and water column versus bedded sediment. Within each dichotomy, there are fundamental differences that drive exposure scenarios and the bioavailability of contaminants. These realities can assist in

Table 1. Effects of clean and contaminated sediments on benthic and water column species in low- and high-energy environments[a]

Low-energy environments				High-energy environments			
Increased input of clean sediment		Increased input of contamination		Increased input of clean sediment		Increased input of contamination	
Benthic fauna and infauna	Water column species	Benthic fauna and infauna	Water column species	Benthic fauna and infauna	Water column species	Benthic fauna and infauna	Water column species
Severe permanent consequences	Severe permanent consequences	Severe and permanent consequences	Minor temporary consequences	Minor temporary consequences	Severe consequences	Moderate consequences	Severe consequences
Smothering of benthic fauna; reduced habitat availability through loss of heterogeneity and increased embeddedness	Altered predator–prey relationships, loss of food, potential smothering of eggs	Historical legacy of contamination causing toxicity and bioaccumulation	(unless frequently resuspended by human activity such as dredging or shipping)	Increased abrasion and scouring of benthic fauna	Blinding predators, clogging filter feeding apparatus	Ongoing input of contamination will increase benthic sediment contamination via embeddedness	Ongoing input of contamination causing toxicity to filter feeders and pelagic species
Loss of biodiversity	Loss of biodiversity? Selection for tolerance to high TSS	Loss of biodiversity; selection for tolerance to contaminants	Subject to bioaccumulation and altered food sources if frequent	Smothering of infauna likely to be temporary, but frequency an issue	Impact will depend on intensity, frequency, and duration of suspension event	More contamination exported— issue for off-shore or catchment habitat	Toxicity may be reduced by addition of TSS and/or DOC with contamination

[a] TSS = total suspended sediment; DOC = dissolved organic carbon.

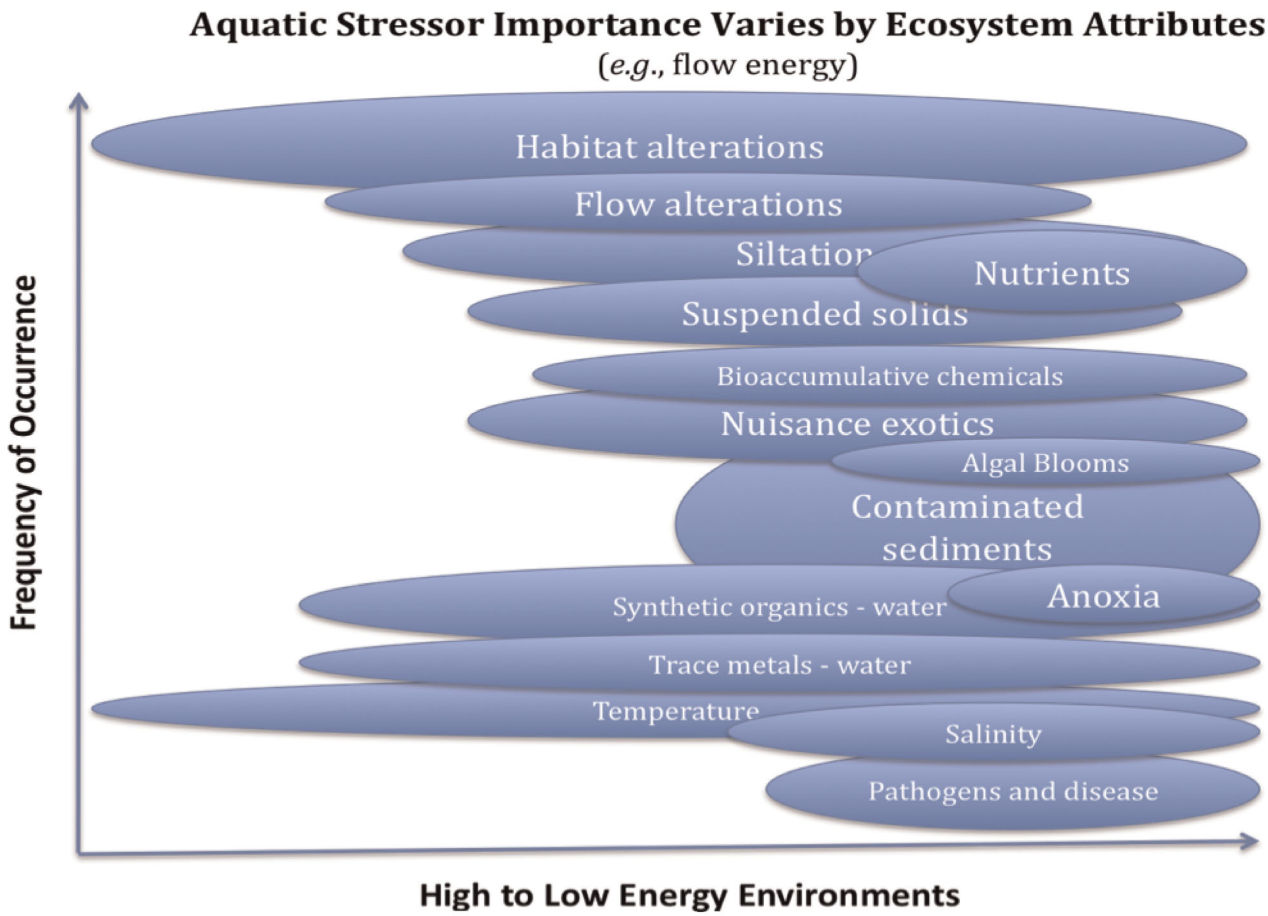

Fig. 1. The frequency of occurrence of common aquatic stressors in high- to low-energy environments. Habitat alterations may consist of a range of physical, chemical, and biological factors, including flow alterations, siltation, and suspended solids. The pathogens and disease stressor relates to the impairment of aquatic life. Rankings are a subjective compilation of several sources ([17,19,304]; http://www.epa.gov/bioiweb1/pdf/EPA-822-R-05-001UseofBiologicalInformationtoBetterDefineDesignatedAquaticLifeUses-TieredAquaticLifeUses.pdf); [305]; http://www.epa.gov/owow/tmdl/results/pdf/aug_7_introduction_to_clean.pdf).

better focusing the risk assessment process and relative ranking of which stressors often dominate the system (Fig. 1). For example, in a high-energy system, such as a mountainous stream, contaminated depositional sediments or siltation are likely not stressors, however, suspended solids may be (Table 1, Fig. 1). Conversely, in a low-energy, human-dominated system, siltation and embeddedness may be degrading habitat and smothering eggs and adults. These same sediments are likely to accumulate contaminants that exert toxicity to infauna, and to water column animals if the sediments are resuspended.

The spatial extent of any disturbance event is of critical importance in determining organism exposure. This issue must be related to the scale of both habitat and benthic community *patches* and will determine the ecological significance of any impacts (discussed below). Stressor exposures also vary in frequency and intensity [30]. Pulse exposures to physical and chemical stressors are common in human-dominated systems, yet predicting the risk of pulse exposures is very difficult and largely dependent on organism sensitivity, the duration and frequency of the pulse [31–33], and the site and season of contaminant release [34].

Understanding the life history, movement, and feeding patterns of an organism can help determine which exposure routes and stressors are likely to be most important to that particular species. For example, ingestion of contaminated sediment particles and overlying waters are important routes of contaminant exposure for some benthic organisms [35,36]. Stressor exposure via overlying waters and food will be strongly influenced by habitat and flow, two factors that also exert direct control over benthic assemblages [37–39]. Dissolved and colloidal phases of contaminants associated with interstitial waters are affected by upwelling and downwelling and must also be considered. Standard sediment toxicity test methods rarely assess the role of overlying water exposures from contaminants that desorb from the sediments [40], so that static or static renewal exposures may be measuring water column toxicity and erroneously interpreting it as sediment-related toxicity.

CLEAN SEDIMENTS AS STRESSORS

Unlike many chemical contaminants, both suspended and bedded sediments are a natural part of marine and freshwater ecosystems. Natural sediments are physically and chemically dynamic; they move downstream or across the seafloor and they change oxidative state and carbon content with depth. They are uplifted by many natural and anthropogenic processes such as tidal flow, bioturbation, dredging, vessel activity, storms, and waves [41]. Sediment movement and dynamic chemical cycling within sediments is critical to the healthy functioning of aquatic ecosystems. However, anthropogenic activities may change the chemical constituency of sediments and the rate, frequency, and extent of sediment transport, deposition, and resuspension. In these environments, sediments may function as both physical and chemical stressors. Hence, it becomes important to define the baseline environmental conditions, and the dominant flux processes for sediments in each aquatic system, so that deviations from reference conditions can be clearly identified, measured, and managed [42]. In many ecosystems, for example, fine-grained sediments are both natural and essential, such as in freshwater and coastal wetlands and marshes. The determination of whether sediments are a stressor is particularly driven by whether or not the ecosystem is high- versus low-energy, if a high-suspended solids concentration predominates, and the expectation of *reference condition* for that ecosystem.

Clean suspended and deposited sediments have been identified as a major stressor of aquatic ecosystems [18,19,43–47]. Natural disturbances are important processes that contribute to maintaining biotic diversity [48], so we must be careful only to attempt to manage clean sediment processes in locations where anthropogenic change has clearly modified them (e.g., construction-related erosion, agricultural erosion, dredging, locks, and dams). The manner in which clean sediments act as a pollutant/stressor will differ from site to site, but frequently occurs in the form of an increase in the mean total suspended sediment (TSS) load, more frequent resuspension events, or greater scouring, erosion, and deposition [18]. Baseline rates of sediment deposition, erosion, and suspension under natural and disturbed conditions can be crudely established by comparing impacted water bodies with suitable reference locations at multiple times. Sediment stability and flux is site-specific, variable, and complex [49,50], and should be characterized using field and/or laboratory evaluations of resuspension [51–54], otherwise, the accuracy of estimates of resupension may be off by orders of magnitude. At times, natural sediment processes will be extreme and represent a natural disturbance or turbid flow period [55]; guidelines for suspended sediment concentrations during these periods may be less restrictive than during clear flow periods.

Excessive deposition of fine-grained sediments in larger-grained sediment ecosystems can have adverse ecological impacts. This may relate to the smothering of benthic flora and fauna [56,57], reduced light penetration, organism abrasion, clogging of filtering mechanisms, or the loss of habitat complexity [18,43,45,46,58,59]. Multiple deleterious impacts associated with fine sediments have been reviewed and widely documented to habitat, primary producers, macroinvertebrates, and fisheries [45].

A vital physical characteristic of aquatic habitats, particularly lotic systems, is the degree of substrate embeddedness that occurs as fine sediment fills interstitial spaces in streambeds. Lotic environments are typically heterogeneous, with a wide range of sediment grain-size distribution which may be mixed or have variable patchiness. Habitat-specific microdistributions and patch dynamics are particularly important for invertebrate diversity [60–62]. Small-grained sediments (clays) may become embedded between larger grained sands, gravels, and cobble, or settle in depositional areas and become compacted to varying degrees. This not only results in a loss of habitat to species that require interstitial space, such as mayflies and stoneflies, but increases the susceptibility of certain invertebrates to predation by fish [3]. Elevated fine sediment loadings have different effects on hyporheic exchange and associated ecological processes (e.g., nitrogen and carbon cycling) depending on local hydrologic and geomorphic conditions [60,63–65]. Although alteration of streambed habitat is recognized as one of the most important stressors of benthic organisms, the reliability of findings from existing embeddedness methodologies have been questioned due to substantial variation in their quantification and assumptions [66].

Increased suspension, delivery, and deposition of clean fine sediments also causes problems in estuarine and marine ecosystems. Strong links exist between infaunal ecology and sediment grain size, such that changes to grain size through deposition or erosion can have biological consequences [60,67,68]. Sandy habitats are transformed to muddy ones in human-dominated environments, and the rate at which this happens is predicted to increase with global climate change. High-flow events and wave activity during intense storms allow suspended sediment to breach natural filters such as sandbars, mangroves, and marshes. Under these conditions, sediment is transported further out into bays and the open coast. When resuspended, the fine particles reduce light availability to organisms such as macroalgae, seagrass, phytoplankton, and animals hosting algal symbionts such as corals [69]. When the sediment settles, this can result in short-term, catastrophic smothering events, but also long-term habitat change, as has already been observed in many human-dominated watersheds [70]. Sediment- and flow-related stress in human-dominated harbors, lakes, and large rivers may also be driven by navigation activities. Navigation-induced physical forces resuspend sediments and affect fish swimming performance and the filter feeding capacity of invertebrates [46,71].

The suspension of clean sediments can cause serious effects through mechanical damage or clogging of the breathing or respiring organs of aquatic invertebrates [72,73]. Laboratory-based experiments have been used to assess the effects of resuspension of uncontaminated sediments on aquatic biota, predominantly fish [72]. These studies have found that the duration, intensity, and frequency of exposure to suspended sediment are important and have resulted in the development of water quality guidelines for total suspended solids [55]. *Daphnia magna* and some other daphnid species have been found to be some of the more sensitive species to elevated TSS concentrations [9,74–76]. However, these lentic species are less commonly found in lotic systems and may be an inappropriate surrogate for stream environments. River flow indexing using benthic invertebrates was recommended in Britain for setting hydroecological objectives [38]. Community structure in limestone streams was predicted well by summer flow, the amount of impervious area, and the degree of regulated or augmented flow. All of these factors influence sediment exposures for benthic organisms in particular.

Long-term exposure to elevated levels of sediment deposition and suspension due to activities such as agriculture or mining, often result in permanent changes to the stream or seabed geomorphology and ecology [45]. While uncontaminated sediments are usually more biologically diverse than contaminated sediments [77], human activities that modify sediment regimes may still result in selection for a small group of sediment-tolerant species. It is likely that many estuarine organisms in low-flow depositional environments are adapted to high-frequency deposition of small volumes of sediment. In a mesocosm study, nematodes were more affected by single large deposits of sediment than by frequent small deposits, regardless of whether the sediment was contaminated or clean [57]. The relative ecosystem impact and recovery following fine sediment exposures may be largely dependent on the duration of exposure [45,75]. Robinson et al. [75] observed a dose-dependent decrease in survival of *Daphnia magna* with continuous exposure to suspended clays; however, when exposures were reduced from 24 h to 12 h, there were no detectable effects on survival or reproduction.

Increased input of organic matter associated with fine sediments often represents the addition of a food source that may result in the domination of communities by a few opportunistic species. The increased organic matter may also lead to increased sediment oxygen demand and ammonia production. Sediment can also be extensively modified using ecosystem engineering processes that directly and indirectly impact food webs and chemical processes. For example, the proliferation *of Dreissena* mussel clusters in lakes has increased benthic algal primary productivity, microbial and benthic diversity, and submerged aquatic vegetation due to the presence of their shell habitat and filtering of plankton [78–80].

CONTAMINATED SEDIMENTS AS STRESSORS

Bed sediments and contaminant flux

Streams, rivers, and coastal areas that are characterized as nondepositional environments due to high flows and currents (i.e., high energy; Fig. 1, Table 1) are less likely to be impacted by contaminated sediments. An exception to this is directly below mining operations where larger grained mine tailings may exist, or where periphyton have accumulated high metal concentrations from the overlying water [81–84]. The dominant contaminant exposure pathways for aquatic organisms in high-energy systems will tend to be runoff, outfalls, upwellings and downwellings, and food [9,75,85–92]. In low-energy, depositional environments, bed sediments become a major route of exposure for many benthic organisms. A number of reviews have been written regarding methods for assessing contaminated sediments, including sediment toxicity and bioaccumulation tests of single species, indigenous benthic community indices, and the use of sediment quality guidelines [5,13]. However, these reviews have not emphasized the importance of better characterizing exposures to bioavailable fractions of chemical contaminants that are dictated by a number of flux processes. The primary flux processes are resuspension and deposition, bioturbation, advection, upwelling/downwelling, diagenesis reactions, and diffusion.

Metal flux out of sediment has been shown to be a dominant process ($Cd > Zn > Co = Ni = Cu > Pb$) in marine harbors over periods of days and related to benthic oxygen demand for some metals [93]. Carbonaro et al. [94] created a one-dimensional reactive and transport model on metal fate in sediment that incorporates metal-sulfide formation/oxidation and partitioning to organic carbon and Fe oxyhydroxides. Nickel and Zn fluxed into overlying water, followed by slow decreases, and were related to the initial pore water concentrations that overwhelmed sulfide complexation. They suggest a further need to incorporate Mn cycling and Mn oxyhydroxide sorption, and binding to pyrite with metal speciation calculations as a logical extension of their model. Similar results have been observed in the field with Ni spiked freshwater sediments with rapid loss of sediment Ni during the initial one to two weeks of exposure [95]. The loss varied from 11 to 89% depending on the sediment's complexation capacity. The rapid scavenging and complexation of potentially toxic metals by Fe and Mn oxyhydroxides reduces their bioavailability during resuspension or bioturbation events [96–98]. In a marine harbor, overlying water pH and sediment mixing were found to be the dominant processes controlling metal (Cd, Cu, Pb, Zn) release and sequestration rates [99]. As pH decreased, oxidative precipitation of released Fe and Mn greatly influenced sequestration rates of released Pb and Zn. Additionally, sequestration was reduced at lower dissolved oxygen levels.

A rapidly growing body of literature documents the common occurrence of the diurnal flux of metals and organometals between sediments, biofilms, and overlying fresh and marine waters due to a number of processes [82,83,100–107]. Diurnal concentration ranges of several hundred percent have been documented [107]. Fluctuation varies by metal type and site conditions, and are driven by processes such as bioturbation, surface water and hyporheic flow, redox and photochemical reactions, precipitation/dissolution of solid mineral phases, and sorption processes. Many of these are interlinked with photosynthesis, pH, and temperature. Most of these processes are not accounted for in sediment transport models or in assessments of organism exposures and related ecotoxicity. However, 96-h in situ exposures of cutthroat trout to nightly increases of Cd and Zn (originating from sediments and periphyton) by as much as 61 and 125%, respectively, did not affect survival [108]. The authors speculated this was due to the short-term nature of the exposure and the lower temperatures reducing uptake.

Significant ground water–surface water interactions commonly occur near the banks of riverine systems and intertidal areas. These may be upwellings of groundwater or advection through sediments into overlying surface waters or vice versa. While such exposure routes tend to occur in larger grained sediments, they have also been shown to be important in some depositional areas, for example, New Bedford Harbor, Buzzards Bay, Massachusetts, USA [93,109]. Quantifying ground water–surface water interaction exposures improves stressor-causality linkages with benthic macroinvertebrates [10,110]. Ecosystems with altered flow conditions may also experience periodic rewetting of metal contaminated soils and sediments. Rewetting of oxidized acid-sulfate soil produces acidic pulses that alter sediment metal bioavailability [111,112]. Lower pH exposures can both increase and reduce exposure [111], whereas reoxidation can result in the release of metals and increased exposure [112]. Heavy rains also result in the release of dissolved Cu and Zn from sediments into overlying waters. Such dynamic responses may be reflected in periphytic algae concentrations that followed free Zn and exchangeable Cu concentrations in the water [113].

Factors affecting chemical sequestration

It is apparent from the above discussion that predicting metal bioavailability and toxicity in sediments is difficult, given their complex and often dynamic interactions with organic and inorganic compounds. Therefore, it would seem that the idea of equilibrium partitioning as a foundation for sediment metal and organic chemical criteria would be flawed. Nevertheless, many laboratory and field studies have been performed since the 1990s, describing the role of the organic carbon and the procedurally defined acid volatile sulfide (AVS) fraction in the binding of nonpolar organics and metals in depositional sediments and their relationship to benthic macroinvertebrate toxicity [95,114,115]. Acid volatile sulfides are found only in anoxic, depositional sediments, which are the sediments of greatest concern because these are where chemicals accumulate. Significant focus has been directed on why the AVS to simultaneously extracted metal (SEM) relationship might not be predictive of toxicity levels in benthic organisms [36,116–127]. For example, most burrowing animals have oxygenated burrows and would rarely interact with the sulfide-rich anoxic sediment component [128]. Given the limitations, it is interesting that such a wealth of laboratory and field studies have shown AVS to SEM relationships (e.g., SEM:AVS ratio and SEM-AVS) to be a very effective predictor of the absence of toxicity to diverse benthic communities where depositional sediments exist [114]. Two possible explanations for this growing body of evidence, despite the reality that AVS does not exist at the sediment–water interface where many organisms are residing, are the upward flux of metals through diffusion and advection, and the AVS-SEM acid extraction process which will also extract surficial Fe- and Mn-oxides that have metals associated with them. In high-energy oxic environments, where larger grained sediments exist, AVS will be absent and not useful for predicting benthic organism exposures.

The bioavailability of metals that are complexed readily by dissolved organic matter, such as Cu and Ni, is affected by sunlight. Photo-oxidation of dissolved organic matter may release free metal. The extent and significance of this phenomenon is not known, but may increase in significance due to climate change alterations in ultraviolet and runoff-terrestrial dynamics [129].

Marine sediment toxicity is influenced by several factors that control chemical bioavailability, particularly for testing of metals, as compared to freshwater sediments. In general, the higher ionic strength of marine waters will reduce metal ion activity with more chloride complexation, and higher sulfate concentrations [130,131]. However, protons are known to compete with some metals for uptake, so under the more alkaline conditions of seawater, metal ion activity might increase. Potentially, a larger fraction of the metals are associated with Fe and Mn oxides in easily reducible fractions in sandy and tidally influenced sediments. The principles governing geochemical processes controlling metal exposure from marine sediments to resident benthic organisms would not be fundamentally different from those that affect exposure in freshwater. However, the factors affecting metal toxicity and bioaccumulation may well be different [132], depending also on the different ion-regulatory physiologies of marine, estuarine, and freshwater organisms [133]. Among the geochemical constituents affecting metal toxicity that change from freshwater to seawater are Ca, Ng, and Na—the cations known to compete with metals for uptake at the gill surface. Binding of metals to DOC may also change, given the varying types and concentrations of DOC that exist throughout aquatic ecosystems.

Microbial biofilms (periphyton) are increasingly recognized as an important compartment in aquatic ecosystems [83,101,104,134,135], and they are a prime site of nutrient and contaminant concentration in streams. The rate of contaminant uptake by biofilms is largely related to their biomass which is strongly affected by streambed morphology, flow, and nutrient availability [134]. Streamside flow-through mesocosms containing Ni-spiked sediments showed Ni rapidly fluxed out of sediments into overlying periphyton mats where the Ni remained trapped in the periphyton, apparently complexed with organic matter and Fe- and Mn oxyhydroxides [95]. This Ni exposure did not appear to cause adverse effects on benthic fauna associated with the periphyton [95]. For some benthic communities, biofilms are a source of contamination to grazing invertebrates [136]. In marine systems, microbial biofilms may act as a source of contaminants to grazing invertebrates through adsorption of metals by extracellular polymeric substances that the microorganisms excrete [137].

Suspended solids and resuspended sediments

Much of the loading of chemical contaminants in aquatic systems is from diffuse sources [138], and suspended sediments have been identified as a major vector for the transport of contamination. Annual suspended sediment and trace metal fluxes were followed in several major drainage basins of the United States, and more than 70% of Zn, Cu, and Ni were transported in association with suspended sediment [139,140]. In Boston Harbor, regular (tidal) low-energy resuspension events contributed up to 60% of the metal flux per year. Some monitoring programs even use measurements of suspended sediments as a surrogate for trace metal movements in contaminated locations [141]. Organic contaminants are also transported with resuspended sediments. Lake Michigan is regularly subjected to intense storms that result in sediment plumes responsible for the redistribution of large amounts of organic contaminants [142].

During sediment resuspension events, toxicity may result both from aqueous contaminant exposure (contaminants released from particulates) and from contaminants associated with suspended sediments that are ingested by organisms [143]. Resuspension-related release of metals and organic compounds has been shown to increase bioaccumulation in freshwater and marine benthic species and caged fish [4,144]. Laboratory studies have also observed increased rates of contaminant (polycyclic aromatic hydrocarbon [PAH] and metals) uptake when sediments are resuspended [46,145–147]. *Daphnia* mortality was increased through ingestion of resuspended contaminated particles and the internal release of bound contaminants [76,145,148]. In situ exposures of amphipods and daphnids during agricultural runoff events showed that TSS-associated pesticides caused acute toxicity, whereas clean TSS or low-flow conditions were not toxic [149]. However, some resuspended sediments may reduce the toxicity of contaminants by rendering them biologically unavailable [150]. In laboratory studies, resuspended clean sediments reduced the toxicity of pyrethroids to *Cerodaphnia* [151] and mayfly nymphs [152]. Field exposures found that photoinduced toxicity related to PAHs was reduced during high-flow events due to suspended solids [153]. So, in summary, suspended/resuspended solids can both increase and decrease the bioavailability and toxicity of contaminants. This creates the difficult issue of determining when suspended solids are beneficial (reducing toxicity) or harmful (physical stress or release of contaminants), and both could be occurring in the same ecosystem. Currently, the science to accurately predict the interaction is limited.

A wealth of literature [86,102,105,154–160] documents the significant and widespread alterations that bioturbation can have on biogeochemical and contaminant flux. In low-energy, depositional environments, bioturbation and diffusion processes are likely to be important, along with episodic resuspensions due to large high-flow events, dredging and vessel activity [161]. Some areas will have dense benthic macroinvertebrate communities, comprised of species (e.g., crayfish, bivalves, Annelids) that can mobilize sediment contaminants through bioturbation from several centimeters depth (or meters in marine systems) to overlying waters. Bioturbation can increase the resuspension rates of contaminated sediments; however, this does not necessarily increase the bioavailability of contaminants [154,162]. For example, mayfly (*Hexagenia rigida*) and Asian clam (*Corbicula fluminea*) had less uptake of Cd and Zn in bioturbated systems than in unbioturbated systems [163]. However, bioturbation by amphipods significantly increased

TSS concentration in the overlying water and, consequently, the total aqueous concentration of sediment-bound fluoranthene, which was subsequently accumulated by filter-feeding mussels [164].

The importance of resuspension events on contaminant release will be a function of several factors. For metals, some of the key controls on release rate will be the length of oxidation period, the occurrence of fresh Fe or Mn oxyhydroxides, and the SEM to AVS ratio and other physico-chemical parameters [96,165–167]. Only a small fraction of the metal or ammonia may be released during resuspension events, but it may be toxic [147,163,168–171]. Some challenges to current paradigms have arisen from models of contaminant release in aquatic systems. The kinetic model of Birdwell et al. predicted that dissolved concentrations of metals will be highest at some distance (2.5–7 km) downstream from a point of dredging [172]. Several field, laboratory, and modeling studies have investigated organic contaminant release under various resuspension scenarios [142]. However, Birdwell et al. [172] argue that there is currently no tool to predict the release rates of organic contaminants from particulates during a resuspension event.

OTHER COMMON STRESSORS AND THEIR INTERACTIONS WITH SEDIMENTS

Stressor interactions and stressor dominance

As noted above, several important sediment properties and processes influence benthic and pelagic communities, whether or not chemical contamination exists. In addition, increasing evidence suggests important physical, biological, and chemical linkages aside from the traditional upstream–downstream and benthic–pelagic couplings that act through a range of interfaces including terrestrial–aquatic, surface–subsurface, lake–stream, river–floodplain, and marine–freshwater [26]. Human activities may interfere with these linkages causing changes to hydrological processes that affect the balance between groundwater and surface water, the permanence of water bodies, nutrient cycling, acidification, and runoff [173–176]. Modifications to these processes may result in cascading effects from multiple stressors [177,178]. Unless these various ecosystem linkages are understood, then protecting and restoring impaired ecosystems will be fraught with uncertainty [26].

One assumes that known stressors to aquatic ecosystems would behave in a cumulative manner, with exposures to additional stressors resulting in additional adverse effects. For example, clean sediment resuspension reduced algal growth, but this was accentuated when irgarol antifouling paint particles were included in the sediment [179]. However, stressors may behave in additive, antagonistic (less than the sum of effects), or synergistic (greater than the sum of the effects) manner. This means the widely used assumption by regulatory programs of additivity based on single chemical thresholds may be flawed in some situations. Hill et al. [180] found that exposure to dissolved Zn reduced the toxicity of dissolved Cu to an Antarctic polychaete. Clements [181] demonstrated causal relationships between metal contamination (Zn, Cd, Cu) and benthic macroinvertebrate community responses. The combination of the three metals produced synergistic effects impacting the mayfly and stonefly populations to the greatest degree, with highly significant effects on macroinvertebrate drift and community respiration. Chemical and physical interactions that increased effects were observed on *Daphnia magna* between Ni and TSS, but not DOC [182]. Conversely, PAH-related photo-induced toxicity was reduced in the presence of TSS [153]. Mayflies exposed to pyrethroids had increased drift, unless there was high flow or the insecticide was associated with particles and drift was reduced showing an antagonistic response [152]. Strongly synergistic effects were observed between metal and PAH mixtures on marine meiobenthic copepods [183]. Kashian et al. [184] showed that ultraviolet-B radiation reduced community metabolism and other benthic indices. When there were simultaneous exposures to metals, synergistic effects and decreased community tolerance were observed. A mix of chemical contaminants is quite common in urban contaminated sediments and suggests that sediment quality guidelines based strictly on empirical guidelines for single contaminants may not be protective, unless safety factors are used.

The importance of effects due to stressor interactions, however, may be lessened when individual stressors have large effects [185]. In Hokkaido (Japan) streams, extensive experimentation of deforestation, channelization, erosion-control dams, biological invasions, and climate change revealed that most of these stressors caused 30 to 90% declines in foraging, growth, or abundance of aquatic or terrestrial predators [184]. In addition, the indirect effects of stressors crossed the aquatic-terrestrial boundary and cascaded throughout the terrestrial food web. Each stressor alone dramatically reduced food web components, whereas additional stressors had little effect. It may be that in heavily human-dominated watersheds, one stressor dominates the system, such as habitat or flow, or many stressors are tightly linked, such as impervious area, flow flashiness, and contaminated runoff [186,187]. For example, note the widely reported relationship between degraded aquatic communities in watersheds where impervious areas are greater than 8 to 20% of the landscape [174,175,188–192]. These areas are subject to multiple dominant stressors of habitat degradation, flashy flows, and elevated temperature, sunlight, solids, metals, and synthetic organics. Sciera et al. [174] demonstrated that, in urban and agricultural watersheds where development was occurring, hydrology and habitat were the most important stressors to monitor. Benthic community impairment was best predicted using a Normalized Disturbance Index quantitatively linked to an increase in the percentage of impervious cover, stormwater runoff, storm-event total suspended solids, and the benthic index of biotic integrity.

Weston et al. [193] and Ding et al. [194] have shown that most of the toxicity to *Hyalella azteca* in urban sediments can now be attributed to pyrethroids and their increasing prevalence. So the question arises, are pyrethroids such a dominant stressor that other stressors in urban depositional sediments are inconsequential? This can only be answered by addressing the tolerance of the resident benthic communities that exist in these urban streams (likely more tolerant than *H. azteca*) and whether or not pollution-sensitive communities could exist in the absence of the pyrethroids. *Chironomus riparius* responded more than a mayfly to the sediment nutritional levels than associated contaminants, suggesting that in human-dominated systems they would not be an appropriate surrogate for benthic species protection [195]. The question of which organism or community is the most appropriate test species will exist in any human-dominated ecosystem when attempting to rank the relative importance of the existing stressors. We therefore reiterate the call for a more prescriptive, experimental approach to establishing stressor importance that links controlled laboratory studies with field-based ecological manipulations and studies of exposure and response relationships between key physical, biological, and chemical conditions [14,24,181,185,196–198].

Climate change

It is difficult to predict how sediment stressor activities will interact with climate change. Climate change is likely to have location-specific effects on sediment processes that are strongly related to rainfall and storm activity. In general, where rainfall is predicted to decrease, we would expect reduced TSS loads, and where storm activity is set to increase, we would predict greater and more frequent sediment resuspension events. Altered flows due to drought or more frequent and severe weather will change harbor and estuarine salinity patterns, lake and marine hypoxia episodes, nutrient inputs, food availability, and reproductive windows of opportunity [129,185,197,199–202]. In addition, climate-related changes to ecosystems (temperature, precipitation patterns, ocean acidification, nutrient cycling, food inputs, migration patterns, range expansions and contractions, pathogens, and disease) will result in novel communities, which may lead to changes in ecosystem functioning and other unexpected biological feedbacks or *ecological surprises* [14,25,197].

Climate change will also alter exposures to chemical contaminants in water, food, and sediments through a variety of mechanisms of which many documented examples are unfolding [200]. Exposures will be altered through changes in physicochemical and biological fate processes, such as fugacity, biodegradation, and metabolism [203]. Some of these changes will result in increased exposures to some species or life stages, whereas others are decreased such as noted in metal type, concentration, and temperature interactions on fish [204]. Temperature has been shown to increase and decrease toxicity in the presence of fungicides and insecticides to some marine and freshwater amphipods [200,205], whereas a complex, concentration-dependent phenomenon has been observed with temperature and metal toxicity [204,206].

APPROACHES FOR ASSESSING STRESSOR IMPORTANCE

Assessment methods and limitations

There are a number of assessment methods and approaches that more effectively characterize exposure, effects, and identify stressor importance. These include field-based species sensitivity distributions for both chemical contaminated sediments, noncontaminated TSS, temperature, salinity, and sediment burial [207–213], improved exposure models of clean and contaminated soils and sediments [214,215], stressor toxicity identification and interactions [9,184,216,217], in situ experimental exposures including biomimetic monitoring [4,218], biological trait-based analysis [219], and data analyses methods for discerning reference versus impairment and stressor-effect relationships [16,220].

It is easy to highlight the limitations of any assessment method, whether they are from ecological or methodological perspectives [1,116]. Nevertheless, all of the popular assessment methods have been used effectively to assess ecosystem quality, when they are used judiciously. While the science has improved, the judicious use of the various methods with suitable acknowledgement of their limitations is not altogether common. In addition, a growing body of recent literature is showing the importance of new chemical stressors, their accumulation in sediments, and their interactions showing greater than additive effects [221–224]. The often common and significant stressors are frequently not characterized from an exposure-and-effects (risk) perspective. So, if the goal is restoring ecosystem quality, beneficial uses, or ecosystem services, how can that occur if the dominant stressors are not identified?

Few studies have actually evaluated the accuracy of assessment methods; however, previous studies usually document large error rates. In Ohio (USA), evaluation of indigenous biota showed 36% of the impaired stream segments could not be detected using water chemical criteria alone [225]. Evaluations of sediment quality guidelines have been better, with prediction rates of 70 to 75% [226,227]. However, error rates of 25 to 30% (and higher for metals) seem unacceptable given the ecological and economic implications of some of the resulting risk management decisions. A comparison of multiple assessment methods (lines-of-evidence) at several sites with metal and organic contaminants, showed accuracy rates ranged from 50 to 60% [40]. This suggests that no one method is adequate and points toward the use of multiple methods to best characterize whether or not sediments are significantly contaminated and the aquatic ecosystem is impaired [1,8].

Tools for experimental designs

Recently, Downes [228] noted that sorting out the effects of multiple stressors would be best accomplished by using the basic components of good experimental design. Experimental designs are available to determine the effects of suspended and deposited clean and contaminated sediments under controlled laboratory conditions [180]. Such studies help identify sensitive species and the causal stressor(s). The experimental designs should include exposing multiple species to different levels and types of suspended or depositional solids alone and in combination with other factors (such as contamination, salinity, and temperature) to determine direct effects of clean sediments versus other stressors. Ideally the frequency and duration of sediment exposures is also investigated [57]. In addition, it is important to consider the connectivity between organism traits (see below) and their exposure, because this is essential when establishing actual exposures and casuality.

Mesocosms can be used in the laboratory or field to determine effects on populations and communities. Stream-side mesocosms and artificial streams with multiple interaction stressor treatments provide another way to discern which stressors are dominating. Culp et al. [229] constructed a portable mesocosm system that contains 16 circular streams. Here, indigenous benthic biota were introduced to the streams, and natural stream water was combined with pulp mill effluent to simulate discharge dilution effects and separate nutrient-related stress from other factors. Many simple mesocosm designs have been used to assess the role of bioturbation on sediment resuspension and contaminant flux to filter-feeding organisms [119]. It is more difficult to create sustained sediment resuspensions within mesocosms without substantially affecting relevant physico-chemical parameters such as dissolved oxygen.

Sublethal effects or subtle interactions that occur in the field are not easily measured in standardized sediment toxicity tests [230,231]. Laboratory toxicity testing can result in artifacts that may alter exposures found in situ, such as changes in: oxidation, redox, pH, sorption and complexation, microbial activity and their by-products (e.g., ammonia), nonequilibrium conditions, organic material via sieving, predation, food availability, ultraviolet light, flow, and suspended solids [5,9,40,232–235]. For example, seasonal complexation of Cu to DOC was affected by solar irradiation, causing dissolved organic matter to photochemically degrade and release Cu^{2+} [129,236].

Many have called for more population- and community-based approaches looking at food chain-based effects, integrating key ecological factors beyond laboratory-based, single-species testing [6,237,238]. This includes a greater focus on ecologically relevant endpoints not traditionally used in ecotoxicology, such as predator–prey interactions, drift, preference-avoidance, and other behavioral measures. These endpoints have been shown to be more sensitive than mortality, growth, or reproduction measures of sediment toxicity and habitat degradation due to uncontaminated fine sediments [3,6,181,239–242]. In food webs with organisms interacting strongly through predatory or competitive relationships, indirect effects of contamination will arise [6,243]. Manipulative experiments conducted in the field can be used to test for effects of metal-contaminated sediments on multiple components of an ecosystem simultaneously. When sediment fauna are negatively affected by metal contamination, the recruitment of invertebrates to patches of hard substrate directly above the sediments may increase (N.A. Hill, personal communication). This is likely due to a reduction in infaunal predators such as annelids and crustaceans.

Hence, another useful approach for discerning sediment, habitat, and water quality stressors and factors that control bioavailability is by using caging, colonization, and transplant experiments [9,181,244–246]. Because sediments are usually altered chemically, physically, and biologically when they are sampled, there is a growing science that uses in situ observations, which can accurately document exposure and effects. The exposure of caged organisms has been recommended for fish and benthic macroinvertebrates to better characterize site and source exposures, thereby allowing for improved risk predictions or measures of remediation effectiveness [4,15]. Field-based tissue residues have been one of the oldest and most common assessment methods [247]. For sediment quality assessments, both fish and benthic macroinvertebrates, particularly bivalves, have been commonly collected and tissue levels compared to sediment concentrations. This technique was used in a before–after, control–impact experimental design to establish that dredging increased the bioavailability of sediment contaminants [248]. By separating sediment and water exposure compartments and documenting upwelling versus downwelling conditions using mini-piezometers, exposure sources and related toxicity can also be established [9,10].

Some excellent papers on methods for determining free metal ion concentration, labile species fraction, metal complexation capacity in waters, and sediment flux have been published [218,249–262]. A wide range of biomimetic approaches exists [263]. These include organic/plastic fibers, tubes, bags and sheets, gel probes, micro-ion selective electrodes, voltammetric electrodes, and optodes. These methods have provided new insights into controls on organic matter mineralization, benthic fluxes, impacts of redox alterations on metal fate, and geochemical reaction pathways [263]. As nanotechnology improves, electrodes will become smaller and more sensitive, and provide a way to determine chemical speciation of metal (organic and inorganic metal complexes and free metal) in situ by encasing microelectrodes into diffusive gradients in thin film- or diffusive equilibrium in thin film-type membranes [263].

In contaminated sediment scenarios, new stressor toxicity identification methods include whole sediment manipulations or in situ exposures with various stressor partitioning methods and substrates that may reduce the likelihood of artifacts [9,245,264]. The phase I type toxicity identification evaluations use similar resins to those used in the U.S. EPA methods, however organisms and benthic communities are exposed directly to pore or surface waters for 24 to 96 h directly in the field. These methods have been shown to be more sensitive than side-by-side laboratory-based toxicity identification evaluation tests, suggesting manipulation artifacts are causing a loss of toxicity. They effectively identified which stressors dominated at contaminated field sites by separating nonpolar organic, metal, and ammonia fractions. In addition, when combined with modified cages to restrict suspended solids or remove solar ultraviolet radiation, adverse effects from turbidity and photo-induced toxicity are possible [9]. Recent lab-based toxicity identification evaluations have effectively documented the importance of sediment-associated PAHs and pyrethroids as dominant sediment stressors in urban watersheds, and chlorpyrifos dominated in agricultural watersheds [205,265,266]. The importance of sediment metals in urban lakes is declining, as evidenced by a nationwide survey of cores from 1970 to 2001 [267]. Median changes ranged from −3% (Hg, Zn) to −46% (Pb), but remained elevated over undeveloped watersheds. These declines suggest that insecticides, such as the pyrethroids and PAHs will increasingly be the toxicants of concern in urban sediments.

A promising new assessment tool, particularly for benthic invertebrate-based ecosystem quality assessments, is the use of biological traits (morphological or functional) to develop geographically broad (continental) lotic ecosystem assessments of dominant stressors [196,219,268–271]. This approach may assist in discerning effects that are due to the physical presence of solids, as opposed to solids-associated chemicals. Proponents of this approach caution against the blind use of excessive traits for the indication of too may stressors and recommend a focus on mechanistic a priori predictions. In addition, one stressor may affect many traits, thus confounding other stressor-specific relationships [271]. For this field to progress, larger databases and poorly studied taxonomic groups are needed [270,271].

Perhaps the most important aspect of an experimental design aimed at assessing when sediments are stressors, is establishing the appropriate reference conditions. In a regulatory sense, this may be straightforward, whereby impairment is simply based on exceedance of environmental quality benchmarks. However, determining what constitutes ecologically significant impairments from a high-quality state is not simple. Hawkins et al. [16] noted that researchers have become increasingly more sophisticated in their approaches for determining reference conditions using site-specific modeling. These approaches have been based on ecological, thermal, hydrologic geomorphic, and chemical benchmarks. These advances have better linked the spatial and temporal dynamics of biota and their natural environment, and are inextricably linked to how well their environments are characterized in terms of accuracy and precision. Useful approaches that have emerged include variations of the River Invertebrate Prediction and Classification System [272–274], ecoregions [275], landscape and typological classifications [276,277], the Benthic Assessment of Sediment [278,279], and others [16]. Similar approaches in marine and estuarine systems are not as advanced, particularly where taxonomic clarity is not well established. However, extensive efforts are being made around the world to develop integrative tools and methods to assess the ecological health of estuaries, and substantial advances have been made by U.S. and European researchers [280,281].

A large number of landscape approaches to studying aquatic ecosystems have established causal linkages between landscape variables and biota [62,282]. These have ranged from microhabitat patches to regional in scale, with high to low resolution, respectively. As noted by Allan [283] in a review of land use effects on streams, there has only been moderate success in quantifying the underlying mechanisms due to covariation of natural and anthropogenic factors over multiple scales, and due to legacy and nonlinear responses. Hawkins et al. [16] suggest that these schemes tend to produce overly coarse estimates that are lacking in accuracy and precision, and should be replaced by predictive modeling approaches. These will be based on a better understanding of natural variability of ecological, physical, and chemical characteristics, which then allows one to discern significant effects (impairment) from sampling and prediction error.

By using data-rich models that are statistically based, the decision of reference versus impaired areas along with stressor rankings is possible and can enhance the decision-making process. Two such ecoepidemiological approaches were independently applied to the same environmental monitoring dataset of biological, physical, and chemical variables for the State of Ohio. The methods are the effect-and-probable-cause pie diagram method and the WoE-weighted logistic regression method [173,220]. Both methods yield predictions of local impacts and their probable causes, which provided a statistical ranking of the dominant stressors.

Cross-validation of these models demonstrated that the methods yield significantly similar results in the identification of stressors impacting local fish communities and their relative influence [173]. However, key differences were also observed between the methods that reflected the variance in objectives and sensitivities of each. The findings show that scientific interpretation of eco-epidemiological analysis output requires understanding of method distinctiveness, and suggest the potential value of utilizing multiple methods as lines of evidence in an environmental assessment [173]. Marine studies of sediment contamination using the sediment quality triad found that salinity and grain size, not contamination, predicted benthic community responses best [284]. Obviously greater amounts of colocated physical, chemical, and biological data will provide stronger spatial and temporal characterizations, thus a greater certainty of which parameters have the most significant stressor–response relationships.

Another useful eco-epidemiological model is $SPEAR_{organic}$ (average community sensitivity to organic toxicants) that has been demonstrated to distinguish between the effects of natural longitudinal lotic and organic toxicants using principal components analysis [285]. Benthic richness and diversity responses were linked to petrochemical and synthetic surfactant exposures and separated from natural factors (altitude, velocity, temperature width, nitrate, macrophytes, periphyton cover, substrate size, and habitat heterogeneity). Another study using SPEAR found that runoff potential, stream width, and the presence of clay and dead wood in sediments predicted benthic assemblages [286]. This approach provides another useful tool for separating natural and anthropogenic stressor responses.

Role of biota tolerance

Many aquatic organisms live, eat, and breed within contaminated sediments, and hence they experience chronic exposure over multiple generations. Adaptation to contaminants has been observed after only a few generations in aquatic vertebrates and invertebrates [287]. Estuarine benthic fish that display high site fidelity are chronically exposed to contaminated sediments and may develop tolerance [288]. One such fish, the Atlantic mummichog (*Fundulus heteroclitus*), has been extensively studied and a recent review summarizes the knowledge we have gained from this species [289]. Sediment dwelling annelids have also displayed the ability to evolve tolerance. Vidal and Horne [290] investigated variable resistance to Hg among five populations of the sediment dwelling oligochaete *Sparganophilus pearsei.* Worms collected from contaminated sediments were eight to 10 times more tolerant to Hg exposure in the laboratory than reference site worms. For another sediment dweller, the oligochaete *Limnodrilus hoffmeisteri*, genetic adaptation to Cd-contaminated sediments was shown to occur after only a few generations [287] and is thought to be controlled by a single gene [291]; however, resistance was rapidly lost in nine to 18 generations [292]. Tolerance to contamination can have ecological consequences resulting in structural shifts from tolerant to intolerant communities that may be dominated by nonindigenous species [293]. Moreover, it is crucial that we understand the extent to which resistance to contaminated sediments modifies organism responses to stressor exposure because empirically based sediment guidelines (e.g., effects range low derived from sites where chemicals co-occur), which may be partially based on adapted populations, may be either under- or overprotective. This would depend on the area to which the guidelines are being applied and the chemical of focus [294].

Role of sediment and biota patchiness

A recent review of the benthic community patchiness literature stated that the theories have outpaced experimentation [62], which suggests that while we know patchiness is important, we do not know how to measure its significance. It has been posited that if disturbed community patches are smaller than one meter and benthic invertebrate generation times are less than a year, then recovery from disturbance will be rapid [295]. However, as generation times and patch size disturbance increases, recovery times lengthen. Nevertheless, patchy sediment (or biofilm) contamination at a small scale may be ecologically significant for some communities and driven by habitat matrix retention [296]. Roberts et al. [297,298] found that storm water impacts were highly ephemeral, whereas bioaccumulation of stormwater contaminants by macroalgae had ongoing chronic toxic effects on herbivorous epifauna. Moreover, different macroalgal species accumulated contaminants to different degrees within the same site, creating a patchwork of varying contamination loads that organisms could choose from. We will advance our ability to predict the effects of heterogeneously distributed contaminants as we develop the field of landscape ecotoxicology [299], and this may be possible with existing data sets.

The U.S. EPA performed a national sediment quality survey that reviewed data for 19,398 stations in 5,695 river reaches. Sampling bias tended to occur towards stations that likely had sediment contamination [300]. They identified areas of probable concern for sediment contamination where the exposure of benthic and fish communities would occur frequently. These watersheds had 10 or more sampling stations classified as having probable adverse effects. We reanalyzed this dataset using a much higher spatial resolution (1:100,000) compared with reach file version 1 (RF3) used in the U.S. EPA report (RF3 at 1:250,000 to 1:500,000) to better delineate local spatial variation. The patchiness of Cu- and Zn-contaminated sediments exceeding sediment quality guidelines in the United States was conducted for river reaches (16-digit reach level RF3, National Hydrology Dataset) on which four or more samples occurred (Figs. 2 and 3). River reaches that had one or more samples with SQG exceedances for Cu and Zn, were primarily within the lowest sample exceedance ranges (1–40%), indicative of more localized, patchy, sediment contamination.

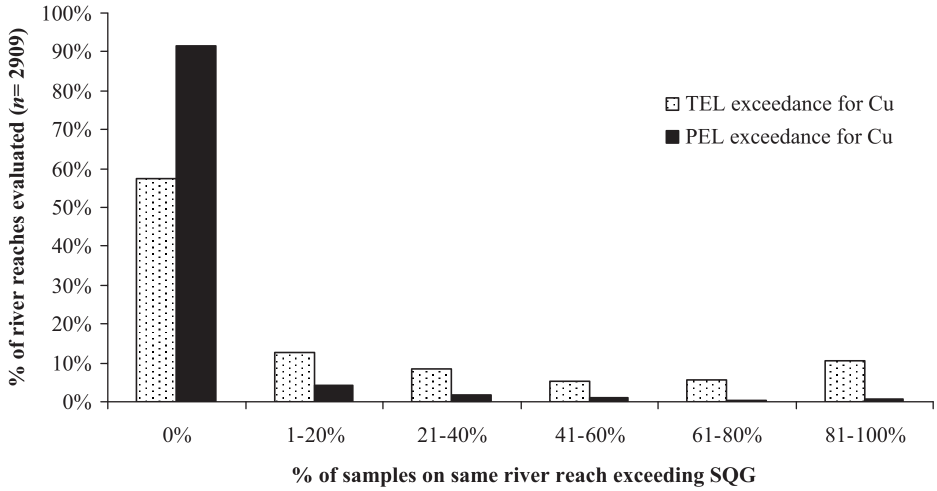

Fig. 2. Distribution of copper sediment quality guidelines (SQG) exceedance rate (per river reach) for all reaches evaluated. Low percentages of samples exceeding an SQG indicate localized (or absence of) contaminated sediment within a given river reach; high percentages indicate more widespread contamination within a given river reach. TEL = threshold effect level; PEL = probable effect level.

An exception was observed at the highest range (80–100% sample exceedance), in which approximately 10% of river reaches had high rates of threshold effect levels exceedances for both Cu and Zn, and 1 and 3% of river reaches had high rates of probable effect levels exceedances for Cu and Zn, respectively. These river reaches are examples of waterbodies potentially dominated by sediment contamination, with a more widespread spatial extent of contaminated sediment as compared to other river reaches. While localized sediment contamination may dominate ecological status on a highly local level, other environmental stressors acting on a wider spatial extent, including habitat quality, flow regime, water chemistry, and point and nonpoint sources of other contaminants, can play a major role in ecological status at the watershed (reach) level. Identification of contaminated sediment as a major (dominant) watershed stressor is, therefore, more convincing for river reaches with high rates (widespread) of SQG exceedances compared with river reaches having low rates (localized) of SQG exceedances. Where contaminated sediments are patchy in their distribution, this will lower their potential to be a dominant stressor in aquatic systems.

IMPLICATIONS FOR REMEDIATION AND RESTORATION

In terms of aquatic ecosystem quality assessment and management, ecotoxicologists, ecologists, and regulators too often have trouble seeing the forest for the trees, with a rather singular, one-at-a-time, focus on individual stressor types (chemical benchmark exceedances, physical disturbance, riparian alteration, flow) or media (sediment). While this can be effective in rather simplistic systems in which one stressor source dominates, it is not practical in complex human-dominated systems where multiple stressors and sources occur. In these multiland use systems, a large number of physical, chemical, and biological stressors are present, with varying exposure regimes, resulting in complex interactions that affect biota directly and indirectly. The biota in these areas may have evolved tolerance to certain stressors (unless development has

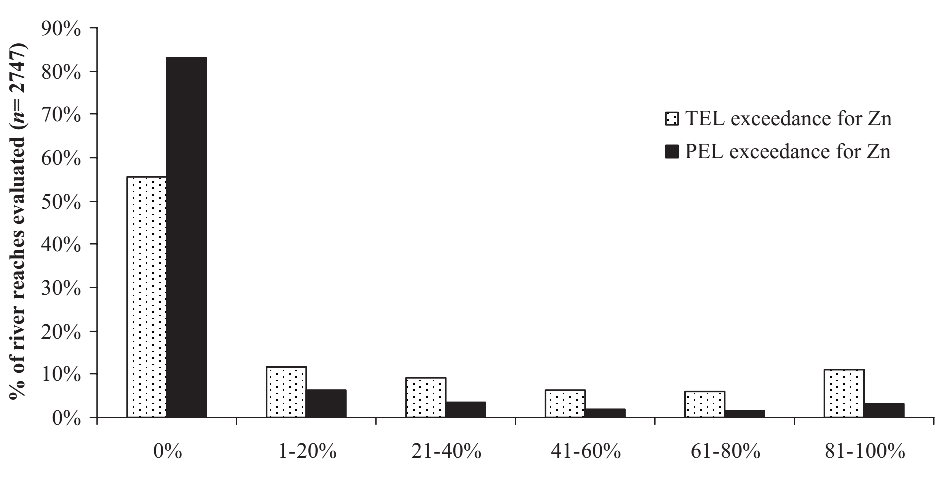

Fig. 3. Distribution of zinc sediment quality guidelines (SQG) exceedance rate (per river reach) for all reaches evaluated. Low percentages of samples exceeding an SQG indicate localized (or absence of) contaminated sediment within a given river reach; high percentages indicate more widespread contamination within a given river reach. TEL = threshold effect level; PEL = probable effect level.

recently occurred) and refugia areas may be limited. Significant difficulties exist in these systems regarding the choice of appropriate reference conditions, spatial-exposure extrapolations, and dominant versus cumulative stressor assessment. Highly focused approaches have not allowed for reliable predictions of how populations and communities will recover if a particular stressor or source is removed from among a plethora of stressor exposures. Yet, it is in these complex environments that billions are being spent on remediation.

Most restoration/remediation efforts in urban dominated watersheds are unsuccessful because important stressors are not removed and/or inadequate refugia exist [4,14,301,302]. Current restoration efforts are directed at single stressor categories and do not take a holistic approach to restoring the biological integrity of our waterways. In many cases, small-scale experimental studies could be used to identify situations in which the reducing the stress associated with physical and chemical attributes of the system are a prerequisite for biological restoration [303]. This process should improve the success of ecosystem restoration efforts, by directing resources towards the most important stressors.

CONCLUSIONS

The ecological risk assessment process for sediments is chemical-centric and does not easily account for major nonchemical stressors in human-dominated aquatic systems or indirect effects. Environmental quality standards or *trigger values* for chemical contaminants may well be protective of ecological impacts. However, habitats that meet these guidelines will not necessarily have good ecosystem quality if other stressors are acting. Increasingly, the importance of co-occurrence of other stressors, both natural and anthropogenic, and their interactions is being realized. Multiple anthropogenic and ecological stressors exert nonadditive effects and unexpected responses. These complex interactions create a high degree of uncertainty surrounding current forecasts of the importance of sediments as stressors.

So when are sediments significant ecosystem stressors? This difficult question is best answered by considering clean versus contaminated sediments in the context of reference conditions and coexisting stressors. This allows for a development of a conceptual model of likely stressors and key populations/communities and sediment properties. If data are missing for possibly important stressors, such as habitat, flow, TSS, or nutrients, then monitoring will be required before appropriate reference conditions can be selected or before accurate decisions can be made regarding the primary causes of ecosystem impairment. If reference conditions cannot be ascertained due to widespread impairment, or there is missing data, it may be possible to use statistical extrapolations, such as Geographic Information System (GIS)-based kriging as a surrogate. Once the key attributes are described, then a stressor ranking analysis (e.g., WoE-GIS) can be performed to guide further experimental manipulations to better establish causality. This, in turn, will allow for more effective decision-making regarding remediation and restoration activities.

For clean sediments, their role as stressors is primarily due to habitat degradation (excessive siltation and scouring) and physical abrasion or clogging (suspended solids). In this review, we do not advocate the management or mitigation of clean sediment stressors unless they are clearly anthropogenically altered in intensity, frequency, or spatial scale. Clean, fine-grained sediments certainly are not stressors in many ecosystems and are useful for restoration purposes. Assessment of anthropogenic influence on these processes is difficult and requires the establishment of suitable well-studied reference locations. In human-dominated watersheds, the erosion of unvegetated or farmed areas often results in increased suspended sediments, while flow energy and flashiness are amplified, affecting large areas of receiving waters. Climate change effects will shift the environmental baseline further and make it increasingly difficult to assess anthropogenic changes to sediment regimes.

Determining when contaminated sediments are significant ecosystem stressors that require management intervention is complex because they usually occur in locations subject to varied levels of anthropogenic modification. We posit that contaminated sediments are more likely to represent a threat to aquatic biota in low-energy, depositional environments. The extent of the impact will be substantially increased if anthropogenic or natural disturbance events act to temporarily resuspend contaminated sediments. In high-energy environments, rapid mixing and dilution of contaminants will result in less intense exposure regimes. The patchiness and relative spatial scale of the contamination needs to be considered in the context of the patchiness and connectivity of the endemic biota. Consideration of the appropriate ecohydraulics is also required to establish the availability of refugia and the likelihood of siltation, deposition, and resuspension.

The relative magnitude and preponderance of contaminated sediments is of particular importance for determining their importance as stressors. Given that we cannot yet quantitatively determine what spatial degree of sediment contamination is significant, it is perhaps more important to establish realistic reference conditions for comparisons and to rank the sediments in terms of coexisting stressors. These many issues and findings all point to the fact that exposures change through space and time, and need to be understood in order to assign causality and determine which stressors pose the greatest risk. Aquatic communities cannot be restored without adequate habitat and food, so these must be considered first in the restoration process. Other stressors (Fig. 1) should be ranked in terms of their relative impact to the receptors and assessment endpoints of concern.

The wide ranging chemically and biologically based methods for assessing when sediments are substantially contaminated have been reviewed elsewhere [5–8,13]. These useful approaches each have their own unique strengths and limitations [1]. Despite the scientific consensus that WoE-based approaches are needed to reliably assess the importance of chemical contamination, regulators continue to rely heavily on point estimates of single chemical exceedances of sediment quality guidelines or probable no-effect concentrations [13]. While this approach is desirable as an easy regulatory bright line of good versus bad, it is flawed in many ways, assuming that: interactions with other nonmetal/synthetic organic stressors are not important; stressor interactions are always additive; resident organisms are continuously exposed to a single chemical concentration (that which is exceeding the SQG) over a chronic period; fluctuating environmental factors do not alter bioavailability or stressor interactions; indirect effects do not occur; and, if the concentration of the chemical of concern is reduced below the guideline, the sediment ecosystem will be clean and ecosystem quality will improve. This latter assumption was recently shown to be erroneous in a National Research Council report documenting the uncertainty of dredging effectiveness at Superfund mega-sites [4,15]. For the same reason that WoE methods are recommended to assess sediment chemical contamination, they must be simultaneously employed to put the sediment contamination in the context of other ecosystem stressors. New lines-of-evidence should be incorporated into ecological risk assessment conceptual models and assessments of human-dominated watersheds and coastal areas. These essential lines-of-evidence should address not only clean and contaminated sediments, but interactions between biota, and critical temporal and spatial threshold conditions for potentially coexisting physical and biological stressors.

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