SETAC PRESS

Environmental Toxicology and Chemistry, Vol. 32, No. 7, pp. 1438–1440, 2013

Printed in the USA

ET&C Impact Papers

ASSESSING SEDIMENT TOXICITY: PAST, PRESENT, AND FUTURE

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(Submitted 25 February 2013; Accepted 25 February 2013)

Sediments have a critical role in biogeochemical cycling, and benthic processes often support the base of aquatic food chains. The Anthropocene era has seen dramatic increases in the availability of nutrients, metals, and synthetic organics released into human-dominated watersheds, with evidence of global transport. Many of these chemicals are retained in sediments, which are the largest repository of chemicals globally. Sediments and their stored chemicals are not static with erosion and fluvial processes continually pumping sediments and their associated chemicals into depositional areas of streams, rivers, lakes, reservoirs, and coastal areas. These processes are likely to intensify as more extreme climate events occur in many parts of world. The US Army Corps of Engineers removes several hundred million cubic yards of dredged materials per year, with much of it simply being moved from channels to adjacent aquatic areas. This suggests that tens to hundreds of billions of cubic yards of sediments worldwide must be moved annually for navigation purposes, adding to the overall transport of sediments through our urban and agricultural landscapes. Thus the depositional and sorptive nature of fine-grained sediments has created hundreds of thousands, if not millions, of contaminated sites. Assessing the ecological impact and risk of these sediments has been challenging from both scientific and regulatory perspectives, and never before has it been more important to get it right.

THE PAST

In the United States, there was a period of rapid development of sediment assessment methods in the 1980s to mid-1990s, driven by regulatory attention and funding. One of the classic sediment toxicity methods papers, which guided further development, was Nebeker et al., published in 1984 [1]. This increased focus also occurred in Canada, Europe, Australia, and New Zealand in the 1990s. Several standardized toxicity test assays were developed by regulatory and standards-setting institutions, along with guidance on how to sample, handle, and characterize sediments [2–5]. A number of sediment quality guidelines based on equilibrium partitioning and empirical data were created [6,7]. Publications and presentations on chemically contaminated sediments dramatically increased. All of this activity was fueled by a host of regulatory mandates to assess and remediate contaminated sites. As is the case whenever there are potentially expensive regulatory drivers, a flurry of activity and some controversy followed regarding how to best address these mandates. Some of the controversy did not deal with the science, but rather policy issues, for example, whether we should call these newly devised values sediment quality standards, indicators, criteria, guidelines, benchmarks, alert, target, or quality levels, and so on. These labels represented differences that were subtle to many, yet huge to others. Many scientific points were widely discussed and published, such as the validity of the chemical-specific guidelines, optimal measurement endpoints, sampling-induced artifacts, which species were most sensitive and discriminatory, when bioaccumulation was at steady-state, how many bioassays were enough, which assessment methods or suite of methods should be used, how were decisions made at the end of the process, and what really is weight-of-evidence.

Useful assessment tools were developed that went beyond sediment quality guidelines and laboratory sediment toxicity testing [8,9]. The US Environmental Protection Agency (USEPA) led efforts over several years to develop toxicity identification evaluation (TIE) methods for sediments, first focusing on porewaters, and then on whole sediments [10]. These methods could detect acute toxicity in fractions linking to ammonia, metals, and nonpolar organics. In situ exposure approaches for TIE allowed for more sensitive detection of porewater toxicity, but testing was limited to a few shallow freshwater systems [11]. In situ approaches also revealed photo-enhanced toxicity of polycyclic aromatic hydrocarbons (PAHs). In streams with PAH-contaminated sediments, increased toxicity was observed during low-flow sunny days, whereas toxicity was removed during high-flow, turbid events [12]. Given that bioavailability changes with site conditions and benthic populations, a promising approach (benthic assessment of sediment quality [BEAST]) was developed that statistically identified those relationships across the Great Lakes, allowing for clear designations of reference condition and degrees of impairment [13].

In the period of 1995 to 2005, the assessment process for sediments matured and best practices converged. Several countries developed sediment quality guidelines and guidance for managing contaminated sediments. Two notable expert workshops were sponsored by SETAC and resulted in state-of-the-science books in 1997 and 2005 [14,15]. The 2005 publication focused on the most widely used assessment tool—sediment quality guidelines—while discussing other methods. A framework was proposed that acknowledged the importance of developing accurate conceptual models streamlined for widely varying hydrologic systems, and using a multi-assessment, weight-of-evidence–based approach. The sediment quality guidelines were recommended as a tier 1 screening approach that explicitly recognized their strengths and

All Supplemental Data may be found in the online version of this article. See Table S1 for the number of citations and rank of all the "Top 100" papers, which in this essay are references [1,6–9].

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Published online in Wiley Online Library (wileyonlinelibrary.com).
DOI: 10.1002/etc.2250

limitations. Despite these advances in understanding sediment contaminant exposure and receptor effect relationships, most regulation-motivated assessment and remediation efforts were driven by a few single chemical guideline exceedances that did not account for bioavailability or accurately determine exposures.

As more and more contaminated sediment sites were identified and slated for remediation, it was apparent that the problem and the fix were often extremely expensive and controversial. For example, at the Coeur d'Alene site in Idaho and Washington (USA), there are millions of cubic yards of contaminated sediments and tailings. The most recent cost estimate for partial remediation is US $1.4 billion. The Hudson River (New York, USA) is currently being remediated to remove polychlorinated biphenyl (PCB)-contaminated sediments, and costs may approach US $1.5 billion. These stark realities were the driving force behind Congress requesting the US National Research Council to study dredging effectiveness at megasites [16]. This report found that virtually none of the large remediation projects in the United States had adequate pre- or postmonitoring to determine remediation effectiveness. Clean-up goals were simply driven by removal of sediment mass, and limited documentation existed to show that a clean-up PCB concentration was met. The same report recommended, among other things, that pre- and postmonitoring be conducted at all contaminated sediment megasites to evaluate remedy effectiveness, that monitoring of benthic organisms and using passive sampling devices were better than fish as indicators of ecologic effects, and that the research and development of rapid field monitoring techniques and benthic organism methods were needed.

THE PRESENT

As noted, only a handful of new sediment toxicity bioassays have been developed in the past 20 yr. There are still surprisingly few standardized bioassays for benthic species compared with pelagic species. For example, in the past decade the American Society for Testing and Materials has developed freshwater sediment toxicity standard methods for mussels (E2455) and amphibians (E2591), and has updated or is updating existing methods for other freshwater, estuarine, and marine invertebrates. No new test species have been standardized in the past decade in Canada, Australia, or New Zealand, or by the OECD. Part of this inactivity can be explained by a scarcity of research funding by governmental agencies for exploratory studies of contaminated sediments. An additional factor that may be slowing the development of new benthic bioassays is the fact that few benthic species can be cultured easily. However, freshwater unionid mussels and snails have recently been used in sediment toxicity assays. Unionid mussels are difficult to culture, but their early life stage is both very sensitive and of ecological importance, as many mussel species are threatened, endangered, or have gone extinct. The limited suite of sediment toxicity assays is problematic for the assessment and regulatory processes, such as developing species sensitivity distributions from which sediment guidelines are derived (e.g., predicted no effect concentrations [PNECs]), or extrapolating findings to widely varying benthic populations and communities.

Sediment quality guidelines have also largely remained unchanged in the past 2 decades. The exception is that new PNECs are currently being developed as part of the European Commission's Registration, Evaluation, Authorization, and Restriction of Chemicals (REACH) and Water Framework Directive programs. Development of these standards has been challenging, as scientists now have a better understanding of bioavailability and the dynamic changes possible in sediment physicochemical conditions. A better understanding of metal spiking issues and dynamics allowed for improved determinations of metal threshold levels for Cu and Ni [17]. Recent field and laboratory studies are showing the importance of considering iron and manganese hydrous oxides for controlling the bioavailability of divalent cationic metals such as Cu, Zn, and Ni [18]. These fractions dominate the sediment–water interface in oxic waters, where acid-volatile sulfides are often nonexistent. This process is accentuated by periphyton, which causes daily dissolved oxygen and pH shifts to alter metal speciation and release in and out of surface waters and underlying sediments [19].

As in water, no sediment guidelines exist for contaminants of emerging concern (CECs), such as pyrethroids, polybrominated diphenyl ethers, pharmaceuticals, and personal care products. Many of the CECs, like most chemicals, tend to sorb to solids and therefore will accumulate in sediments. Pyrethroids are of particular concern for sediments in human-dominated watersheds due to their propensity to sorb to particles, degrade slowly in sediments, and are extremely toxic [20]. Limited research has delved into the impacts of sediment-associated nanomaterials due to the confounding issues involving quantification and matrix interferences.

Another critical aspect of sediment assessments that is presently being studied is that of resuspension of contaminated sediments. It is somewhat perplexing that, although many of the contaminated sediment sites in the world are in harbors, where resuspension is a frequent daily event, little is known about the ecological consequences of these events. Perhaps tackling the issue has been too daunting—given that bedded sediments are inherently complex and the addition of resuspension would introduce even more spatial and temporal complexity. The literature suggests that these events are toxic, yet most studies have been conducted under laboratory conditions that poorly simulate reality. Under more realistic conditions, initial findings suggest that released chemicals are quickly recomplexed. When this process is paired with exposure periods of minutes to hours, the resulting biological effects appear small. Of greater concern may be the settled, rebedded sediments that may smother early life stages or have greater concentrations of contaminants available for epibenthic feeding organisms.

THE FUTURE

It has been 22 yr since my review of freshwater sediment toxicity testing [8]. The science and its use in managing contaminated sites have improved in many ways but there is a long way to go before such testing may be considered straightforward, efficient, and effective. Many still consider contaminated sediment assessments too complex and expensive and revert back to managing based on concentrations of a single chemical, or cubic yards of sediment removed, or both. Sadly, this approach is both inefficient and ineffective, wasting millions of dollars again and again, and not significantly protecting and restoring our ecosystems.

Continued growth of urban areas and megacities is predicted [21]. This growth will be greatest in developing countries, where wastewater infrastructure is lacking. Nevertheless, even in the United States, infrastructure receives a D+ from the American Society of Civil Engineers, and billions of gallons of raw sewage spill into the Great Lakes (and other waterways) each year through combined sewer overflows [22]. Most of our water quality problems are due to nonpoint source pollution, which is associated with all human-dominated watersheds. I have never studied a contaminated sediment site where stormwater inputs did not contribute to contamination. All of this suggests that contaminated sediments will increase in importance as our populations and economies grow. It will be increasingly difficult to remove and dispose of contaminated sediments due to the cost and sheer magnitude of the problem. This will lead to greater contamination of food webs and fish. The rising use of CECs in the population and their deposition to sediments will elevate concerns for ecological and human health. The impacts of climate change will continue to unfold and have already resulted in more extreme events in the Great Lakes, resulting in greater nutrient and sediment runoff, massive harmful planktonic and benthic algal blooms, and lake hypoxia. Nutrients accumulating in sediments will contaminate overlying waters for years to come.

These interwoven human impacts are becoming all too common around the world, and suggest that we cannot simply focus resources and regulatory efforts on single chemicals [23]. The human-dominated waterways where these chemicals have accumulated have a multitude of other stressors that are primary drivers for ecological processes. Effective restoration is not possible without improved assessments and feasibility and remediation approaches that rank and deal with the stressors that matter most.

Bringing together the thoughts of many colleagues, I offer the following suggestions for improving the sediment assessment: 1) create a better linkage of spatial and temporal exposure with effects, thereby reducing uncertainty in hazard and risk assessments; 2) improve field method sensitivity, discriminatory power, and practicality; 3) use context-based assessments [24] that link key sensitive indigenous receptors with stressors; 4) consider ecosystem stress from habitat, nutrients, solids, and hydrology in the context of water column (baseflow and stormflow) and sediment contaminants; and 5) validate efficient biomarker relationships to population and community effects. Currently, the field of sediment ecotoxicology is addressing these issues, and noteworthy advances are more evident with each passing year. The more looming challenge may be in translating this science for regulators and in improving ecosystem management policy.

SUPPLEMENTAL DATA

Table S1. (49 KB PDF).

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