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TOXICITY OF SEDIMENTS

Overview

As water quality has improved over the past three decades in North America, diffuse sources of pollution such as stormwater runoff and sediments are now recognized as long-term, widespread pollutant sources to aquatic systems. Substantial impacts on the ecosystem from sediment-associated contaminates range from direct effects on benthic communities to substantial contributions to contaminant loads and effects on upper trophic levels through food chain contamination (e.g., McCarty and Secord, 1999).

Sediment contamination primarily occurs because many chemicals bind to organic or inorganic particles that eventually settle to the bottom of our streams, rivers, reservoirs, lakes, estuaries, or marine waters. Once contaminants bind to a particle surface or sorb into its interior matrix, they become less likely to be biotransformed and desorption is usually very slow; therefore, sorbed contaminants will reside for long periods in the sediment. Sediment-associated contaminants tend to accumulate onto small, fine-grained particles and settle in depositional areas. This is promoted largely by the very high surface area and the tendency for higher concentrations of organic matter in the fine particles.

From a contamination perspective, we understand that sediments are extremely important to the food web and serve as a habitat for the benthic community as well as contaminant reservoir for bioaccumulation and trophic transfer. Once chemical contamination reaches a concentration whereby it causes adverse effects to biota, then it is considered polluted. The greater the evidence of sediment contamination within a watershed, the more likely a fish consumption advisory exists (US EPA, 1997).

Extent of the problem

There are over 9 billion cubic meters of surface sediments (upper 5 cm) in the United States of which approximately 1.2 billion can currently be considered contaminated to levels that pose potential risks to fish and to humans and wildlife who eat fish (US EPA, 1997). This estimate is derived from a literature search on sediment contamination and represents 65 percent of the watersheds in the continental US. However, only 11 percent of the nation's rivers had data with any toxicity information available and of those 77 percent had at least one station that was sufficiently contaminated such that adverse effects are probable or possible. This leaves 89 percent of the data sets with no available toxicity data (US EPA, 1997). Therefore, the true extent of sediment contamination remains unknown.

Sediment associated contaminants are found in every type of aquatic environment within the US from mountain streams to large rivers, from small lakes to the Great Lakes and in estuaries and bays (US EPA, 1997). Further, recent investigations have found sediment pollution in areas previously not thought to be contaminated (e.g., estuarine areas of North Carolina, Pelley, 1999). While the true extent of sediment contamination is not known, it is apparent that huge quantities of sediments in industrialized countries are contaminated with metal and organic chemicals at levels that pose risks to both aquatic life and to humans consuming aquatic species.

Effects on ecosystems and components

Despite a concentrated research effort over the past two decades, our ability to assess the impact of contaminated sediments on ecosystems remains a challenge that is inherent in the very nature of sediments. The most obvious effect in most assessments of sediment contamination is that of a direct adverse impact on the benthic (or bottom-dwelling) community, (e.g., Peterson *et al*., 1996). Because alteration of the benthic community structure can result from the mixture of contaminants present as well as from potential for confounding factors, such as poor habitat, high flows, or low dissolved oxygen in the water column, it is often difficult to establish cause and effect for sediment associated contaminants. Studies of field sites where direct biological effects are obviously contaminant-induced, are often located in watersheds containing high levels of industrial and shipping activities. Here, sediments are likely to be visibly contaminated with petroleum products and may contain a myriad of metals, pesticides, and other synthetic non-polar organic chemicals, e.g., Grand Calumet River, Indiana. Thus, assigning cause-effect between single, specific chemicals and benthic communities becomes nearly impossible. Most of the detailed biological assessments dealing with contaminated sediments have dealt with freshwater systems, although several sediment risk assessments are currently underway in coastal harbor areas. Unfortunately, most of these risk assessments are only published in the "grey" literature and difficult to obtain.

It is not just infaunal (burrowing) benthic invertebrate organisms that demonstrate effects from sediment-associated contaminants. Many bottom-feeding fish have hepatic and epidermal neoplasms (tumors). There is a strong link between contaminated sediment and fish neoplasms in all the major water bodies of the US (Harshbarger and Clark, 1990). The sediment link for these neoplastic growths was clearly demonstrated in the Black River in Lorain, OH. The high incidence of neoplasms in brown bullhead showed a high correlation with high PAH sediment concentrations. These neoplasms resulted in age specific mortality in the bullheads. Once most of the PAH contaminated sediments were removed by dredging, the incidence of neoplastic growths declined sharply (Baumann and Harshbarger, 1995).

Sediment-associated contaminants have also become linked via food web transfer to impacts on upper trophic levels. Such transfer occurs with mercury and some organochlorines, such as PCBs and DDT that are poorly biotransformed and are hydrophobic; however with other chemicals these connections are more difficult to establish. From modeling exercises, food web transfer of persistent contaminants is important for maintaining the chemical concentrations observed in upper tropic levels and the benthic component is essential to account for the observed concentrations (Thomann *et al*., 1992; Morrison *et al*., 1996). Trophic transfer of sediment-associated contaminants has been documented in both freshwater systems (e.g., Lester and McIntosh, 1994) and marine systems (e.g., Maruya and Lee, 1998). In Saginaw Bay, Lake Huron, tree swallows were found to accumulate PCB from sediments. In some areas of the Great Lakes and in the Hudson River, NY, system reproductive damage has been observed for this species directly linked to PCB (Bishop *et al*., 1999; McCarty and Secord, 1999). Further, impacts to birds were observed in the Saginaw Bay system with reproductive effects on the Caspian terns following a 100 year flood event suggesting impacts through food web transfer from a latent sediment source (Ludwig *et al*., 1993). Thus, fresh pulses of contaminated sediments from the watershed can recharge the system resulting in impacts on the receiving system.

Developing cause–effect links

The complexity of the distributions of contaminants, nutrients, other sediment/habitat characteristics and resident biota make accurate determinations of exposure and effects difficult, but not impossible. This reality suggests that accurate assessments of contaminant effects use a weight-of-evidence approach (i.e., gathering supporting data from several lines of investigation) to determine the extent or magnitude and cause of the contamination problem. The initial ideas on the weight-of-evidence approach took data from sediment chemistry, benthic community structure and sediment laboratory bioassays to establish a triad of information that could be used to determine hazard (Chapman, 1986). This approach is continually improving with additional development on sediment bioassays (toxicity tests), interpretability of sediment chemistry, and the addition of new types of information such as *in situ* toxicity tests and residue (tissue chemical concentration)-effects information.

Sediment toxicity testing on whole field collected sediments and laboratory dosed sediments are an essential line-of-evidence that the contaminants in sediment can produce the effects that are observed in benthic communities. The earliest sediment bioassay was performed in the early 1970s and demonstrated avoidance behavior by amphipods for contaminated sediments. However, much of the work demonstrating the effect of sediment-associate contaminants came in the 1980s. By the mid 90's, standardized methods for whole sediment toxicity testing occurred within the US EPA, American Society for Testing and Materials (ASTM), and Environment Canada. These tests measured acute (short-term $\leq$10 days) toxicity in benthic macroinvertebrates such as the amphipods *Hyalella azteca, Rhepoxynius abronius, Ampelisca abdita, Eohaustorius estuarius* and *Leptocheirus plumulosus* and the midges *Chironomus tentans* and *Chironomus riparius* (US EPA, 1994a,b; ASTM, 2000a). The primary measured response was mortality, but in the case of the midge, growth was included and reburial was an additional endpoint for the *Rhepoynmius abronius*. Unfortunately, most sediment toxicity test methods are focused on acute (short-term exposure) and not chronic (long-term exposure) toxicity. The measures of acute toxicity are not often adequate to detect the impacts on benthic communities. For instance, the 10-d test with *Rhepoxynius abronius* was not sensitive enough to describe the loss of amphipods from Lauritizen Channel in San Francisco Bay (Swartz *et al*., 1994). In reality, chronic toxicity is the more pervasive problem and it is the chronic responses, such as changes in reproduction that lead to population level responses. While recently developed chronic methods (US EPA, 2000, 2001) will greatly aid our ability to determine if sediments are toxic, their long duration and increased costs may impede their widespread adoption.

A large number of other species have been used for determining the toxicity of sediments, ranging from bacteria to fish and amphibians (Burton, 1991). The commonly used test species encompass trophic levels ranging from decomposers and producers to predators and representing multiple levels of biological organization. Comparisons of their sensitivities have shown a wide range of responses to different types of sediment contamination, with an equally wide range of discriminatory power (ability to detect differences between samples) (Burton *et al*., 1996). This reality suggests that more than one or two species may be necessary to determine with certainty whether or not sediment contamination is ecologically significant.

The freshwater species that have been used successfully and most often in assessments of sediment toxicity include both water-column and benthic species: *Selenastrum capriconutum, Daphnia magna, Ceriodaphnia dubia, Pimephales promelas, H. azteca, C. tentans* and *C. riparius*, and *Hexagenia limbata*. Standard guides for whole sediment toxicity testing have also been published by the American Society for Testing and Materials for invertebrates, including *H. azteca, C. tentans, C. riparius*, the cladocerans *Daphnia magna* and *Ceriodaphnia dubia*, the mayfly *Hexagenia*, the Great Lakes amphipod *Diporeia*, and the oligochaete *Tubifex tubifex* (ASTM, 2000a–e). Dredged material evaluations in the US follows the protocols published by the US EPA and US Army Corps of Engineers; in 1991 for dredged material proposed for ocean disposal, and protocols published in 1998 for dredged material proposed for disposal in ‘‘inland’’ waters of the US (USEPA–USACE, 1998). Canada’s Ontario Ministry of the Environment has also published solid-phase test methods (Bedard *et al*., 1992).

Toxicity tests for marine and estuarine species also range from fish to bacteria. The marine and estuarine species frequently and successfully used in studies of sediment contamination include: the fish *Cyprinodum variegatus* and *Menidia beryllina*, and amphipods *Rhepoxynius abronius, Ampelisca abdita, Eohaustorius estuarius, Grandidierella japonica* and *Leptocheirus plumulosus, the* bivalves *Haliotis rufescens*, *Crassostrea gigas* and *Mytilus* sp. the echinoderms *Strongylocentrotus purpuratus, Dendraster excentricus*, and *Arbacia punctulata*, the polychaetes *Dinophilus gyrociliatus* and *Neanthea arenaceodentata*, the mysid *Americamysis bahia* (formerly *Mysidopsis bahia)*, the algal *Champia parvula*, and the bacterial assay Microtox using *Vibrio fischeri*. As with freshwater tests, several ‘‘standard’’ methods for marine and estuarine organisms exist. The US EPA has standardized protocols for toxicity testing with the amphipods *Rhepoxynius, Ampelisca, Eohaustorius, and Leptocheirus* using a 10-d survival assay, a chronic 28-d survival, growth reproduction assay with *Leptocheirus*, and 28-d bioaccumulation assays with *Macoma* and *Nereis* (US EPA 1994b, 2001). The European OECD (Organization of Economic Cooperation and Development) is discussing the possibility of developing standardized sediment tests. Environment Canada has developed methods for sediments, elutriates, and leachate samples using *Vibrio fisheri* luminescence, sea urchins and sand dollars (echinoids), and seven amphipod species. Methods were recently published by the Chesapeake Bay Program for benthic testing using *Lepocheirus plumulosus, Ampelisca abdita, Lepidactylus dytiscus, and Monoculodes edwardsi*. Dredged materials in the US is tested using US EPA and US Army Corps of Engineers protocols. The American Society for Testing and Materials has standard procedures for amphipods, fish, mysids, echinoderm, and oyster early life stages, polychaetes, and algae (ASTM, 2000b–e).

Another component that is useful in establishing causality is linking sediment contaminant exposure and tissue-residue effects in organisms. Because many factors appear to alter the bioavailability of contaminants in sediments (Hamelink *et al*., 1994), approaches to establish links between the body-residue concentrations and effects in aquatic organisms provide the insight to better link the toxic response directly to contaminants. Data has been amassing over the course of the past several years that allow the direct comparison of some residue levels with acute and chronic effects (www.wes.army.mil/el/ered; Jarvinen and Ankley, 1999). However, the data base is very limited at this time, thus there is still need to establish a weight-of-evidence approach for developing the link between the observed response and the presence of contaminants in sediments.

The second recent development to assist in the formation of the link between sediment contaminants, responses in the laboratory and observed responses in the field is that of *in situ* tests. These are tests that are conducted in the field. One approach uses confined organisms, such as traditional surrogates (e.g., *Daphnia magna, Ceriodaphnia dubia, H. azteca, C. tentans, Lumbriculus variegatus, Pimephales promelas*) or indigenous species in chambers, cages, bags, or corrals. Organisms are placed in test chambers, which optimize exposures to sediments, pore waters, and/or overlying waters, and are exposed in the field from time periods ranging from a day to months. This approach has a number of advantages, in that it can better simulate real-world exposures, which may fluctuate dramatically over a period of hours, reduces sampling/experimental related artifacts, and allows for more natural interactions of potentially critical physical and chemical constituents than do laboratory conditions (e.g., Chappie and Burton, 2000).

Traditionally sediment contamination was determined by assessing the bulk chemical concentrations of individual compounds often comparing to some background or reference value. The desire to have chemical parameters to evaluate the hazard of sediments has focused primarily on two approaches: (1) a statistical approach to establish the relationship between sediment contamination and toxic response; and (2) a theoretically based approach that attempted to account for differences in bioavailability through equilibrium partitioning.

The original sediment quality guidelines (SQGs) that were compared to a reference or to background provided little insight into the impact of sediment contaminants. Therefore, SQGs for individual chemicals were developed that relied on paired field sediment chemistry with field or laboratory biological effects data. They are often based on frequency distributions and account for the impact of all chemicals present, but do not establish cause and effect. These approaches have also been shown to be useful and predictive of biological effects in many (but not all) marine and freshwater systems (e.g., Long *et al*., 1998).

However, the issue of bioavailability remains (that portion of the chemical that is available for biological uptake and subsequent adverse effects). The equilibrium partitioning approach attempts to address this issue specifically. This approach suggested that interstitial water concentrations represented the equivalent chemical activity for exposure of aquatic organisms, and chemical concentrations could be determined from an equilibrium partitioning calculation (Di Toro *et al*., 1991). The equilibrium partitioning calculation was particularly important for attempting to account for the changes in bioavailability that occur with changes in amounts of organic matter in sediments.

This method has also been applied to metals by accounting for the interactions with acid volatile sulfide. Five metals, Cd, Ni, Pb, Zn, and Cu, form insoluble sulfides and so their toxicity is limited by the amount of sulfide in the sediment. Toxicity is observed when the amount of metal stoichiometrically exceeds the amount of sulfide that can bind it. A clear demonstration of the utility of this approach was shown for Cd toxicity to amphipods, *Ampelisca abdita* and *Rhepoxynius hudsoni* in marine sediments. Like the case for organic contaminants, the toxicity predicted from this approach has been sometimes over predicted usually because of the presence of other ligands that bind the metals (Ankley *et al*., 1993).

When comparisons have been made between the equilibrium-partitioning (EQP) method and other measures of sediment quality to estimate toxic response, the sediment quality guideline values and their predictive ability were comparable, i.e., in greater than 70 percent of samples toxicity or absence of toxicity were correctly predicted. However, when comparing the predictive ability for field sites, EQP consistently gave more type II errors, false negatives, than other sediment quality guidelines (Ingersoll *et al.*, 1997). This likely results from failure to account for the wide variety compounds that exist in sediment and their interactions.

Risk-assessment for sediment-associated contaminants is a relatively new field and is developing rapidly (Ingersoll *et al.*, 1997). The strengths and limitations of the various assessment methods dictates that a weight-of-evidence approach define the significance and role of sediment-associated contaminants on the aquatic environment and also through food web transfer to terrestrial species. This approach should include an integrated approach, which identifies and ranks stressors using habitat, physical, and chemical measures, biological community structure, toxicity, and bioaccumulation descriptors. Use of these tools can provide essential characterizations of key watershed sources, sensitive receptors, natural variation, and both natural and anthropogenic stressors. Only with the use of multiple tools and an understanding of their interactions can reliable determinations of sediment pollution and long-term consequences be made. Then cost-effective, environmentally protective management decisions can be made about the type, extent, and need for sediment remediation.

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