SETAC PRESS

Environmental Toxicology and Chemistry, Vol. 34, No. 1, pp. 6–21, 2015

Printed in the USA

Critical Review

CRITICAL REVIEW OF MERCURY SEDIMENT QUALITY VALUES FOR THE PROTECTION OF BENTHIC INVERTEBRATES

JASON M. CONDER,*† PHYLLIS C. FUCHSMAN,‡ MELISSA M. GROVER,†
VICTOR S. MAGAR,§ and MIRANDA H. HENNING‖
†ENVIRON International, Irvine, California, USA
‡ENVIRON International, Burton, Ohio, USA
§ENVIRON International, Chicago, Illinois, USA
‖ENVIRON International, Portland, Maine, USA

(Submitted 7 February 2014; Returned for Revision 29 March 2014; Accepted 1 October 2014)

Abstract: Sediment quality values (SQV) are commonly used—and misused—to characterize the need for investigation, understand causes of observed effects, and derive management strategies to protect benthic invertebrates from direct toxic effects. The authors compiled more than 40 SQVs for mercury, nearly all of which are "co-occurrence" SQVs derived from databases of paired chemistry and benthic invertebrate effects data obtained from field-collected sediment. Co-occurrence SQVs are not derived in a manner that reflects cause–effect, concentration–response relationships for individual chemicals such as mercury, because multiple potential stressors often co-occur in the data sets used to derive SQVs. The authors assembled alternative data to characterize mercury-specific effect thresholds, including results of 7 laboratory studies with mercury-spiked sediments and 23 studies at mercury-contaminated sites (e.g., chloralkali facilities, mercury mines). The median (± interquartile range) co-occurrence SQVs associated with a lack of effects (0.16 mg/kg [0.13–0.20 mg/kg]) or a potential for effects (0.88 mg/kg [0.50–1.4 mg/kg]) were orders of magnitude lower than no-observed-effect concentrations reported in mercury-spiked toxicity studies (3.3 mg/kg [1.1–9.4 mg/kg]) and mercury site investigations (22 mg/kg [3.8–66 mg/kg]). Additionally, there was a high degree of overlap between co-occurrence SQVs and background mercury levels. Although SQVs are appropriate only for initial screening, they are commonly misused for characterizing or managing risks at mercury-contaminated sites. Spiked sediment and site data provide more appropriate and useful alternative information for characterization and management purposes. Further research is recommended to refine mercury effect thresholds for sediment that address the bioavailability and causal effects of mercury exposure. *Environ Toxicol Chem* 2015;34:6–21. © 2014 SETAC

Keywords: Sediment quality guidelines (SQGs) Benthic toxicity Mercury Empirical SQGs

INTRODUCTION

Most major regulatory programs that govern sediment site investigation and sediment management require characterization of risks to benthic invertebrates [1–4]. Predicting sediment toxicity with a reasonable level of confidence is critically important in benthic invertebrate risk assessments because risk managers use information from such risk assessments to inform risk-management decisions that can affect many aspects of the existing aquatic communities. Conceptually, risk-management decisions are intended to protect key elements of the food web (e.g., benthic invertebrates), if warranted. Overprotection should be avoided, however, because invasive sediment management practices (e.g., dredging, capping) can disrupt existing biological communities, potentially to a greater extent than the impairment caused by the chemicals of concern, and at a high financial and social cost [5,6].

Risks posed to benthic invertebrates often are initially screened by comparing chemical concentrations in sediment with sediment quality values (SQVs, also known as sediment quality guidelines or sediment quality benchmarks). Often, SQVs are developed using paired toxicity and chemistry data and typically are reported as a lower bound statistic that is believed to be associated with the absence of an effect (i.e., no effect) and an upper bound statistic that is believed to be associated with an adverse effect [5,7]. Over the past 2 decades, several dozen SQVs have been developed to help characterize the potential for adverse effects of chemicals on benthic invertebrates [3,5,8–13]. Although most authors of such SQVs report that their intended use is for screening evaluations, the use of SQVs has been extended to a variety of decision-making steps associated with the management and regulation of dredged materials and sediments, including the determination of dredged material disposal options, cleanup and monitoring of sediments, and in support of total maximum daily load development or other source reduction approaches.

Often, SQVs are applied in multiple lines of evidence decision-making frameworks, such as the sediment quality triad [14]. This decision-making tool also considers toxicity testing and benthic community surveys in determining the severity of sediment contamination and potential adverse biological effects. In this context, comparisons of measured chemical concentrations in site sediments to SQVs are used as a line of evidence to identify the potential for toxic effects or impairment of benthic community composition at the site [5,7,15,16], although in some cases they can be misapplied within such analyses to attribute potential causality to single chemicals.

The present study compiles and reviews SQVs that have been developed for characterizing mercury risks to benthic invertebrates. Mercury is a naturally occurring element that has been released from geologically stable forms into the environment through human activities. The methylated form of mercury

All Supplemental Data may be found in the online version of this article.
* Address correspondence to jconder@environcorp.com.
Published online 15 October 2014 in Wiley Online Library (wileyonlinelibrary.com).
DOI: 10.1002/etc.2769

biomagnifies through the food web, and much attention has focused on its potential effects on humans and wildlife that consume aquatic organisms. Mercury also has the potential for direct toxic effects on lower trophic levels, including benthic invertebrates. The majority of the compiled SQVs for mercury were derived using co-occurrence methods, which are calculated from databases of paired sediment chemistry and (usually) lethal effects data [15]. A small number of SQVs have been developed for mercury using a variation of equilibrium partitioning (EqP) methods, which have the potential to incorporate single-chemical cause–effect information [17–19]. For comparison, we also review data relevant to understanding causal relationships between mercury concentrations and sediment toxicity, specifically spiked sediment toxicity studies and biological investigation results from sediment sites where mercury is a predominant contaminant. Although we do not derive a specific alternative SQV for mercury, the present review summarizes the current understanding of sediment exposure–response relationships for mercury that can inform effective investigation and management of mercury risks to benthic invertebrates.

REVIEW OF SQVs FOR MERCURY

We compiled SQVs from multiple reviews of SQV approaches [3,5,12,15,20] and other sources. We identified 40 co-occurrence SQVs for mercury (Supplemental Data, Table S1). The definitions of these SQVs vary; some are intended to predict a lack of toxicity ("low SQVs"), whereas others are intended to predict probable, moderate, or severe effects ("high SQVs"). We have maintained the classification of co-occurrence SQVs in 2 qualitative categories. The first category includes SQVs equivalent to concentrations of mercury that have a minimal probability of co-occurrence with adverse effects, classified as "low"; these values are generally associated with terms such as "threshold," "probable no effect," or "low effect." Generally, low SQVs represent the concentration in sediment that is associated with the 5th to 10th percentile of concentrations found in toxic sediments in the study (i.e., 5–10% of the toxic sediments in the respective databases exhibit concentrations below the SQV). Most authors note that sediments exhibiting concentrations below low SQVs rarely exhibit observed effects and often assume sediments with concentrations less than low SQVs to be "safe" or nontoxic [15,21,22]. It is not possible to express the probability of toxicity (or degree of effect) for a sediment that exceeds a low SQV value. However, consistent with the intended use of low SQVs, many sediments exhibiting concentrations above these SQVs also do not exhibit adverse effects [15,22].

The second category includes SQVs equivalent to concentrations of mercury that have a moderate or high probability of co-occurrence with adverse effects, classified as "high"; these values are generally associated with terms such as "median range," "probable," "moderate," "occasional," "apparent effect," or "severe effect." Generally, high SQVs represent the concentration in sediment that is associated with the 50th to 80th percentiles of concentrations found in sediments exhibiting adverse effects in the study (i.e., 50–80% of sediments in the respective databases with adverse effects exhibit concentrations below the high SQV). Most authors note that adverse effects are often expected when high SQVs are exceeded [15,21–23]. This low/high classification scheme has been used elsewhere (e.g., MacDonald et al. [12]) to distinguish SQVs designed to minimize false-negative predictions (low SQVs) from those intended to approximate toxic thresholds (high SQVs).

Consensus SQVs [12] have also been developed as aggregations of multiple individual SQVs; in the case of mercury, these are based on aggregations of co-occurrence SQVs. Additionally, a small number of SQVs for mercury are based on an adaptation of equilibrium partitioning theory, whereby water quality values are extrapolated to sediment using empirical partition coefficients. Sediment quality values derived by multiplying other SQVs by arbitrary numerical values (e.g., Manz et al. [24]) were not included in the present review.

Unless otherwise stated, all concentrations of mercury in sediment are expressed as milligrams of total mercury (inorganic mercury plus organomercury) per kilogram of sediment on a dry weight basis. Sediment quality values (and other effect concentrations for mercury in sediment) were not segregated according to salinity (with the exception of SEDTOX data compilation; see *Predictive ability of mercury SQVs within the SEDTOX database*). No clear differences in mercury SQVs or toxicity data were observed between saltwater and freshwater exposures, although further research would be needed to definitively evaluate salinity-related differences in mercury toxicity.

Limitations of co-occurrence SQVs

The majority of the available SQVs for mercury are co-occurrence SQVs. It is widely recognized in the scientific community that co-occurrence SQVs in general do not represent cause–effect concentration–response relationships for individual chemicals [3,5,7,15,25–28]. This limitation stems from the derivation of co-occurrence SQVs from large databases of paired data on bulk sediment chemical concentrations and biological effects (i.e., toxicity test results or benthic invertebrate community observations). The underlying databases predominantly represent field-collected sediments from urban ports and harbors contaminated with multiple chemicals. Co-occurrence SQVs are derived using various methods, but all of the methods rely on associations between concentrations of single chemicals considered individually and biological effects from all chemical and nonchemical stressors acting together. Co-occurrence SQVs for single chemicals thus do not represent a concentration that would be expected to cause adverse effects in sediment where only that chemical is present [15,28]. Co-occurrence SQVs also do not necessarily represent all sites with multiple stressors because the types and levels of stressors differ from site to site.

The co-occurrence approach provides a weak basis for identifying single-chemical toxic thresholds not only because it assigns effects to selected chemicals in unknown mixtures but also because it relies on correlations rather than controlled experiments designed to determine causality [26]. Co-occurrence methods identify values that are within the range of the data evaluated, regardless of whether a particular chemical actually contributed significantly to toxicity in the evaluated sediments [27,29]. This limitation is greatly exacerbated by the fact that chemical concentrations tend to covary in sediments [10,26], leading to specious correlations between effects and higher concentrations of any given chemical. Conversely, an absence of effects is observed only in cases in which all chemical concentrations are below toxic thresholds. In fact, if an SQV were equal to the actual chemical-specific toxicity threshold, it would not successfully predict a lack of effects due to all other chemicals; to achieve such an objective, the SQV must be similar to background conditions so that the concentrations of other covarying chemicals are also generally low and nontoxic.

Numerous other factors affect SQV derivation by confounding the relationships between chemicals of interest and toxic effects. First, the considerable variation in bioavailability among sediments poses a challenge to any approach that attempts to predict toxic effects based on bulk sediment chemical concentration alone, including co-occurrence approaches [3,5,15,30,31]. Mercury geochemistry has been extensively studied with respect to the potential for net methylmercury formation [32–34], yet a comprehensive understanding remains elusive [34]. Multiple physicochemical and biological factors interact to affect the potential for mercury methylation by sulfate-reducing bacteria, such as organic carbon and sulfate concentrations, sulfide concentrations and forms, redox conditions, and bacterial consortia [33,34]. Factors affecting inorganic mercury speciation and bioavailability to benthic invertebrates are less well characterized than those affecting methylation, although most or all forms of mercury may influence mercury-related toxicity at the base of the food web. In any case, it is clear that mercury geochemistry is quite complicated, and a wide range of effect thresholds can be expected when exposures are expressed simply on the basis of total mercury in bulk sediment.

Second, chemicals that were unmeasured in the databases used to develop co-occurrence SQVs (such as current-use pesticides, ammonia, and sulfide) have been shown to contribute significantly to observed toxicity in other data sets and likely affected some of the co-occurrence database sites as well [3,35].

Third, benthic invertebrate community observations also were likely affected by physical habitat variables in many cases [19,36,37]. Thus, some samples may have been identified as "toxic" when impacts were not actually attributable to the toxicity of the chemicals analyzed but rather physical habitat.

Taken together, these factors dictate that co-occurrence SQVs, including those for mercury, should not be used as chemical-specific toxicity thresholds [5,22,27]. Multiple authors have demonstrated that it is possible to generate mock sediment benchmarks similar to existing co-occurrence SQVs using a real data set of ambient sediment chemistry and either randomly simulating biological responses or conducting constrained random sampling of chemical concentrations independent of any hypothetical biological responses [36,38]. The fact that SQVs derived using different co-occurrence approaches show a degree of consistency has been considered to be an indication that the SQVs reflect an underlying concentration–response relationship [12]. However, as the data compiled in the present review demonstrate, the consistency among co-occurrence SQVs for mercury is a reflection of typical ambient environmental concentrations in the urban sediments represented in many SQV databases, rather than the inherent toxic potential of mercury in sediment.

Given the general limitations with co-occurrence SQVs, it is not surprising that co-occurrence SQVs have limited utility in predicting sediment toxicity in general and specifically for mercury. For example, O'Connor and Paul [39] compiled a data set of 2475 samples with paired sediment chemistry and amphipod toxicity data. Among the 453 samples with at least 1 effects range median (ERM) exceedance, fewer than half were toxic (41%). O'Connor et al. [40] concluded that ERM exceedances, by themselves, "should never be taken to mean that sediment is exerting a toxic effect on the environment or that there would be any benefit to decreasing its chemical content." O'Connor [22] estimated the probability of toxicity associated with effects range lows (ERLs) as approximately 10%, with no abrupt increase in toxicity on exceedance of an ERL. Similarly, Bay et al. [3] compiled paired sediment chemistry and amphipod toxicity data from 151 studies of California marine embayments. Toxicity predictions based on a variety of co-occurrence SQVs yielded low agreement with toxicity test results, only slightly better than would be expected if toxicity were predicted purely by chance [3].

A lack of predictive ability has been observed for SQVs for many chemicals, and the performance of SQVs for mercury is especially poor. MacDonald et al. [9] derived sediment guidelines (threshold effects level and probable effects level) for multiple chemicals and noted that "the reliability of the guidelines for mercury, nickel, total PCBs [polychlorinated biphenyls], chlordane, lindane, and total DDT [dichlorodiphenyltrichloroethane] was low." Similarly, Long et al. [21] indicated that the ERL and ERM benchmarks had differential predictive success of toxicity, depending on the chemical. They noted that "relatively poor relationships were observed between the incidence of effects and the concentrations of mercury, nickel, total PCBs, total DDT, and p,p′-DDE [dichlorodiphenyldichloroethylene]."

Comparison of SQVs to background concentrations

Most SQVs were developed without consideration of the natural geological presence of metals at background levels. To evaluate mercury SQVs in the context of typical background concentrations, the present study compared the SQVs with present-day background concentrations of mercury in soil and sediment compiled from a variety of sources (Supplemental Data, Table S2). As shown in Figure 1, co-occurrence low SQVs for mercury are consistent with central tendency background concentrations, whereas co-occurrence high SQVs are consistent with upper-bound background concentrations. The fact that co-occurrence SQVs are within the range of background concentrations for mercury (and other metals) strongly suggests that the SQVs are not cause–effect toxicity thresholds [5,27]. It is empirically evident from decades of toxicity testing that natural,

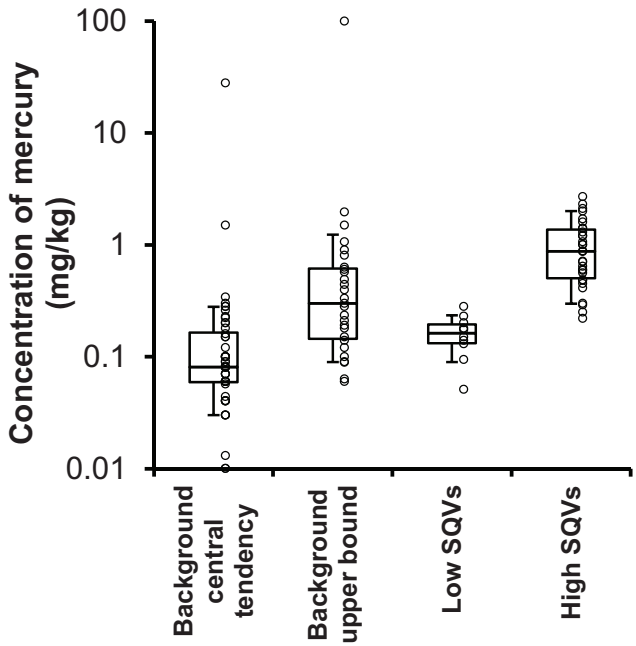

Figure 1. Mercury background levels and co-occurrence sediment quality values (SQVs). Boxes, horizontal bars, and error bars (whiskers) indicate interquartile ranges, medians, and 10th and 90th percentiles, respectively.

unpolluted sediments bearing detectable concentrations of mercury (and other metals) are not likely to be toxic to benthic invertebrates. Mercury is a naturally occurring metal, and invertebrates have evolved various mechanisms to adapt to and thrive in the presence of background mercury levels [41–44]. Although surface sediment mercury concentrations have increased globally because of atmospheric deposition [45], the increase appears limited (approximately 3-fold) relative to the geographic variability of background mercury concentrations, which ranges over more than 1 order of magnitude. From a practical perspective, SQVs that are equivalent to background conditions (especially central tendency background) do not efficiently accomplish the objective of screening because very few sites can be "screened out," regardless of the actual likelihood of toxicity.

Predictive ability of mercury SQVs within the SEDTOX database

To illustrate the lack of relationship between mercury concentrations and toxicity in a large co-occurrence database, paired toxicity and sediment mercury data were retrieved from the SEDTOX database, a large database of concurrently collected sediment chemistry and toxicity data from sites in North America [46]. The SEDTOX database incorporates multiple databases, several of which were developed for deriving co-occurrence SQVs. Most of the information presented herein is transcribed directly from the SEDTOX database and could not be validated for the present study. Underlying documents and information for many of the studies indexed in SEDTOX are not widely available and could not be reviewed to fully confirm the data or site conditions. These constraints are important because in 1 case a units error in the SEDTOX database was found for 1 of the studies (1995–1996 Dade County, Florida study), causing mercury concentrations to be overestimated by 3 orders of magnitude. Nevertheless, our use of the SEDTOX data is consistent with previous uses of this database by other authors (e.g., Fairey et al. [13]).

In the SEDTOX data set, mercury concentrations consistent with central tendency background concentrations or lower (less than or equal to approximately 0.1 mg/kg) tend to indicate clean, nontoxic sediments (Figures 2 and 3). When mercury concentrations are higher than this level, however, no relationship is discernible between mercury concentrations and mortality. A lack of toxicity (survival $\geq$80%) is common at concentrations

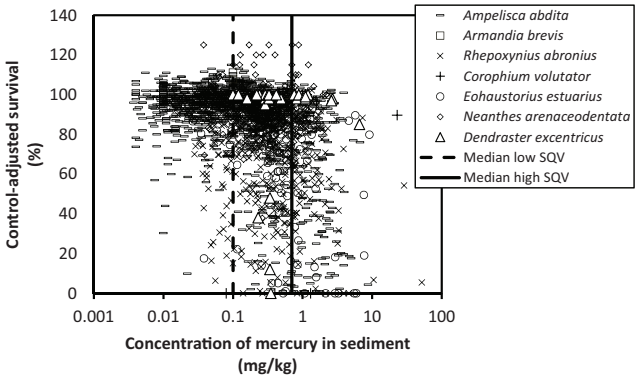

Figure 2. Lethal toxicity test results (survival) and concentrations of mercury in saltwater sediment from the SEDTOX database. Median low (dashed line; 0.1 mg/kg) and high (solid line; 0.7 mg/kg) estuarine and saltwater sediment quality values (SQVs) illustrate the low predictive value of SQVs when mercury concentrations are above background levels (on the order of 0.1 mg/kg).

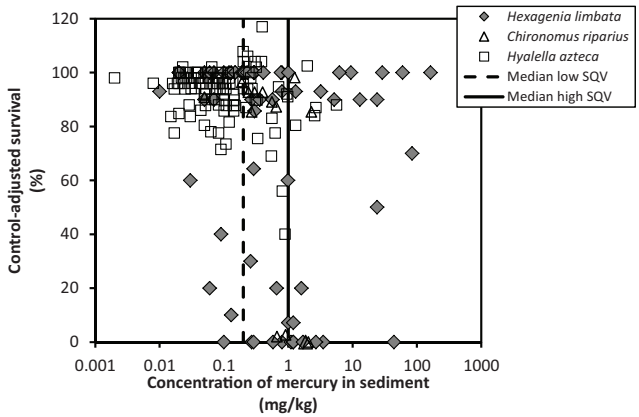

Figure 3. Lethal toxicity test results (survival) and concentrations of mercury in freshwater sediment from the SEDTOX database. Median low (dashed line; 0.2 mg/kg) and high (solid line; 1 mg/kg) estuarine and saltwater sediment quality values (SQVs) illustrate the low predictive value of SQVs when mercury concentrations are above background levels (on the order of 0.1 mg/kg).

1 to 2 orders of magnitude higher than SQVs and background levels. For example, toxicity test organism mortality was less than 20% for nearly half (49%) of the saltwater sediments with concentrations greater than 0.7 mg/kg (the median of the saltwater high SQVs reviewed in this compilation; Figure 2) and more than half (57%) of the freshwater sediments with concentrations greater than 1 mg/kg (the median of the freshwater high SQVs reviewed in this compilation; Figure 3). Even the highest SQVs in the compilation were inaccurate as mercury-specific toxicity thresholds. Nearly 40% of the 65 freshwater (Figure 3) and saltwater (Figure 2) sediment samples with concentrations of mercury greater than 2.7 mg/kg (the highest co-occurrence SQV) exhibited toxicity test organism mortality less than 20%. Although this discussion focuses on mortality as opposed to sublethal endpoints, mortality data are the primary basis of most SQVs. Thus, although co-occurrence SQVs have been described as useful for predicting the occurrence of toxicity, if not its cause [7], the utility of SQVs for mercury as non-causal indicators of potential toxicity is limited. This conclusion is consistent with previous findings that mercury SQVs are less reliable predictors of toxicity than SQVs for some other chemicals [9,21].

Equilibrium partitioning SQVs

In contrast to the large number of co-occurrence SQVs, only a small number of EqP SQVs were identified for mercury [17–19], with concentrations ranging from 0.63 mg/kg to 10 mg/kg (Supplemental Data, Table S3). Equilibrium partitioning SQVs are derived using information on chemical partitioning between sediment particles (organic carbon, solid mineral matrices, etc.) and porewater [15] to translate a threshold concentration of the chemical of interest in water (e.g., a concentration assumed to be safe because of a lack of effects) into a concentration in sediment. In contrast to co-occurrence SQVs, EqP effects information is derived from single-chemical exposures, albeit usually in water. Equilibrium partitioning benchmarks have been developed for nonpolar organic chemicals and divalent metals [47] and are considered useful for understanding chemical-specific thresholds for toxicity [3]. These EqP models attempt to account for key factors affecting chemical bioavailability by estimating sediment porewater partitioning as a function of organic carbon and/or sulfide concentrations. In

contrast, the EqP SQVs for mercury are empirical and do not account for differences in bioavailability among sediments.

As has been generally observed for other metals and organic compounds in sediment, the EqP approach for mercury assumes bioavailability is directly related to the freely dissolved concentration of mercury in sediment porewater, because this measurement best represents the portions of sediment-associated chemicals that can readily diffuse into dermal, respiratory, or digestive tissues in contact with sediment [15,48]. The processes affecting concentrations of bioavailable mercury in sediment porewater are controlled by sorption of mercury to sediment solid and mineral phases, which in turn is greatly affected by sediment chemical and physical conditions such as pH, redox potential, and presence of sulfate, sulfides, iron, and organic matter [31,49,50]. Exposure and fate of mercury in sediment and sediment porewater also are greatly influenced by the form of mercury [51]. In most aquatic systems, mercury exists in several forms, including elemental mercury (Hg0), inorganic mercury compounds (usually divalent mercury, Hg[II]), and organomercury compounds, such as methylmercury or dimethylmercury [34,52–55]. In sediments, Hg0 is typically a small proportion of total mercury and is not directly available for organism uptake [56,57]. Inorganic Hg(II), present as a cation (Hg^{2+}), usually predominates in most mercury-contaminated sediment. Only a small portion of Hg^{2+} is present in a truly dissolved, bioavailable form; the majority is bound in mercury–ligand complexes with chloride, dissolved organic matter, and reduced sulfur (e.g., organic thiols and sulfhydryl groups [58–60]) or associated within or adsorbed to solid mineral particles [34].

These complexities imply that EqP SQVs for mercury do not provide the level of certainty associated with mechanistic models for divalent metals [61] or nonpolar organic chemicals [62]. Existing models are capable of predicting mercury fate but usually are developed empirically (rather than mechanistically) on a site-by-site basis and require considerable amounts of site-specific data [49]. Nonetheless, 3 European jurisdictions have adopted EqP SQVs for mercury based on empirical measurements of bulk sediment porewater partitioning for mercury [17–19]. Differences among the EqP SQVs reflect variability in both the selected empirical sediment porewater partitioning coefficients and the aquatic toxicity information used in each value's derivation. Because these SQVs are based on empirical rather than mechanistic predictions of chemical partitioning, they do not account for differences in sediment porewater partitioning and bioavailability among sites and must be used with caution.

CAUSAL MERCURY EFFECTS DATA FOR BENTHIC INVERTEBRATES

To evaluate an alternative to co-occurrence SQVs, we compiled matched mercury chemistry and effects data from lethal and sublethal toxicity studies using mercury-spiked sediment and from biological data for field-collected sediment from sites where the only or the primary stressor is presumed to be mercury (i.e., "mercury sites"). For the purposes of the present article, mercury sites are defined as sites primarily contaminated by mercury as a result of clearly identified mercury sources, such as chloralkali processing, mercury or gold mining, or other documented point source releases of mercury.

Data compilation focused on biological endpoints with clear ecological relevance, such as survival, reproduction, and growth (as measured by laboratory toxicity testing), and benthic community attributes, such as abundance, diversity, and richness (as measured by benthic census of field sediment)—that is, the same endpoints most commonly used in co-occurrence approaches. Documentation of effects was verified by examining the data, statistical evaluations, and methods, as provided by the authors of the original study. Effects were considered ecologically significant if the results were significantly different from the control or reference measurement (typically $\alpha = 0.05$) and at least 20% different from the same control or reference measurement [22,63]. In a few cases where applicable statistical analysis was not presented, only the 20% criterion was used to identify adverse effects.

Toxicity test results were identified from whole-sediment exposure durations equal to or greater than 7 d. The time course of toxicity in benthic invertebrates depends on the dose present at the sites of toxic action, which is largely a function of uptake. Mercury may be accumulated more slowly than other metals and thus may require longer periods for adverse effects to become evident. For example, Lawrence and Mason [51] estimated that steady state is achieved in the amphipod *Leptocheirus plumulosus* after 50 d, whereas Stephenson and Turner [64] reported that steady state was attained in 14 d by *Hyalella azteca*. Amirbahman et al. [65] found that uptake by *L. plumulosus* and the bivalve *Macoma nasuta* plateaued after 14 d, whereas a plateau in uptake in the polychaete *Nereis virens* was observed after 7 d. In contrast, Kennedy et al. [66] found that concentrations appeared to reach steady state for *N. virens* within 7 d, whereas steady state for *M. nasuta* was not observed until approximately 70 d to 90 d. For the purposes of the present review, 7 d was considered to be a reasonable minimum exposure duration requirement for quantification of toxicity via whole-sediment exposures, recognizing that longer exposures could potentially yield more sensitive responses for some species.

Data were evaluated with the goal of deriving study-specific no-observed-effect concentration (NOEC) and/or lowest-observed-effect concentration (LOEC) values reflecting concentrations of total mercury in sediment. We interpreted each study according to the following principles. First, studies in which no adverse effects were detected, the highest concentration of mercury was selected as an unbounded NOEC (i.e., the true NOEC value is greater than or equal to the unbounded NOEC). Similarly, if adverse effects were found at all tested concentrations, the lowest concentration of mercury was selected as an unbounded LOEC. Second, for studies in which adverse effects were found and were consistent with an exposure–response relationship (i.e., effects associated with higher mercury concentrations and lack of effects associated with lower mercury concentrations), the lowest concentration in the sediments exhibiting adverse effects was selected as the LOEC and the highest concentration in sediments without detectable adverse effects was selected as the NOEC. Third, in studies in which adverse effects were found but were not consistent with an exposure–response relationship, the highest concentration of mercury observed in the nontoxic sediment was generally selected as an unbounded NOEC. There is some uncertainty in this approach in cases in which bioavailable mercury concentrations were not determined (i.e., an exposure–response relationship might have existed for bioavailable mercury). Therefore, studies that fell in this category were considered usable for identification of threshold mercury concentrations only if other likely causes of observed toxicity were clearly identifiable (e.g., concentration–response relationships with other stressors), effects were also observed at

reference sites (i.e., not caused by mercury), or effects were observed in only a few samples. All such interpretations are described below or in the Supplemental Data. 4) For studies in which multiple endpoints were measured but no effect was documented, effects were noted as combined endpoints (i.e., "survival and growth") in summary tables and represented as single-point values in figures. In studies for which different LOECs could be estimated for at least 1 or more endpoint, metrics for each endpoint are shown separately (e.g., NOEC and LOEC for growth, NOEC and LOEC for survival).

In general, we attempted to be inclusive with regard to the variety of laboratory and field studies summarized in the present review. We included "gray literature," and thus all studies are not equivalent in terms of the extent of available documentation. Studies were included only if the documentation and data quality were sufficient to support confident interpretation. Considering the wide variety of sediment and site conditions, experimental designs, and study objectives, no single study can be identified as the most robust study of mercury effects on benthic invertebrates.

Mercury-spiked sediment toxicity studies

Data from 7 mercury-spiked sediment studies are available for 5 taxa, as shown in Table 1. The results of these mercury-spiked sediment studies suggest mercury toxicity thresholds that are generally higher than SQVs but lower than toxicity thresholds from aged, field-contaminated sediments (for further discussion, see *Comparison of SQVs for mercury and compiled effects information*). No mercury-spiked sediment studies evaluating reproduction as an endpoint could be located (i.e., only survival and growth effects data were available).

Swartz et al. [48] evaluated lethal toxicity of mercuric chloride to the amphipod *Rhepoxynius abronius*. Amphipod survival varied widely among sediments. In unaltered, field-collected sediment spiked with mercury, a clear dose response enabled the estimation of NOEC (10.2 mg/kg), LOEC (12.8 mg/kg), and median lethal concentration (LC50; 13.1 mg/kg) values. Effect concentrations varied widely when the spiked sediment was manipulated to alter bioavailability. For example, in a second experiment conducted with 2 sediments spiked at the LC50, a mercury concentration of 13 mg/kg caused 100% mortality in the original sediment but only 1% mortality when the same sediment was enriched with fines. Porewater mercury concentrations were more consistent with amphipod mortality in that experiment (see *Research Recommendations* for additional discussion).

Peluso et al. [67] measured amphipod (*Hyalella curvispina*) growth and mortality in artificial sediment and a field-collected sediment spiked with inorganic mercury. One of the artificial sediments used in the study was excluded from the present review because it was formulated to contain no organic carbon, which is not representative of natural sediments. The authors calculated growth inhibition based on differences between initial and final lengths. On this basis, growth in the artificial sediment was significantly inhibited by approximately 20% in amphipods exposed to 5.1 mg/kg mercury (LOEC), whereas no significant effect was observed following exposure to 3.3 mg/kg (NOEC). For the mercury-spiked field-collected sediment, the lowest

Table 1. Concentrations of mercury in sediment and associated effects in mercury-spiked sediment

Species	Water salinity	Spiked compound	Equilibrium time (d)	Exposure duration (d)	Measured effect	Endpoint	Mercury concentration[a] (mg/kg, dry wt)	Organic carbon content	Study
Amphipod (*Rhepoxynius abronius*)	Salt	Mercury (II) chloride	1.7	10	Survival	NOEC	10.2[b]	0.22%	[48]
						LOEC	12.8[b]		
						LC50	13.1[b]		
Amphipod (*Hyalella curvispina*)	Fresh	Mercury (II) chloride	7	21	Growth	NOEC	3.3[c]	3.5%	[67]
						LOEC	5.1[c]		
					Survival	Unbounded NOEC	11[c]		
					Growth	NOEC	7.6[d]	12%	
						LOEC	9.4[d]		
					Survival	Unbounded NOEC	9.4[d]		
Midge (*Chironomus riparius*)	Fresh	Mercury (II) chloride	7	14	Growth	NOEC	0.93	2.5%	[70]
						LOEC	2.42		
					Development rate	NOEC	0.59		
						LOEC	0.93		
					Survival	NOEC	2.42		
						LOEC	3.84		
Midge (*Chironomus riparius*)	Fresh	Mercury (II) chloride	"Minimal"	21–35	Growth	Unbounded NOEC	2.86	0.7%	[71]
Nematodes	Salt	Mercury (II) chloride	7	60	Benthic community effect indices	Unbounded LOEC	0.084	1.32%	[72]
Mayfly (*Hexagenia rigida*)	Fresh	Mercury (II) chloride	5	15	Survival and growth	Unbounded NOEC	10.5	2%	[69]
		Methylmercury chloride	5	15	Survival and growth	Unbounded NOEC	1.1		
Mayfly (*Hexagenia rigida*)	Fresh	Methylmercury chloride	3	15	Survival and growth	Unbounded NOEC	6.3	1.5%	[68]

[a]Mercury concentrations are measured values, except as noted.
[b]Mercury concentrations are nominal. The LC50 value based on measured concentrations in sediment was reported as 15.2 mg/kg; however, the NOEC and LOEC based on measured values were not presented.
[c]Mercury concentrations are the average of initial and final measured concentrations in artificial sediment.
[d]Mercury concentrations are the average of initial and final measured concentrations in field-collected sediment.
LC50 = median lethal concentration; LOEC = lowest-observed-effect concentration; NOEC = no-observed-effect concentration.

mercury exposure resulting in statistically significant growth inhibition >20% was 9.4 mg/kg (LOEC, 33% inhibition), with <20% inhibition at the 7.6 mg/kg (NOEC) exposure level. If growth were evaluated based on the percent difference in final lengths between spiked and control samples (rather than change in length during the exposure period), then none of the spiked samples would be considered >20% different from the control. Neither sediment elicited significant amphipod mortality (unbounded NOECs of 11 mg/kg and 9.4 mg/kg for artificial and field sediments, respectively).

Odin et al. [68] evaluated spiked sediment toxicity of methylmercury to the mayfly *Hexagenia rigida*, with no effects on growth or survival observed at concentrations up to 6.3 mg/kg (unbounded NOEC). In an earlier study using both inorganic mercury and methylmercury [69], exposure to 10.5 mg/kg inorganic mercury and 1.1 mg/kg methylmercury was also nontoxic (unbounded NOEC for growth and survival). These studies focused on mercury bioaccumulation and did not attempt to establish a toxicity threshold for total mercury or methylmercury in sediment.

Two studies examined mercuric chloride effects on the midge *Chironomus riparius* in mercury-spiked artificial sediments. Chibunda [70] reported a 54% reduction in growth of midges exposed to 2.42 mg/kg (LOEC), with an NOEC of 0.93 mg/kg. Gremyatchikh et al. [71] found no significant growth reduction at 2.86 mg/kg (unbounded NOEC). Chibunda [70] found that mortality was a less sensitive endpoint than growth (NOEC of 2.42 mg/kg, 84% mortality at LOEC of 3.84 mg/kg), whereas development rate was more sensitive (NOEC and LOEC of 0.59 mg/kg and 0.93 mg/kg, respectively). Although Gremyatchikh et al. [71] noted effects on survival in the range of 0.5 mg/kg to 1.5 mg/kg, quantitative data were not reported, so we could not derive effect concentrations for mortality from that study.

In a mesocosm study, Hermi et al. [72] observed changes in Mediterranean nematode community structure at all spiked concentrations of mercuric chloride, as low as 0.084 mg/kg (unbounded LOEC). Nematode abundance and species richness were reduced by approximately 30% to 40%, and certain nematode species responded consistently to mercury exposure, suggesting particular sensitivity or tolerance. Nematodes are considered meiofauna based on their small size and often are not included in benthic macroinvertebrate community investigations (because they can pass through the sieves typically used to separate invertebrates from sediment). However, effects on nematode community function (e.g., diminished availability as prey for larger invertebrates), if ecologically adverse, would be observable in macroinvertebrate community condition. The LOEC from Hermi et al. [72] is a low outlier compared with the other spiked sediment and mercury site studies (including macroinvertebrate community studies) identified in the present review.

Invertebrate effects studies at mercury sites

Sediment toxicity and/or benthic invertebrate community study results from 23 investigations of 14 mercury sites are summarized in Table 2. The basis for the selected NOEC and LOEC values is outlined below, with further details provided as Supplemental Data. Mercury exposures are shown in Table 2 based on total mercury in sediment; however, additional measures of mercury exposure were collected in many cases, such as methylmercury analyses, porewater analyses of total mercury or methylmercury, and tissue analyses. Where available, results of these analyses as they relate to the applicable biological results are discussed in the Supplemental Data.

At the majority of sites, a lack of adverse effects on benthic invertebrates caused by mercury was evident. The highest whole-sediment mercury concentrations exerting no adverse effect were reported from Clear Lake (CA, USA), a former mercury mine site. There, Suchanek et al. [73,74] found no population- or community-level effects on invertebrates associated with mercury concentrations up to 1200 mg/kg. This finding was supported by observations of low mercury concentrations in chironomid tissues across a range of sediment exposures, indicating low mercury bioavailability [73]. Environment Canada identified no adverse effects from mercury in sediments of Peninsula Harbour or the St. Clair River (both in Ontario, Canada), based on a battery of chronic and subchronic sediment toxicity tests with amphipods, midges, mayflies, and oligochaetes, as well as benthic community census results [75,76]. Sediment mercury concentrations at these sites ranged up to 19.5 mg/kg and 49 mg/kg, respectively. Additional sediment quality triad studies indicated a lack of adverse effects attributable to mercury in sediments from Lavaca Bay (Texas, USA), the Androscoggin River (New Hampshire, USA), and Lake Maggiore/Toce River (Italy), despite maximum mercury concentrations ranging from 0.3 mg/kg to 5.2 mg/kg in sediment [77–79]. In a particularly lengthy experiment, exposure to New York Harbor sediment (NY, USA) for 100 d caused no effects on survival of polychaete worms, clams, or grass shrimp at sediment mercury concentrations up to 35 mg/kg [80]. In the Whatcom Waterway (WA, USA), sediment toxicity tests with amphipods, polychaetes, bivalves, and sand dollars conducted from 2002 to 2008 yielded no adverse effects in 37 of 38 samples, with mercury concentrations up to 2.6 mg/kg [81,82]. Toxicity tests at this site in earlier years (1996–1998) showed a greater incidence of effects, but the observed toxicity was correlated with concentrations of phenolic compounds related to wood waste (which subsequently declined), rather than mercury [81]. Sediments from the Sudbury River (MA, USA) and connected reservoirs and wetlands had no effect on mayfly survival, and most samples (12 of 14) did not affect mayfly growth, despite sediment mercury concentrations up to 22.1 mg/kg [54]. No relationship was evident between mayfly growth and various measures of mercury exposure, including total and methylmercury in sediment and mayfly tissue, as well as methylmercury in overlying water [54].

At 5 mercury sites, some degree of adverse effect was observed that may be attributable to mercury, although possible contributions of other stressors cannot be excluded. A sediment quality triad study of the South River (VA, USA) indicated no adverse effects at any location, with sediment mercury concentrations up to 18.9 mg/kg [83]. On the other hand, Bundschuh et al. [84] observed an inhibition of amphipod feeding rate in sediment containing 10.3 mg/kg mercury from the South River, although the ecological implications of this test endpoint are uncertain. Similarly, exposure to sediments from the Brunswick estuary (GA, USA) resulted in decreased leaf consumption by amphipods; sediment mercury concentrations were 18 mg/kg to 25 mg/kg [85]. Brunswick estuary sediments did not cause significant amphipod mortality at concentrations up to 551 mg/kg [85–87], although exposure to sediment containing 972 mg/kg mercury (and 27 000 μg/g [organic carbon weight basis] polychlorinated biphenyls) caused 73% mortality [87]. Other evaluations of sublethal endpoints have been conducted for Brunswick estuary sediments but are not suitable

Table 2. Concentrations of mercury in sediment and associated effects at mercury sites

Study location (mercury source)	Water salinity	Cocontam.	Species	Exposure duration (d)	Measured effect	Endpoint	Mercury conc. (mg/kg, dry wt)	OC content	Study
Androscoggin River, NH, USA (chloralkali plant)	Fresh	Potentially other metals and organic compounds	Amphipod (*Hyalella azteca*), midge (*Chironomus dilutus*), benthic community census	28 (amphipod) and 20 (midge)	Survival and growth	Unbounded NOEC	0.3	2.5%	[78]
Augusta Bay, Italy (chloralkali plant)	Saltwater	PAHs	Amphipods (*Rhepoxynius abronius*)	10	Survival	NOEC	22	14.1%	[94]
						LOEC	37	8.3%	
			Amphipod (*Leptocheirus plumulosus*)	10	Survival	NOEC	38	9.2%	
						LOEC	373	13.3%	
Brunswick estuary, GA, USA (chloralkali plant)	Estuarine	Metals, PAHs, and PCBs	Amphipod (*L. plumulosus*)	28	Survival	NOEC	551	7.3%	[87]
						LOEC	972	8.4%	
			Amphipod (*H. azteca*)	10	Survival	Unbounded NOEC	24.7	5.90%	[85]
					Feeding rate	Unbounded LOEC	18		
			Amphipod (*L. plumulosus*)	14	Behavioral abnormalities and survival	Unbounded NOEC	170	0.78%	[86]
Clear Lake, CA, USA (mercury mine)	Fresh	DDD and methyl parathion	Benthic invertebrate community	NA	Benthic invertebrate population and community-level effects	Unbounded NOEC	1200	Not reported	[73,74]
Lake Maggiore and Toce River, Pieve Vergonte, Italy (chemical production factory)	Fresh	DDT	Midge (*C. dilutus*), benthic community census	28	Emergence, survival, and growth	Unbounded NOEC	5.2	1.76%	[79]
Lavaca Bay, Point Comfort, TX, USA (chloralkali plant)	Estuarine	PAH	Polychaete (*Neanthes* sp.), amphipod (*Leptocheirus* sp.), benthic community structure	20 (polychaete), 28 (amphipod)	Survival (amphipod, polychaete), growth (polychaete), benthic community metrics	Unbounded NOEC	4.6	0.51%	[77]
Mabubi River, Tanzania (artisanal gold mine)	Fresh	Arsenic	Midge (*Chironomus riparius*)	10	Growth	NOEC	1.6	0.91%	[95]
						LOEC	2.3	0.56%	
					Development rate	NOEC	0.23	1.2%	
						LOEC	1.6	0.91%	
					Survival, emergence	Unbounded NOEC	2.3	0.56%	
New York Harbor, NY, USA (assumed industrial point source)	Estuarine	PCBs and cadmium	Annelid worm (*Nereis virens*), bivalve (*Mercenaria mercenaria*), shrimp (*Palaemonetes pugio*)	100	Survival	Unbounded NOEC	34.89	22.3%	[80]
Peninsula Harbour, Lake Superior, Ontario, Canada (chloralkali plant)	Fresh	PCBs and wood waste	Benthic community assessment, amphipod (*H. azteca*), midge (*C. riparius*), mayfly (*Hexagenia* sp.), worm (*Tubifex tubifex*)	28 (amphipod and worm), 10 (midge), 21 (mayfly)	Benthic census metrics, survival and growth (amphipod, midge, and mayfly), survival and reproduction (worm)	Unbounded NOEC	19.5	6.8%	[75]
Ria de Aveiro coastal lagoon, Portugal (chloralkali plant)	Estuarine	PCBs	Benthic community census	NA	Abundance and species richness	NOEC	3	Not reported	[91]
						LOEC	7		
			Mysid (*Mesopodopsis slabberi*)	NA	Population growth, biomass, and density	Unbounded NOEC	200	Not reported	[92]
			Mudsnail (*Peringia ulvae*)	NA	Growth, biomass, and density	Unbounded NOEC	82	Not reported	[93]
					Population age structure	NOEC	24		
						LOEC	82		
			Amphipods	NA	Density and productivity	NOEC	2	Not reported	[90]

(*Continued*)

Table 2. (*Continued*)

Study location (mercury source)	Water salinity	Cocontam.	Species	Exposure duration (d)	Measured effect	Endpoint	Mercury conc. (mg/kg, dry wt)	OC content	Study
						LOEC	11		
			Isopods	NA	Density and productivity	Unbounded NOEC	206	Not reported	
			Mysids	NA	Density and productivity	Unbounded NOEC	206	Not reported	
South River, VA, USA (textile plant)	Fresh	None reported	Amphipod (*H. azteca*) midge (*C. dilutus)*, benthic community structure	10	Survival and growth, various benthic macroinvertebrate community metrics	Unbounded NOEC	18.9	Not reported	[83]
			Amphipod (*H. azteca*)	7	Feeding rate	NOEC	5.9	Not reported	[84]
						LOEC	10.3		
St. Clair River, Ontario, Canada (chloralkali plant)	Fresh	Hexachlorobenzene, octachlorostyrene (both at low levels)	Benthic community assessment, amphipod (*H. azteca*), midge (*C. riparius*), mayfly (*Hexagenia* sp.), worm (*T. tubifex*)	28 (amphipod and worm), 10 (midge), 21 (mayfly)	Benthic census metrics, survival and growth (amphipod, midge, and mayfly), survival and reproduction (worm)	Unbounded NOEC	49	1.5%	[76]
Sudbury River, MA, USA (industrial complex/ Nyanza Chemical Waste Dump Site)	Fresh	Multiple	Mayfly (*Hexagenia* sp.)	21	Survival and growth	Unbounded NOEC	22.1	16.6%	[54]
Whatcom Waterway, Bellingham Bay, WA, USA (chloralkali plant)	Estuarine	Phenol, 4-methylphenol, 2,4-dimethylphenol	Amphipod (*Eohaustorius estuarius*), sand dollar (*Dendraster excentricus*) or bivalve (*Mytilus* sp. or *Crassostrea* sp.), polychaete (*Neanthes arenaceodentata*)	10 (amphipod), 2 (bivalve or sand dollar), 20 (polychaete)	Survival (amphipod and sand dollar or bivalve), growth (poly-chaete)	Unbounded NOEC	2.6	4.1%	[81,82]

Cocontam. = cocontaminants; Mercury conc. = mercury concentration; OC = organic carbon; LOEC = lowest-observed-effect concentration; NOEC = no-observed-effect concentration; PAH = polycyclic aromatic hydrocarbon; PCB = polychlorinated biphenyl; DDD = dichlorodiphenyldichloroethane; DDT = dichlorodiphenyltrichloroethane; NA = not available.

for identifying Horne et al. [86] observed shifts in benthic feeding guilds along a gradient of mercury, polychlorinated biphenyls, and organic carbon concentrations. Although results for sediment containing 34 mg/kg mercury showed no effect, the cause of observed effects at other locations is uncertain, and the sample size was relatively small ($n = 4$). Wall et al. [88] found a disruption in the size-versus-fecundity relationship for grass shrimp; however, the authors did not provide mercury data that could be used to identify a threshold concentration for this effect, and absolute fecundity was not affected. Additionally, post-remedy chronic toxicity monitoring of Brunswick estuary sediments using amphipods and grass shrimp [89] yielded no concentration–response relationships, and poor performance in uncontaminated reference sediments suggested possible methodological issues. Overall, there are only "subtle indications" of possible adverse effects on benthic effects in Brunswick estuary sediments [88]; therefore, the toxicity test results for mortality and amphipod feeding rate provide a reasonable representation of mercury effects at the site for the purposes of the present analysis. At the Ria de Aveiro (Portugal), multiple studies have evaluated benthic invertebrate responses across a gradient of mercury exposures. The most sensitive responses were observed in amphipod populations (density, productivity) [90] and benthic communities (abundance, richness) [91], with NOECs of 2 mg/kg to 3 mg/kg and LOECs of 7 mg/kg to 11 mg/kg. Less sensitive taxa included mysids [90,92], isopods [90], and snails [93]. Sediments from Augusta Bay (Italy) caused significant mortality in 2 amphipod species, with LOECs of 37 mg/kg and 373 mg/kg [94]. Although porewater and tissue analyses indicated limited mercury bioavailability and significant polycyclic aromatic hydrocarbon exposures ([94]; J.M. Conder, unpublished data), these LOECs are included in the present analysis as potential effects of mercury in the interest of conservatism. Finally, sediments from the Mabubi River (Tanzania) downstream of an artisanal gold mine impaired the growth and development rate of midges at mercury concentrations as low as 1.6 mg/kg to 2.3 mg/kg [95]. Although other confounding factors cannot be ruled out, mercury bioavailability in the Mabubi River could be relatively high because the mercury source is recent and ongoing. Thus, mercury bioavailability in this system might be more similar to that observed in spiked sediment toxicity tests than in many sites contaminated by historical sources.

Two additional mercury sites provide supporting information that is generally consistent with the studies described above but less conclusive with respect to identification of specific NOEC or LOEC values. In the Calcasieu estuary (LA, USA), sediment toxicity was found to be caused primarily by hexachlorobutadiene, but targeted testing was also conducted with samples containing elevated mercury concentrations and lower levels of organic contaminants. Some adverse effects were observed, but they were not related to total mercury concentrations in sediment (up to 4.1 mg/kg) [96]. Similarly, multiple contaminants complicate the interpretation of benthic invertebrate survey results for Eight Day Swamp, along Berry's Creek (NJ, USA). At that site, benthic community diversity varied widely but showed no relationship with total mercury in sediment up to a concentration of 68 mg/kg [43]. In these 2 studies [43,96], the highest concentrations without mercury-related effects were considerably elevated above SQVs, but the cause of effects at lower concentrations could not be clearly determined (for additional description, see Supplemental Data). Thus, without measurements of bioavailable mercury exposures, the possibility of effects attributable to bioavailable mercury associated with lower total mercury concentrations could not be definitively excluded for these 2 studies.

Comparison of mercury SQVs and compiled effects information

The cumulative frequency distributions for co-occurrence low and high SQVs and concentrations associated with effects/lack of effects in mercury-spiked and mercury site sediment (Tables 1 and 2; Supplemental Data, Table S1) are shown in Figure 4. Where available, LOECs are paired with the respective NOECs. Values for sediments spiked with methylmercury are included, although no SQVs for methylmercury were identified in the present review. Cumulative frequency distributions are used here only as a visualization tool, not as quantitative species sensitivity distributions. Thus, multiple data types are combined (freshwater and saltwater, lethal and sublethal), and in some cases, multiple results are presented for the same species.

As shown in Figure 4, co-occurrence SQVs poorly predict the likelihood of toxicity attributable to mercury. Most co-occurrence SQVs for mercury (0.05–2.7 mg/kg) were 1 to 2 orders of magnitude less than concentrations of mercury in sediment associated with absence of effects at mercury sites and in mercury-spiked sediments. The low SQVs were 1 order of magnitude to several orders of magnitude lower than the single-chemical NOECs for mercury in both mercury-spiked sediment and field studies, with medians (± interquartile range) of 0.16 mg/kg (0.13–0.20 mg/kg), 3.3 mg/kg (1.1–9.4 mg/kg), and 22 mg/kg (3.8–66 mg/kg), respectively. The highest co-occurrence SQV (2.7 mg/kg) was <75% of the NOECs from studies at mercury sites (0.23–1200 mg/kg) and 67% of the NOECs identified in the mercury-spiked sediment toxicity tests (0.08–11 mg/kg).

Only 4 of the 14 bounded LOECs from the mercury-spiked and field studies (0.93 mg/kg, 2.3 mg/kg, 1.6 mg/kg, and 2.4 mg/kg) were lower than the highest co-occurrence SQV. These LOECs were associated with growth and development rate endpoints, not the lethal endpoints that comprise the basis of the co-occurrence SQVs. In contrast, it is striking that there are 6 sublethal LOECs that are higher than the range of high co-occurrence SQVs (in some cases, an order of magnitude higher).

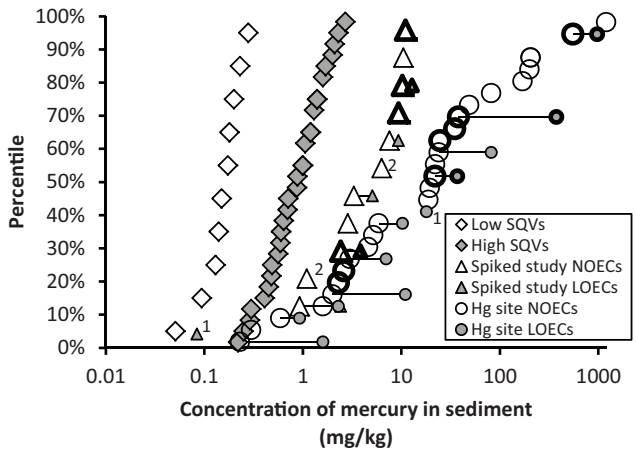

Figure 4. Co-occurrence sediment quality values (SQVs) compared with effect/no effect concentrations of mercury in mercury-spiked and mercury site sediment. Horizontal lines connect bounded no-observed-effect concentrations (NOECs) and lowest-observed-effect concentrations (LOECs). Spiked and mercury site symbols with heavy outlines indicate data in which only lethal endpoints were monitored (other data include sublethal endpoints). 1 = unbounded LOECs; 2 = methylmercury-spiked sediment study results.

In general, it is assumed that sublethal effects occur at exposures lower than lethal effects; and within single studies with the same species and test methods [67,70], sublethal endpoints were more sensitive to mercury (although more research would be justified). Additionally, the lowest laboratory and field LOECs for lethality, 3.8 mg/kg and 37 mg/kg, respectively, were greater than all high SQVs, indicating that high SQVs are well below thresholds for lethal effects. Thus, even the least conservative co-occurrence high SQVs are not meaningful thresholds for predicting lethal or sublethal toxicity caused by mercury.

Equilibrium partitioning SQVs show somewhat better agreement with single-chemical effects data than do co-occurrence SQVs. The 4 EqP SQVs obtained in the present review (range, 0.6–10 mg/kg; Supplemental Data, Table S3) were lower than ranges of effect concentrations shown for mercury-spiked sediment and mercury site sediment. The highest EqP SQV (10 mg/kg) identified for the present study exceeded the least conservative co-occurrence SQV (2.7 mg/kg; Supplemental Data, Table S1) by more than 3-fold. As noted previously, uncertainties related to partitioning/speciation and potential differences in effects measurement for water-only and sediment exposures limit the applicability of existing EqP SQVs to sediments contaminated with mercury.

Sediments spiked with mercury chloride generally indicated effects approximately in the 1 mg/kg to 10 mg/kg range (Figure 4), substantially lower than many of the NOECs and LOECs identified for mercury sites. The median of NOECs from mercury sites (22 mg/kg) was an order of magnitude greater than that from mercury-spiked sediment tests (3.3 mg/kg). The true difference may be even greater because of the likelihood that the field thresholds are conservative. Some of the adverse effects in the field studies may have been the result of cocontaminants, despite our attempt to include data only from sites with a clear mercury contamination source.

The disparity in effect values from field and mercury-spiked studies is likely attributable to higher mercury bioavailability in the spiked sediments than in field sediments. Concentrations of metals in porewater and overlying water of spiked sediment tests can be orders of magnitude higher than levels associated with sediments obtained from sites containing equivalent mercury concentrations in bulk sediment [28]. A primary reason for the enhanced bioavailability of spiked mercury may be the limited equilibration of spiked sediment systems prior to organism addition, because time may be needed for highly available mercury ions in the salt spiking solution to transform to less available mercury forms. The time period between sediment spiking and addition of organisms ranged from "minimal" to 7 d in the experiments reviewed in Table 1, whereas most of the mercury sites reviewed in the present study (Table 2) received mercury inputs several decades before effects were evaluated. Although sorption of inorganic mercury to solid particles can occur on the order of minutes to days [56,97], the overall processes of mercury sorption and speciation may take weeks or months to approach a steady-state exposure concentration for benthic invertebrates [98]. Thus, the short chemical equilibration periods in the mercury-spiked sediment studies to date may have been insufficient to allow dissolved mercury in the spiking solution to sorb to solid sediment phases, resulting in exaggerated mercury bioavailability and toxicity in the spiked sediments relative to field conditions. Additionally, among the mercury-spiked sediment studies identified, none controlled for low pH effects, which often result from spiking sediment with metal salts [28]. Such pH effects could have stressed test organisms (potentially increasing sensitivity) or affected mercury geochemistry. If mercury-spiked sediment information is to be used effectively to predict mercury risks at contaminated sediment sites, the effects of aging on spiked mercury speciation, bioavailability, and toxicity warrant further research.

RESEARCH RECOMMENDATIONS

Risk assessors and risk managers need an alternative to co-occurrence SQVs to better characterize the potential for mercury in sediment to cause adverse effects on benthic invertebrates. Two research recommendations are particularly evident: first, development of a better metric for bioavailable mercury exposure of benthic invertebrates; and second, additional controlled dose–response experiments that involve quantifying effects in sediment primarily or solely contaminated with mercury.

As noted above, chemical bioavailability varies greatly among sediments with differing physicochemical properties. As such, the concentration of mercury in bulk sediment is a very rough substitute measure of the dose at the site(s) of toxic action within an organism. Although the complex geochemical factors affecting mercury bioavailability in sediment are beyond the scope of the present review, it is clear that bioavailability varies widely in a manner that cannot be predicted from analysis of total mercury in bulk sediment. Concentrations associated with observed effects for similar species tested at multiple mercury sites indicate that effect concentrations range over several orders of magnitude (Table 2). For example, the NOEC to LOEC range for survival in *L. plumulosus* was 550 mg/kg to 972 mg/kg at 1 site [87] and 38 mg/kg to 373 mg/kg at another [94]. Unbounded NOECs for *Hyalella azteca* survival range over 2 orders of magnitude [75,76,78,83,85]. For other metals (e.g., copper, nickel), researchers have responded to this high variability with targeted research focusing on the effects of geochemistry and bioavailability on effect concentrations [99–101]. A similar effort for mercury would be helpful to better understand mercury-specific risks to benthic invertebrates.

Stakeholders' understanding of the effects of mercury on benthic organisms would be enhanced by the development of practical techniques to quantify concentrations of dissolved mercury in sediment porewater. Two of the spiked sediment studies (Table 2) expressed toxicity on the basis of concentrations of mercury in porewater, in addition to bulk sediment [48,70]. Porewater mercury appears to be a more precise measure of dose than concentrations in bulk sediment and may better reflect bioavailability. In the Swartz et al. [48] amphipod experiments with sediments differing in fines content, the concentration of mercury in porewater for a lethally toxic sediment (1200 μg/L) was much higher than the concentration in porewater of a nonlethal sediment (260 μg/L), despite the fact that both sediments exhibited similar concentrations of mercury in bulk sediment. Concentrations in porewater associated with lethality in sediment were also approximately consistent with a porewater LC50 of 1750 μg/L observed in an initial experiment using a sediment spiked with a range of mercury levels [48]. In a study with *Chironomus riparius*, Chibunda [70] reported porewater NOEC and LOEC values for mortality of 142 μg/L and 316 μg/L, respectively. These concentrations are roughly comparable to water-only toxicity test results for *C. riparius* exposed to inorganic mercury in water-only exposures (4-d LC50s of 100–547 μg/L [102]). For *C. riparius* growth, the porewater NOEC and LOEC were 85 μg/L and 142 μg/L, respectively [70]. The similarity of effect concentrations in water

and sediment porewater suggests that concentrations in porewater may be a more meaningful dose metric than concentrations in bulk sediment.

However, routine chemical analysis of mercury and methylmercury in porewater cannot currently distinguish truly dissolved concentrations from mercury bound to colloids or dissolved organic carbon. Thus, measurements of mercury in sediment porewater may include bound fractions that are not truly bioavailable [78]. Additionally, artifacts may result from the methods used to obtain sediment porewater samples (e.g., centrifugation of bulk sediment or peepers). Such artifacts may relate to changes in physicochemical conditions that alter mercury species during sampling as well as overestimation of dissolved mercury caused by the presence of colloid-associated mercury [34]. Future analytical refinements to sediment porewater techniques are recommended. Passive sampling methods that involve absorption of labile or dissolved porewater mercury while minimizing the disturbance of sediment physicochemical conditions (e.g., diffusive gradient in thin films using functionalized thiol groups as the binding agent) are likely to aid in understanding mercury in porewater and may be useful as an exposure metric [34,65,103]. These tools are still in development and/or may not be as widely (commercially) available as porewater analyses.

Another commonly used approach for estimating bioavailable mercury is to express concentrations of mercury exposure on an organic carbon–normalized basis under the assumption that mercury bioavailability is controlled primarily by the amount of organic carbon present in sediment [51,53,104]. We found that organic carbon normalization of the effect concentrations in mercury-spiked toxicity tests and mercury site sediment (Tables 1 and 2) did not reduce the variability in effect concentrations (i.e., no reduction in ranges or coefficients of variation of effect concentrations). Organic carbon normalization also did not yield a more precise or discernible relationship between survival and sediment mercury for data obtained from the SEDTOX database, although this is not surprising given the nature of the SEDTOX data (see *Limitations of co-occurrence SQVs*). However, more research is justifiable to explore the role organic carbon may play in affecting mercury bioavailability and toxicity to invertebrates. For example, controlled bioavailability and toxicity studies in which carbon is added to sediment [31,105,106] would aid in evaluating the utility of organic carbon normalization.

Mercury availability is also influenced by redox potential, which can influence methylmercury generation and binding to sulfide [60]. The latter process also factors largely in influencing the bioavailability of other divalent metals to the extent that bioavailability and toxicity can be reasonably predicted using an analytical technique involving determination of acid volatile sulfides (AVS) and dilute acid-extractable metals (simultaneously extracted metals) [61]. The direct application of this approach to understand toxicity for mercury has not been shown to yield the same predictive success as for other divalent metals, although the presence of AVS in sediment is related to mercury bioavailability [53,107]. Measurement of AVS may be considered as a supporting line of evidence for mercury bioavailability studies.

Concentrations of mercury in whole organisms or particular tissues provide an exposure metric alternative to concentrations in abiotic exposure media. Concentrations in tissue provide data that explicitly consider bioavailability while circumventing the need to understand the complicated fate mechanisms occurring outside the organism [108,109]. Effect-based tissue concentrations (e.g., critical body residues, lethal body burdens) have been relatively successful for evaluating the toxicity of lipophilic organic compounds that act via nonspecific narcosis [108] and are advocated as a promising tool for application to metals and metalloids [110]. However, site-specific tissue data are only available at sites with sediment that is not lethally toxic to organisms (i.e., wild organisms are available for collection and/or laboratory organisms survive a bioaccumulation experiment). Additionally, there are few controlled sediment studies that are useful for deriving effect-based tissue concentrations of mercury for aquatic invertebrates. Many studies are uncontrolled field exposures in sediments containing numerous cocontaminants or water-only exposures that may not represent routes of exposure for infaunal benthic invertebrates [111]. Additional research is recommended to explore relationships between mercury in invertebrate tissue and effects.

There is a pressing need to rigorously evaluate the comparative toxicity and bioavailability of inorganic and methylmercury to benthic invertebrates. Few spiked sediment toxicity tests have employed methylmercury, and none have established effect concentrations for the same sediment spiked separately with methylmercury and inorganic mercury. Concentrations of methylmercury in sediment usually are orders of magnitude lower than concentrations of inorganic mercury. However, sediment-associated methylmercury is considered to be more bioaccumulative and bioavailable than inorganic mercury forms [112–114]. In some cases, methylmercury can comprise the majority of the total mercury present in invertebrate tissues [115]. Several studies have shown inorganic mercury to be less toxic than methylmercury in both vertebrate and invertebrate neuronal cell model systems [116]. Aqueous effect concentrations are also lower for methylmercury than for inorganic mercury for many pelagic aquatic invertebrates [104]. It is unclear, however, whether the toxicity of methylmercury to invertebrates exceeds that of inorganic mercury by a large enough margin to contribute significantly to sediment toxicity, given the minuscule proportion of mercury in sediment that is present as methylmercury. Further research to quantify the relative toxicity and bioavailability of methylmercury and inorganic mercury to benthic invertebrates might help resolve the great differences in mercury effect values shown in Figure 4. Such research also would support an improved understanding of how to quantify mercury exposure and effects in sediment.

CONCLUSIONS

The present review of available SQVs for mercury and information from mercury sites and mercury-spiked sediment experiments demonstrates that the available co-occurrence SQVs for mercury poorly predict mercury-specific risks to benthic invertebrates. Many authors caution that co-occurrence SQVs are useful only for initial screening steps or as part of a weight-of-evidence approach that includes toxicity and benthic community census data [3,5,15,30,117]. In cases in which sediments exhibit concentrations that are below co-occurrence SQVs (and thus several orders of magnitude below mercury-specific thresholds in sediment), mercury-driven effects are extremely unlikely. However, exceedance of co-occurrence SQVs indicates nothing about mercury-specific risks to benthic invertebrates, because their use was not originally intended as such. Furthermore, because many co-occurrence SQVs reflect background chemical levels, even their use as a screening tool is not particularly robust. It is clear from the present review that thresholds for "safe" concentrations of mercury in sediment are

likely at least 1 to 2 orders of magnitude greater than co-occurrence SQVs. Maintaining SQVs so far below actual thresholds provides the assurance of extreme conservatism; however, this attribute comes at the potential price of diverting attention and resources from real environmental problems.

Many risk managers or agency guidelines often endorse (or even require) the use of co-occurrence SQVs for mercury and other chemicals beyond initial screening stages [20,118–120]. It is our experience that, under the guise of the precautionary principle, co-occurrence SQVs often receive weight equal to or greater than site-specific biological or toxicological lines of evidence. Given that, beyond their potential use in initial screening stages, co-occurrence SQVs for mercury contribute little toward our understanding or management of benthic mercury risks, there is no scientific support to continue this approach. Thus, co-occurrence SQVs should not be used as remediation goals or as a basis for quantifying natural resource injuries. Possible exceptions to this recommendation include instances in which the costs required to conduct site-specific investigation outweigh the costs associated with remediation [121] or when concentrations of mercury in field sediment are below SQVs or background levels (in those cases, mercury-driven effects are extremely unlikely).

One possible reason for the continued misuse of SQVs is the perceived lack of alternatives—indeed, the available data to characterize cause–effect concentration–response relationships for mercury had not been conveniently compiled and interpreted before the present review. The data provided in the present review demonstrate that information from sites contaminated primarily with mercury and mercury-spiked sediments provides a more appropriate basis than existing co-occurrence SQVs for interpreting risks to benthic invertebrates exposed to mercury, provided that differences in mercury bioavailability between mercury-spiked sediments and in-place sediments are considered. Total mercury concentrations in bulk sediment are available from all such studies and provide a more realistic perspective on the range of observed mercury toxicity thresholds. Such data provide a logical alternative to co-occurrence SQVs for informing ecological risk assessment and management of mercury-contaminated sediments. However, as our understanding of mercury geochemistry, mercury bioavailability, and mercury-specific toxicity increases, we hope that this data set can be improved on (or supplanted entirely) by more precise and accurate metrics with which to predict the likelihood of mercury toxicity.

A better mechanistic understanding of mercury-related risks to benthic invertebrates is needed. Further research is recommended to clarify which measures of mercury exposure best account for differences in mercury bioavailability and toxicity among sediments, as the present review indicates that concentrations of mercury in bulk sediment are a crude measure of mercury bioavailability to benthic invertebrates. A more robust exposure–response data set for bioavailable mercury based on these metrics should be developed to support more precise and accurate measures of mercury dose for use in the assessment and management of mercury risks. In the interim, risk assessors are encouraged to develop site-specific exposure–response relationships or use effects-based mercury values from studies conducted at mercury sites or with mercury-spiked sediment.

Acknowledgment—Funding was provided by the Sediment Management Work Group. M. McCulloch, S. Brown, W. Gala, K. Christensen, T. Parkerton, S. Nadeau, R. Hoke, and 3 anonymous reviewers provided thoughtful comments on earlier drafts of the manuscript. The views expressed herein are solely those of the authors. We thank J. Arblaster for assistance with data review.

SUPPLEMENTAL DATA

Tables S1–S3. (106 KB DOC)
Review of mercury site studies. (45 KB DOC)

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