SETAC PRESS

Environmental Toxicology and Chemistry, Vol. 36, No. 9, pp. 2503–2513, 2017

Printed in the USA

LENTIC, LOTIC, AND SULFATE-DEPENDENT WATERBORNE SELENIUM SCREENING GUIDELINES FOR FRESHWATER SYSTEMS

David K. DeForest,[a,*] Kevin V. Brix,[b,c] James R. Elphick,[d] Carrie J. Rickwood,[e] Adrian M.H. deBruyn,[f] Lucinda M. Tear,[a] Guy Gilron,[g] Sarah A. Hughes,[h] and William J. Adams[i]

[a]Windward Environmental, Seattle, Washington, USA
[b]EcoTox, Miami, Florida, USA
[c]Department of Marine Biology and Ecology, Rosenstiel School of Marine & Atmospheric Science, University of Miami, Miami, Florida, USA
[d]Nautilus Environmental, Burnaby, British Columbia, Canada
[e]CanmetMINING, Natural Resources Canada, Ottawa, Ontario, Canada
[f]Golder Associates, Vancouver, British Columbia, Canada
[g]Borealis Environmental, North Vancouver, British Columbia, Canada
[h]Shell Health–Americas, Shell Oil, Houston, Texas, USA
[i]Red Cap Consulting, Lake Point, Utah, USA

(Submitted 4 November 2016; Returned for Revision 9 December 2016; Accepted 9 March 2017)

Abstract: There is consensus that fish are the most sensitive aquatic organisms to selenium (Se) and that Se concentrations in fish tissue are the most reliable indicators of potential toxicity. Differences in Se speciation, biological productivity, Se concentration, and parameters that affect Se bioavailability (e.g., sulfate) may influence the relationship between Se concentrations in water and fish tissue. It is desirable to identify environmentally protective waterborne Se guidelines that, if not exceeded, reduce the need to directly measure Se concentrations in fish tissue. Three factors that should currently be considered in developing waterborne Se screening guidelines are 1) differences between lotic and lentic sites, 2) the influence of exposure concentration on Se partitioning among compartments, and 3) the influence of sulfate on selenate bioavailability. Colocated data sets of Se concentrations in 1) water and particulates, 2) particulates and invertebrates, and 3) invertebrates and fish tissue were compiled; and a quantile regression approach was used to derive waterborne Se screening guidelines. Use of a regression-based approach for describing relationships in Se concentrations between compartments reduces uncertainty associated with selection of partitioning factors that are generally not constant over ranges of exposure concentrations. Waterborne Se screening guidelines of 6.5 and 3.0 μg/L for lotic and lentic water bodies were derived, and a sulfate-based waterborne Se guideline equation for selenate-dominated lotic waters was also developed. *Environ Toxicol Chem* 2017;36:2503–2513. © 2017 SETAC

Keywords: Selenium Water quality guideline Tissue-based guideline Trophic transfer Sulfate

INTRODUCTION

Selenium (Se) toxicity to fish is primarily manifested via exposure of adult females to dietary organic Se and subsequent maternal transfer to the eggs, which, at sufficiently high concentrations, can result in edema, larval deformities, and mortality in their offspring [1]. The bioaccumulation potential of Se from water into aquatic food webs is highly dependent on site-specific biogeochemistry and food web characteristics; therefore, a wide range of waterborne Se concentrations across different sites can yield a given Se concentration in fish tissue [2–4]. As such, there is general consensus that Se concentrations in fish tissue, especially in eggs or ovaries, are the most appropriate measures of risk to fish from Se [5–9].

Nevertheless, it is still valuable and cost-effective to identify environmentally protective waterborne Se screening guidelines that, if not exceeded, reduce the need to directly measure Se concentrations in fish tissue. The US Environmental Protection Agency (USEPA), for example, recently finalized Se criteria that consist of water column elements and fish tissue elements, with the fish tissue criterion elements overriding the water column criterion elements [9]. In concept, complying with the water column criterion elements would preclude the need for measuring Se concentrations in fish tissue. The USEPA's water column criteria are 3.1 and 1.5 μg/L in lotic (flowing) and lentic (standing) waters, respectively. The British Columbia Ministry of Environment (BCMOE) also provides Se guidelines for both the water column and fish tissue, as well as for sediment and the dietary pathway (invertebrate tissue); but no hierarchy by which one guideline overrides the other is recommended [7]. The BCMOE water column guideline is 2 μg/L (an "alert concentration" of 1 μg/L is also provided). Australia, New Zealand, and South Africa have a chronic water column Se guideline of 5 μg/L; but they do not currently have fish tissue–based guidelines [10,11].

The first objective of the present study was to develop water Se screening guidelines for lotic and lentic waters. These water Se screening guidelines are conceptually similar to the water column criterion elements recently finalized by the USEPA [9]. Both our approach and the USEPA approach used a multistep Se partitioning model to link an Se criterion or guideline for fish eggs to water column concentrations in lotic and lentic water bodies, but different methodologies for compiling and evaluating the model input data were applied. It should be emphasized that the water Se screening guidelines that we derived, as well as the water column criteria and guidelines recommended by the USEPA and the BCMOE, are not site-specific and are intended to be protective of reasonable worst-case site conditions and food-web types (i.e., high Se bioavailability and trophic transfer

This article includes online-only Supplemental Data.
* Address correspondence to DavidD@windwardenv.com
Published online 14 March 2017 in Wiley Online Library (wileyonlinelibrary.com).
DOI: 10.1002/etc.3793

potential). Development of screening guidelines differs from more rigorous methods for translating between fish and water Se concentrations that are based on site-specific information, such as the Bayesian Monte Carlo approach described in Brix et al. [2] and the ecosystem-scale Se model approach described in Presser and Luoma [12].

The second objective was to develop a methodology for deriving sulfate-dependent waterborne Se screening guidelines for selenate-dominated streams. Increasing sulfate concentrations in water reduce selenate bioavailability at the base of the food web [13,14] and hence to higher–trophic level organisms, including fish. A sulfate-dependent waterborne Se screening guideline for selenate-dominated streams would provide a more meaningful trigger for determining the need to monitor Se concentrations in fish tissue in such waters. A sulfate-dependent guideline for Se would be analogous to hardness-dependent water quality guidelines or criteria for divalent metals [15,16].

METHODS

Terminology

In the remainder of the present study, the term "guideline" is used in a generic context that is not intended to reflect any specific regulatory definition. In addition, for simplicity, the term "fish eggs" is used to refer to both fish eggs and fish ovaries, unless noted otherwise. Lastly, the term "particulate" refers to the base of the food chain and may include both primary producers, such as algae and macrophytes, and detritus.

Se partitioning model

The relationship between Se concentrations in fish tissue or bird eggs and waterborne Se has been studied extensively over the last 25+ yr, leading to development of Se bioaccumulation models. Most of these Se bioaccumulation models are partitioning models relating waterborne Se to fish tissue or bird egg Se concentrations via either multiple food chain steps [12,17–19] or a single step [2,20,21]. Multistep models account for partitioning of Se from water to 1 or more food chain components (e.g., algae, benthic macroinvertebrates) and then into fish tissue or bird eggs, while single-step models directly relate waterborne Se concentrations to colocated fish tissue or bird egg Se concentrations (the latter is typically termed a "bioaccumulation factor" [BAF]).

The key step in these models is the partitioning of Se from water into particulates at the base of the food web (e.g., detritus, algae), termed the "enrichment factor" (EF) [3]. The enrichment factor is an explicit term of a multistep model and an implicit component of the BAF in a one-step model. In multistep partitioning models, trophic transfer factors (TTFs) represent successive steps in the food chain, such as Se transfer from particulates into invertebrates and from invertebrates into fish. Following Presser and Luoma [12], the multistep Se partitioning model is expressed as

$$C_{fish} = C_{water} \times EF_{part} \times TTF_{invert} \times TTF_{fish} \quad (1)$$

where C_{fish} is the fish Se concentration, C_{water} is the waterborne Se concentration, EF_{part} is the enrichment factor (the dissociation constant, K_d, is also used), while TTF_{invert} and TTF_{fish} are invertebrate and fish trophic transfer factors, respectively.

If C_{fish} is set equal to a fish tissue guideline and enrichment factors and trophic transfer factors are defined, then the C_{water} predicted to result in C_{fish} can be calculated by rearranging Equation 1

$$C_{water} = \frac{C_{fish}}{EF_{part} \times TTF_{invert} \times TTF_{fish}} \quad (2)$$

A potential limitation of this equation is that the Se enrichment factors and trophic transfer factors selected to represent a given exposure and food-web scenario are treated as constants across all water and fish Se concentrations. However, the use of constants is not supported by empirical relationships or our understanding of Se biodynamics. Stewart et al. [3], for example, note that enrichment factors and trophic transfer factors are dependent on concentration in a nonlinear manner. The basis for this nonlinearity can be explored further by considering the factors that ultimately influence the magnitudes of Se enrichment factors and trophic transfer factors. In particulates (e.g., phytoplankton), steady-state Se accumulation is determined by a combination of the uptake rate constant ($k_{u,\ water}$), elimination rate constant ($k_{e,\ water}$), and growth rate constant (g; Equation 3); and in consumers, the steady-state accumulation of Se via the diet is a function of assimilation efficiency (AE), ingestion rate (IR), elimination rate constant ($k_{e,\ diet}$), and g (Equation 4) [22–24].

$$C_{Se,part} = \frac{k_{u,water} \times C_{Se,water}}{k_e + g} \quad (3)$$

$$C_{Se,consumer} = \frac{AE \times IR \times C_{Se,diet}}{k_e + g} \quad (4)$$

Although the influence of Se concentration on some of these key terms that influence Se biodynamics has not been evaluated, there are data supporting the empirical enrichment factor and trophic transfer factor observations. For example, short-term (i.e., 3 h) Se uptake data for *Chlamydomonas reinhardtii* suggest that $k_{u,water}$ decreases with increasing waterborne Se concentration [25]. In diet-borne Se exposures, Guan and Wang [26] observed a concentration-dependent decrease in assimilation efficiency when the Se concentration in algal diets increased.

Consequently, it is clear that enrichment factors, trophic transfer factors, and the factors that influence them are inversely related to the exposure concentration [3,27], as has been observed for divalent metals [28]. In the present study, a multistep Se partitioning model was used in which quantile regression methods were applied to establish relationships for each step in the model to back-calculate waterborne Se screening guidelines from a fish tissue Se guideline. The multistep model is illustrated conceptually in Supplemental Data, Figure S1.

Compilation of Se partitioning data

To develop robust water Se screening guidelines, Se enrichment factors were compiled from a wide range of locations and exposure conditions and trophic transfer factors for invertebrates and fish over a wide range of diet types and exposure concentrations. This data set was assumed to be representative of Se enrichment factors and trophic transfer factors over a broad range of exposure concentrations and,

hence, served as the lower and upper bounds of Se enrichment and trophic transfer potential.

Particulates. Selenium enrichment factors and BAFs tend to be greater in lentic than in lotic systems [2,4,12,29], although there is substantial overlap. Nevertheless, the field enrichment factor data compiled were categorized as either lotic or lentic because some degree of distinction between these 2 categories was apparent when considering relationships between water and particulate Se concentrations. Lotic water bodies included creeks, rivers, and, conservatively, sloughs. The latter may possess more lentic characteristics. In addition, laboratory data in which particulates were exposed to selenate or selenite were compiled separately, allowing for the derivation of waterborne Se screening guidelines for lotic and lentic waters and selenate-dominated and selenite-dominated waters.

Sources of colocated field data compiled in Presser and Luoma [12] were initially consulted and then augmented with field-based data from additional studies. Selenium concentration data for bulk sediment were not included because they were considered to have limited relevance to Se trophic transfer; however, Se concentration data based on fine sediments (e.g., silt) were included, where available. When Se data for multiple particulate types were available for a given site, the geometric mean Se concentration was used. Likewise, if Se data were available for multiple water samples from a site, the geometric mean Se concentration was used. Temporally colocated samples were generally collected within 1 mo of each other or within the same season of the same year.

Invertebrates. Paired Se concentrations in aquatic invertebrates and particulates (i.e., their diets) were compiled from the published and gray literature (primarily government agency reports). Data were compiled from laboratory trophic transfer studies and field studies with colocated measurements of Se in invertebrates and their putative diets. Invertebrate Se concentration data were compiled from a range of invertebrate taxa, including amphipods, cladocerans, crayfish, and insects (e.g., caddis flies, chironomids, crane flies, damselflies, mayflies, stone flies, water boatmen).

Fish. Paired Se concentrations in fish tissues (i.e., eggs and whole body) and their diets were compiled from the published and gray literature. The egg-based data were of greater interest because the fish tissue–based guideline being considered in this evaluation is an egg-based guideline; however, whole-body Se trophic transfer factors were also compiled because more data are available for whole-body fish tissue. Given the generally higher mobility of fish species, the fish Se data were compiled from laboratory studies so that dietary Se concentrations were accurately measured.

Se toxicity guideline for fish tissues

We applied an Se guideline of 20 μg/g dry weight for fish eggs [30]. This guideline is based on the 5th percentile of a species sensitivity distribution (SSD) of predominantly fish egg Se 10% effect concentrations (EC10s) for reproductive effects. For comparison purposes (see *Discussion* section), the USEPA's fish egg Se criterion of 15.1 μg/g dry weight was also applied [9].

Water Se screening guideline development

Quantile regression was used to back-calculate waterborne Se screening guidelines from the fish egg Se guideline of 20 μg/g dry weight and the USEPA criterion of 15.1 μg/g dry weight. To minimize the influence of laboratory control and field reference site data, quantile regressions were applied only to Se "exposure" data, which were defined as waterborne Se concentrations >1 μg/L and dietary Se concentrations >2 μg/g dry weight, consistent with definitions in Seiler et al. [31] (control and reference site data reduced the slopes of quantile regressions because they tended to fall below inflection points in the relationships between Se concentrations in food chain model steps). Quantile regression analyses were conducted using the quantile regression package ("quantreg") in the software program *R* [32,33]. We used ln-transformed data because log–log relationships were more linear, had more homogeneously distributed relationships, and fit the data better at lower concentrations. Quantile regression was used to estimate median (50th) and upper (75th, 90th) quantiles of the response variables (i.e., Se concentration in particulates, invertebrates, or fish eggs/ovaries) as conditional linear functions of the independent variables. Ultimately, the 75th quantile was used in the multistep models because the 90th quantile compounded conservatism at each step in the food chain such that back-calculated waterborne Se concentrations (i.e., 0.3–0.5 μg/L) overlapped with or were less than background concentration ranges [31,34] and the 50th quantile was not deemed adequately protective for screening guideline development.

Quantile regression relationships were derived to link fish egg Se to invertebrate (dietary) Se and then invertebrate Se to particulate (dietary) Se (quadrants 1 and 2, respectively; Supplemental Data, Figure S1). Particulate Se was then linked to waterborne Se using quantile regression relationships for 4 different data sets (quadrant 3; Supplemental Data, Figure S1), as follows: 1) lotic field, 2) lentic field, 3) laboratory selenite, and 4) laboratory selenite.

Sulfate-dependent screening guideline for selenate-dominated waters

Data were compiled from laboratory studies in which waterborne selenate and sulfate, along with particulate Se concentrations, were measured. Laboratory data were augmented with field data for Se concentrations in periphyton and colocated waterborne Se and sulfate concentrations; we assumed that waterborne Se was predominantly selenate for field data from oxic mountain streams. Particulate Se concentrations were then used to predict Se concentrations in invertebrates and fish eggs using the quantile regression models described in the section *Water Se screening guideline development*. This generated a data set of predicted fish egg Se concentrations that varied as a function of waterborne selenate and sulfate concentrations. Examples of particulate Se concentrations and predicted Se concentrations in invertebrates and fish eggs, varying as a function of waterborne selenate and sulfate concentrations, are provided in Supplemental Data, Figure S2.

A multiple quantile regression analysis was conducted using predicted fish egg Se concentrations as the dependent variable and waterborne selenate and sulfate concentrations as the independent variables. The 75th quantile of the multiple quantile regression model was used to solve for an environmentally protective, sulfate-dependent guideline for selenate-dominated waters. The 75th quantile was used for consistency with the Se screening methodology described above and because it resulted in ≤5% "false negatives" based on validation with a field data set (see *Results* section).

Validation of waterborne Se guidelines using field data sets

The lotic, lentic, and sulfate-dependent waterborne Se screening guidelines were validated relative to a field-based

Se bioaccumulation database [35] augmented with additional unpublished data (S. Covington, Formation Environmental, Austin, TX, USA, personal communication; Supplemental Data, Tables S7–S9). This database included colocated fish tissue (i.e., whole-body, muscle, egg), waterborne Se data, and, where also available, waterborne sulfate data. To augment the egg Se data, egg Se concentrations were estimated from the more prevalent whole-body and muscle Se concentrations using species-specific relationships. These relationships were developed by deBruyn et al. [36] along with relationships developed for additional fish species (Supplemental Data, Section S1).

For each colocated fish egg and waterborne Se concentration, 2 ratios were calculated: 1) the ratio of waterborne Se concentration to the waterborne Se screening guideline, and 2) the ratio of the fish egg/ovary Se concentration to the tissue guideline of 20 μg/g dry weight. The waterborne Se screening guideline was corroborated by the fish egg Se data when both ratios were <1 or >1; false negatives had a water ratio <1 and a fish ratio >1, while false positives had a water ratio >1 and a fish ratio <1.

RESULTS

Se partitioning data

The Se partitioning data compiled are summarized below, in part, based on enrichment factors and trophic transfer factors because these terms are useful for general comparisons among different levels of the food chain, different site types (lotic vs lentic), and Se species (selenate vs selenite).

Particulates. Overall, 75 and 42 pairs of colocated particulate and water Se data were identified for lotic and lentic sites, respectively (Supplemental Data, Tables S1 and S2). Selenium enrichment factors span almost 3 orders of magnitude and more than an order of magnitude within both lotic and lentic sites (Supplemental Data, Figure S3). In general, lentic enrichment factors are approximately 2 times greater than lotic enrichment factors (e.g., the median lentic and lotic enrichment factors are 1387 and 633 L/kg dry wt, respectively). The field-based Se enrichment factors were also plotted against the corresponding water Se concentrations, and significant inverse relationships ($p < 0.001$) were observed (Supplemental Data, Figure S4). Thus, the highest enrichment factors tend to be associated with relatively low water Se concentrations, and the lowest enrichment factors tend to be associated with relatively high water Se concentrations. Almost all Se enrichment factors above the 90th percentile of the data distribution in both lotic and lentic systems are associated with water Se concentrations <1 μg/L. This highlights the uncertainty and need for caution in selecting point estimate enrichment factors in food chain modeling of Se. The regression-based approach used in the present analysis helps to more appropriately address the concentration-dependent Se enrichment at the base of the food chain.

Paired particulate Se concentrations and waterborne selenate and selenite concentrations from laboratory studies, as well as corresponding waterborne selenate and sulfate concentrations used for the sulfate-dependent waterborne Se screening guideline development, are provided in Supplemental Data, Table S3. As for the field-based enrichment factors, significant inverse relationships between laboratory enrichment factors and corresponding water Se concentrations were observed for selenate ($p < 0.001$) and selenite ($p = 0.03$; Supplemental Data, Figure S5).

Invertebrates. Whereas Se enrichment factors ranged over 2 to 3 orders of magnitude, Se trophic transfer factors for invertebrates generally varied between 0.5 and 3.3, although lower and higher trophic transfer factors were observed in a few cases (Supplemental Data, Table S4 and Figure S6). There were clearly differences among taxa and variables within taxa that influenced the magnitudes of Se trophic transfer factors, but the range in magnitude of invertebrate Se trophic transfer factors was much less than that observed for enrichment factors. There were no clear patterns in how trophic transfer factors vary among taxa because insect taxa, for example, were distributed fairly evenly throughout the data set (Supplemental Data, Figure S6). Given the inverse relationship between the invertebrate trophic transfer factor and dietary exposure concentration observed in several studies, all of the laboratory and field data were pooled and trophic transfer factors were plotted against dietary Se (Supplemental Data, Figure S7). A significant ($p < 0.001$) inverse relationship between Se trophic transfer factors and corresponding diet-borne Se concentrations was observed. The highest trophic transfer factors, such as those greater than the 90th percentile, were generally associated with a dietary Se concentration <1.5 μg/g dry weight, and the lowest trophic transfer factors are generally associated with dietary Se concentrations >10 μg/g dry weight. In comparison, Se concentrations in invertebrates collected at control/reference sites ranged from 0.1 to 5.3 μg/g dry weight, with a geometric mean of 1.1 μg/g dry weight. As for Se enrichment factors, this highlights the uncertainty and need for caution in selecting point estimate trophic transfer factors in food chain modeling of Se and supports the regression-based approach used in the present analysis.

Fish. As for invertebrates, Se trophic transfer factors for fish were much lower in magnitude and much less variable than Se enrichment factors, generally varying between 0.7 and 4.1 for eggs and between 0.3 and 1.5 for the whole body (Supplemental Data, Table S5 and Figure S8). As also observed for Se enrichment factors and invertebrate trophic transfer factors, significant ($p < 0.001$) inverse relationships were observed for both fish egg/ovary and whole-body Se trophic transfer factors versus dietary Se (Supplemental Data, Figure S9).

Se screening guidelines

To develop Se screening guidelines, we first related a fish egg Se guideline of 20 μg/g dry weight to a concentration in the fish diet. Using the 75th quantile of the regression relationship between fish egg and dietary Se, a dietary Se concentration of 10 μg/g dry weight was predicted to result in an egg Se concentration of 20 μg/g dry weight (Figure 1A). This is consistent with dietary Se toxicity data for fish because effect levels of >10% are typically observed at dietary Se concentrations >10 μg/g dry weight (Figure 1B).

Next, we linked the Se concentration of 10 μg/g dry weight for fish diets (represented by invertebrates) to a particulate Se concentration. Using the 75th quantile regression relationship between invertebrate and particulate Se, a particulate Se concentration of 4.7 μg/g dry weight resulted in an invertebrate Se concentration of 10 μg/g dry weight (Figure 2).

Finally, we linked a particulate Se concentration of 4.7 μg/g dry weight to waterborne Se concentrations using field data for lotic and lentic systems and laboratory data for selenate and selenite. Using the 75th quantile regression relationships between particulate and waterborne Se, waterborne Se concentrations of 6.5 μg/L in lotic systems and 3.0 μg/L in lentic systems resulted in a particulate Se concentration of 4.7 μg/g

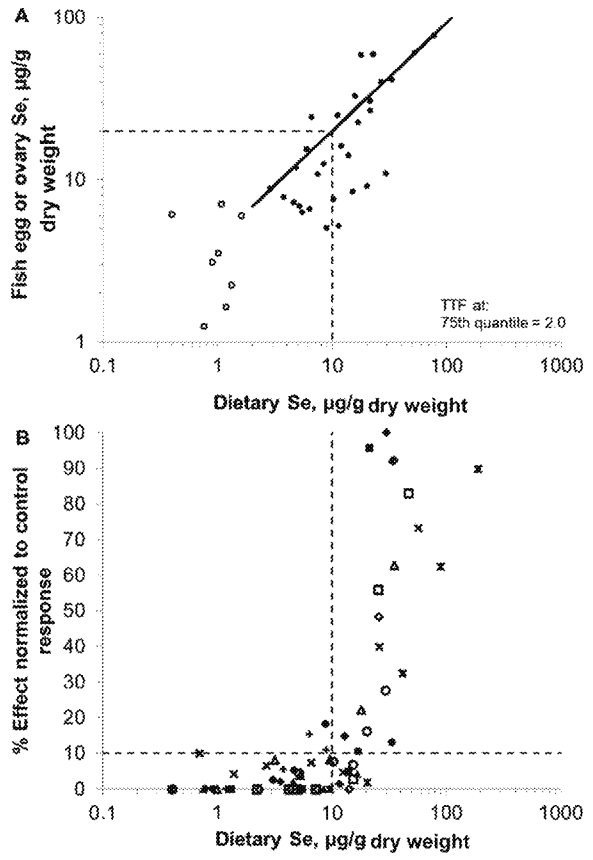

Figure 1. (**A**) Relationship between fish egg or ovary selenium (Se) and dietary Se and (**B**) dietary concentration–response data for fish exposed to organic Se. (**A**) Shown are the 75th quantile (solid line), reference data (○), and exposure data (●). (**B**) Shown are date for bluegill [46] (◆), [47] (■), [48] (▲), [49] (●), and [50] (4–5 °C ◇, 9 °C □); Chinook salmon [51] (Δ); fathead minnow [52] (○); Sacramento splittail [53] (×); white sturgeon [54] (*); and Yellowstone cutthroat trout [55] (+). TTF = trophic transfer factor.

dry weight (Figure 3 and Table 1). Similarly, a selenate concentration of 5.1 µg/L and a selenite concentration of 1.5 µg/L resulted in a particulate Se concentration of 4.7 µg/g dry weight using laboratory data (Figure 4 and Table 1).

For comparison, application of the USEPA's fish egg Se criterion of 15.1 µg/g dry weight to the analysis resulted in back-calculated Se concentrations of 6.5 µg/g dry weight for fish diets and 2.9 µg/g dry weight for invertebrate diets. The

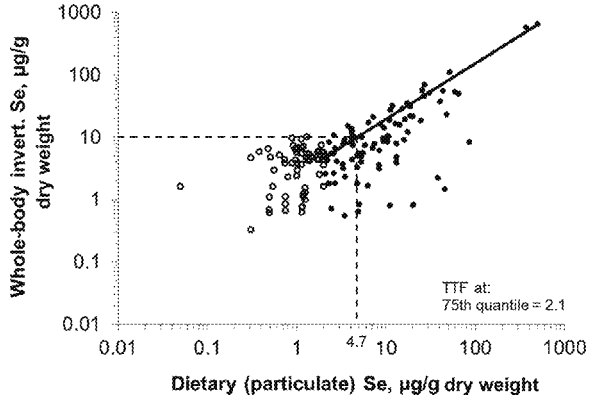

Figure 2. Relationship between whole-body invertebrate selenium (Se) and dietary (particulate) Se. Shown are the 75th quantile (solid line), reference data (○), and exposure data (●). invert. = invertebrate; TTF = trophic transfer factor.

particulate Se concentration of 2.9 µg/g dry weight, in turn, translated to waterborne Se concentrations of 2.8 and 1.7 µg/L for lotic and lentic waters, respectively, and to 3.0 and 0.85 µg/L for selenate and selenite, respectively.

In the field validation for lotic sites, fish Se ratios corroborated waterborne Se ratios 76% of the time, with fish and waterborne Se ratios being both >1 in 18% of the cases and <1 in 58% of the cases (Figure 5). In 18% of the cases, the waterborne Se ratio was >1 but the fish Se ratio was <1 (false positives), whereas in 6% of the cases the waterborne Se ratio was <1 but the fish Se ratio was >1 (false negatives; Figure 5). Overall, therefore, the waterborne Se ratio based on the lotic Se screening guideline was protective in 94% of the samples. For lentic sites, the fish Se ratios corroborated waterborne Se ratios 96% of the time, with 2% of the cases resulting in false positives and 2% of the cases in false negatives (Figure 5).

Sulfate-dependent screening guideline for selenate-dominated waters

Using the multiple quantile regression analysis of fish egg Se concentrations predicted as a function of waterborne selenate and sulfate concentrations to which particulates were exposed (Supplemental Data, Table S6), the following 75th quantile regression equation was derived

$$\text{Mean 75th quantile fish egg Se} = \exp[4.364 - 0.5680(\ln SO_4) + 0.4089(\ln \text{selenate})] \quad (5)$$

The fish egg Se concentrations predicted from waterborne selenate and sulfate concentrations versus fish egg Se concentrations predicted from empirical particulate Se concentrations using Equation 5 are illustrated in Figure 6.

Equation 5 can be rearranged to solve for the waterborne Se concentration which predicts a given fish egg Se concentration at a given waterborne sulfate concentration

$$\text{Waterborne Se screening guideline} = \exp\left[\frac{\ln(\text{fish egg Se guideline}) - 4.364 + 0.5680(\ln SO_4)}{0.4089}\right] \quad (6)$$

where the fish egg Se guideline is 20 µg/g dry weight and SO_4 is the waterborne sulfate concentration (milligrams per liter) of interest.

Equation 6 can be rearranged as follows

$$\text{Waterborne Se screening guideline} = \exp[2.446 \times \ln(\text{fish egg Se guideline}) - 10.67 + 1.389(\ln SO_4)] \quad (7)$$

Examples of waterborne Se screening guidelines as a function of various sulfate concentrations are provided in Table 1.

In the field validation, those data sets containing colocated waterborne sulfate concentrations were used and only data for lotic waters were included, which were primarily mountain streams where selenate is likely to be the predominant form of Se [3]. The fish Se ratios corroborated the waterborne Se ratios 88% of the time, with 7% of the cases resulting in false positives and 5% of the cases resulting in false negatives (Figure 7). Therefore, the waterborne Se ratio based on the sulfate-dependent guideline was protective in 95% of the samples. Results using the 50th and 90th quantiles of the multiple

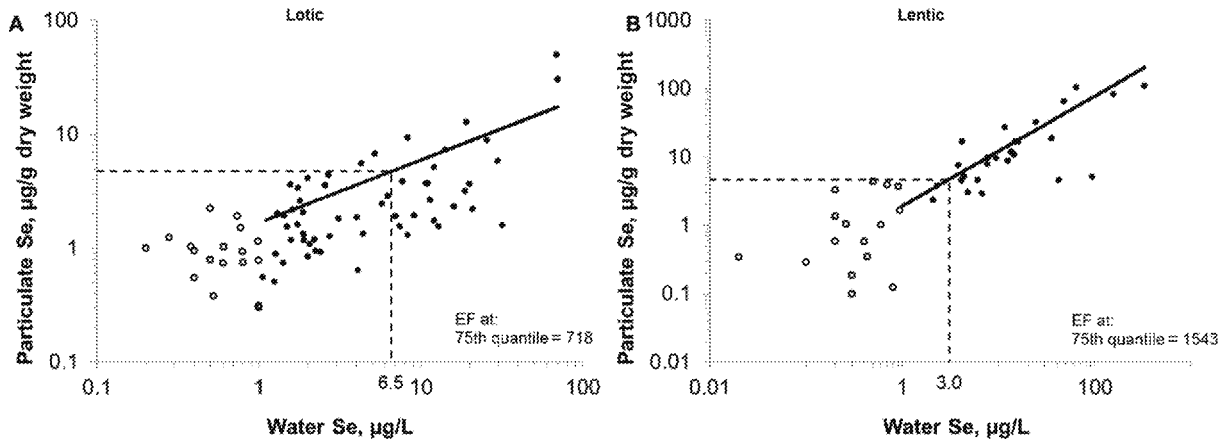

Figure 3. Relationships between particulate and waterborne selenium for (**A**) lotic sites and (**B**) lentic sites. Shown are 75th quantile (solid line), reference data (○), and exposure data (●). EF = enrichment factor; Se = selenium.

quantile regression model for deriving sulfate-dependent waterborne Se screening guidelines are discussed in Supplemental Data, Section S2, for comparison.

The lower bound of the sulfate-dependent model was capped at a sulfate concentration of 43 mg/L, which is the sulfate concentration that results in a guideline of 6.5 µg/L (equivalent to the lotic guideline; Table 1). The lotic Se screening guideline of 6.5 µg/L is based, in part, on the relationship between waterborne and particulate Se in a variety of lotic water bodies. Sulfate concentrations were not always reported in the data sets used to derive the lotic guideline, but reported ranges encompassed low sulfate conditions where available, for example, ranging from <5 mg/L to approximately 100 mg/L [37,38] and from <5 mg/L to approximately 50 mg/L [39]. As such, we expect that the lotic Se screening guideline of 6.5 µg/L would be protective at low sulfate concentrations. The sulfate-dependent guideline appears to have sufficient conservatism such that sulfate concentrations <43 mg/L result in unnecessarily low guidelines. This is supported by the field data used to validate the sulfate-dependent guideline. When the Se guideline is not capped at 43 mg/L, the percentage of sites with observed false negatives decreases only marginally (Figure 7, upper left quadrant), while the number of false positives increases substantially (Figure 7, lower right quadrant). Consequently, capping the guideline based on a sulfate-dependent model does not substantially reduce the level of protection provided but does reduce the number of sites that would be overprotected if the lower bound was not capped.

Table 1. Summary of waterborne selenium (Se) screening guidelines based on 1) field data for lotic and lentic sites, 2) laboratory data for selenate and selenite, and 3) waterborne sulfate concentrations for selenate

Description of approach	Waterborne Se (µg/L)
Field-based water-to-particulate Se data	
Lotic	6.5
Lentic	3.0
Laboratory-based water-to-particulate Se data	
Selenate	5.1
Selenite	1.5
Sulfate-dependent selenate guidelines	
43 mg SO_4/L (lower limit)[a]	6.5
75 mg SO_4/L	14
100 mg SO_4/L	21
150 mg SO_4/L	37
200 mg SO_4/L	55
300 mg SO_4/L	97

[a]The sulfate-dependent model is capped at a lower sulfate concentration of 43 mg/L, which results in the lotic guideline of 6.5 µg/L (see text).

DISCUSSION

Comparisons to other waterborne Se guidelines or criteria

The derived waterborne Se screening guidelines of 6.5 µg/L for lotic systems and 3.0 µg/L for lentic systems are differentiated based on relationships between particulate and waterborne Se concentrations from field data sets. Back-calculated waterborne Se concentrations of 5.1 µg/L for selenate and 1.5 µg/L for selenite were derived from laboratory data. It is unclear why these back-calculated waterborne Se concentrations based on laboratory-based selenate and selenite data are generally less than those derived from field-based lotic and lentic data, but it could be for 1 or more of the following reasons: 1) greater Se bioavailability in laboratory waters; 2) testing of single species in the laboratory that may have a higher Se bioconcentration potential than particulates in the field data sets, where multiple species occur; and/or 3) other conditions associated with laboratory exposures, such as high light intensity and addition of nutrients, that might enhance Se bioaccumulation relative to field sites. The following discussion compares these lotic and lentic guidelines of 6.5 and 3.0 µg/L to other waterborne guidelines and criteria.

The USEPA's recently finalized ambient water quality criteria for Se include a fish egg Se criterion of 15.1 µg/g dry weight as well as water Se criteria of 3.1 µg/L for lotic systems and 1.5 µg/L for lentic systems [9]. In deriving these water column criteria, the USEPA first compiled empirical Se enrichment factors for field sites (26 lentic and 39 lotic sites). The USEPA then modeled Se in the food web using trophic transfer factors based on 1) the fish species reported to be present at each of the sites for which the enrichment factors were compiled, and 2) the diet of each species, which was usually assumed based on typical diets and/or feeding behavior. The fish and invertebrate (dietary) trophic transfer factors were compiled from the literature and government reports. The fish trophic transfer factors were based on whole-body Se concentrations, so a whole-body to egg conversion factor was also applied. The USEPA treated the enrichment factors and trophic transfer factors as constants regardless of the Se concentrations at the site. This produced predictions of

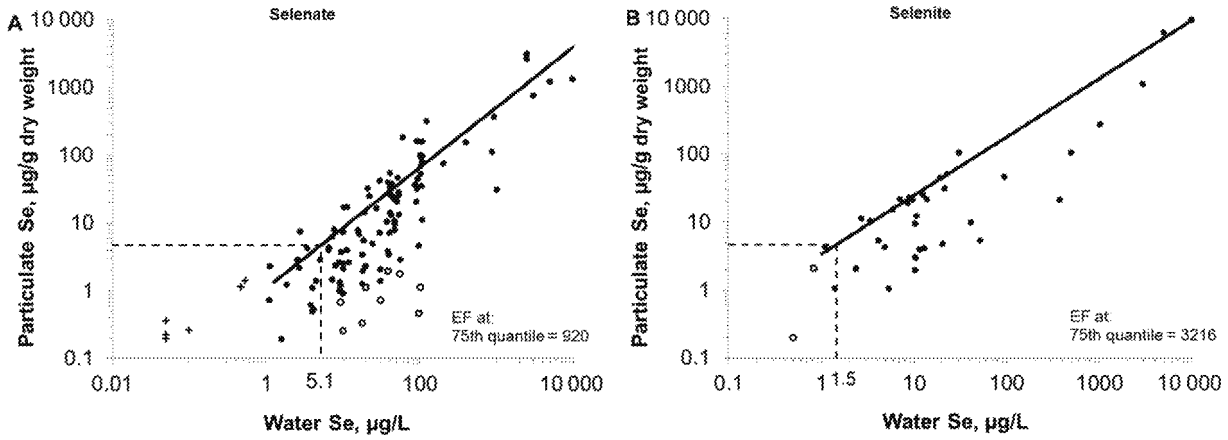

Figure 4. Relationships between particulate and waterborne selenium from laboratory studies with (**A**) selenate and (**B**) selenite. (**A**) Shown are data for 75th quantile (solid line), sulfate ≤267 mg/L (●), sulfate >267 mg/L (○), and controls (+). (**B**) Shown are data for 75th quantile (solid line), controls (○), and exposure data (●). EF = enrichment factor; Se = selenium.

water column Se concentrations that could result in the fish egg Se criterion of 15.1 µg/g dry weight for 39 lotic sites and 26 lentic sites. The water column criteria were then set equal to the 20th percentiles of these distributions for lotic and lentic sites, which were 3.1 and 1.5 µg/L, respectively.

One uncertainty in the approach used by the USEPA [9] is that it can provide counterintuitive results, which is largely a result of the inverse relationship between enrichment factors and water column concentrations. For example, the 2 lentic sites with the lowest back-translated water Se concentrations are reference lakes (Badin Lake and High Rock Lake, NC, USA [40]). The reported water Se concentrations in these lakes were 0.32 and 0.67 µg/L, respectively, and fish Se concentrations were well below criteria (based on measured muscle Se concentrations, the USEPA predicted egg Se concentrations that ranged 3.2–5.8 µg/g dry wt in Badin Lake and 3.1–6.1 µg/g dry wt in High Rock Lake). However, the USEPA's model predicted that water Se concentrations of 0.27 and 0.68 µg/L would be required to achieve the fish egg Se criterion of 15.1 µg/g dry weight. As such, the model is predicting that water column Se concentrations need to be essentially unchanged, in fact lower in Badin Lake, for the fish egg Se criterion of 15.1 µg/g dry weight to be achieved in these 2 reference water bodies. The reason is that the empirical enrichment factors for Badin Lake and High Rock Lake were 12 480 and 4990 L/kg, respectively, and it was assumed that the enrichment factors would be of the same magnitude if the lakes had a fish egg Se concentration of 15.1 µg/g dry weight.

If the USEPA's fish egg Se criterion of 15.1 µg/g dry weight is entered into the quantile regression model described herein, the resulting lotic and lentic water column concentrations are 2.8 and 1.7 µg/L, respectively, which are similar to the USEPA's lotic and lentic criteria of 3.1 and 1.5 µg/L. However, they are mainly similar because, as noted, the USEPA's criteria are based on the relatively nonconservative 20th percentiles of the lotic and lentic sites they evaluated (i.e., by their definition, 20% of sites would not be protected by their water column criteria). If the 5th percentile of their distributions had been used, for example, the lotic and lentic criteria would have been approximately 1.3 and 0.8 µg/L for lotic and lentic water bodies, respectively. To put those concentrations into perspective, of all the field-based water column Se concentrations of 1.5 µg/L and lower ($n = 131$) that were compiled in Appendix I of USEPA [9], just 4 (3%) had an egg Se concentration >15.1 µg/g dry weight. Or, to look at it another way, the mean predicted egg Se concentrations in the same data set averaged 6.6 µg/g dry weight for water bodies with Se concentrations ≤1.5 µg/L (which averaged 0.87 µg/L). The USEPA selected the 20th percentile for deriving water column Se criteria so that the criteria were not within or approaching reference water body ranges.

In addition to differences in the model used herein versus that used by the USEPA, the USEPA's waterborne Se criteria are derived from a fish egg Se criterion of 15.1 µg/g dry weight, whereas a fish egg Se guideline of 20 µg/g dry weight was used in the present evaluation [30]. The key difference between these values is the inclusion of a white sturgeon (*Acipenser transmontanus*) EC10 based on an incomplete concentration–response relationship; this species has the lowest EC10 (15.6 µg/g dry wt) in the database used in the USEPA's SSD, while the second lowest species mean EC10 is 20.6 µg/g dry weight [9]. As noted, inserting the USEPA's fish egg Se criterion of 15.1 µg/g dry weight into our model results in back-calculated lotic and lentic waterborne Se concentrations of 2.8 and 1.7 µg/L, respectively, which are in the same range as those proposed by the USEPA. At first glance, this may seem to suggest that the USEPA's lotic and lentic Se criteria are not inconsistent with what would be derived from our model if the

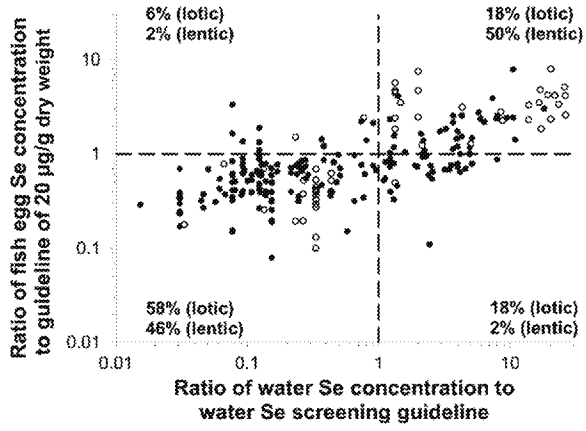

Figure 5. Relationship between mean fish egg selenium (Se) and waterborne Se ratios based on lotic (●) and lentic (○) screening guidelines. Dashed lines indicate waterborne Se ratios (vertical) and egg Se ratios (horizontal). Quadrants show agreement (upper right and lower left) or disagreement (lower right and upper left) between water and egg Se guideline comparisons, with percentage of observations in each quadrant indicated. Upper left quadrant is the false-negative quadrant.

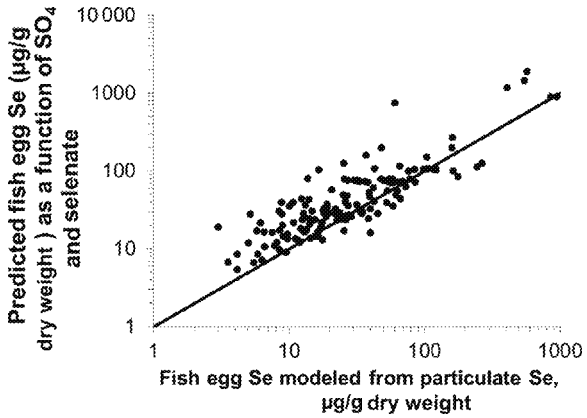

Figure 6. Comparison of 75th quantile fish egg selenium (Se) concentrations predicted from waterborne sulfate (SO_4) and selenate concentrations (*y* axis) versus fish egg Se concentrations predicted from empirical particulate Se concentrations (*x* axis). Solid line represents 1:1 agreement.

egg Se criterion of 15.1 µg/g dry weight is used. As discussed further, however, selection of an appropriate quantile to use in the model may not be independent of the fish egg Se criterion that is used.

We selected the 75th quantile in our model because use of a higher quantile, such as the 90th, resulted in waterborne Se concentrations found in reference areas. We attribute this to compounding quantiles at each step of the model. The 75th quantile was deemed to be a reasonably protective quantile that also provided Se concentrations in the food chain that were toxicologically sensible. For example, the 75th quantile resulted in a back-calculated invertebrate Se concentration of 10 µg/g dry weight, which is consistent with the lower threshold for diet-borne Se toxicity to sensitive fish including the white sturgeon (Figure 1B). These observations were based on an egg Se guideline of 20 µg/g dry weight, but that does not mean the same quantile should be considered for an alternative egg Se

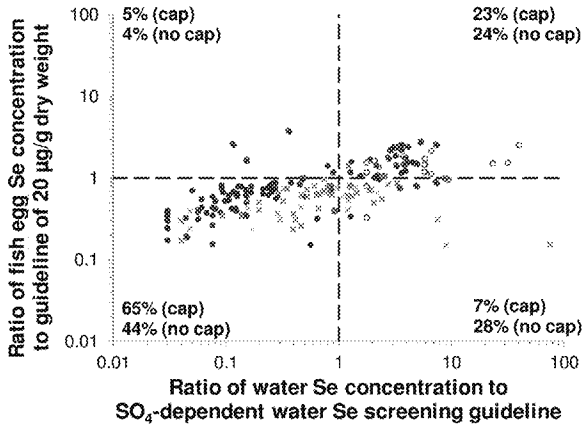

Figure 7. Relationship between mean fish egg selenium (Se) and waterborne Se ratios based on the sulfate-dependent screening guideline. Ratios based on the sulfate (SO_4)–dependent guideline capped at 43 mg SO_4/L are filled symbols (●); ratios based on sulfate-dependent guideline not capped at 43 mg SO_4/L are expressed as open symbols (○) where water Se was >6.5 µg/L and sulfate was >43 mg/L and as crossed symbols (×) where water Se was <6.5 µg/L and sulfate was <43 mg/L. Dashed lines indicate waterborne Se ratios (vertical) and egg Se ratios (horizontal). Quadrants show agreement (upper right and lower left) or disagreement (lower right and upper left) between water and egg Se guideline comparisons, with percentages of observations in each quadrant indicated.

concentration of interest, such as the USEPA's criterion of 15.1 µg/g dry weight. If the USEPA's criterion of 15.1 µg/g dry weight is used, the associated Se concentrations in invertebrates and particulates using the 75th quantile models would be 6.5 and 2.9 µg/g dry weight, respectively. These are below the dietary Se toxicity threshold for fish and near or within the range of reference area invertebrate and particulate Se concentrations in at least some geographic regions [19,29,41,42]. As such, a lower quantile would be justified when considering a lower fish egg Se criterion.

In addition, we note that the diet-to-fish egg trophic transfer factors vary depending on the fish egg Se toxicity threshold used. Based on the 75th quantile of the regression model, a fish egg Se threshold of 20 µg/g dry weight is associated with a trophic transfer factor of 2.0, while a threshold of 15.1 µg/g dry weight is associated with a trophic transfer factor of 2.3. Although we earlier noted the added uncertainty in the white sturgeon EC10 because of the partial concentration–response relationship, it does appear to be among the more sensitive species tested to date. Based on the paired sturgeon egg and diet-borne Se concentrations reported in Linville [43], diet-to-sturgeon egg trophic transfer factors ranged from 0.22 to 0.82. Fish species with relatively low egg Se EC10s may partition a lower proportion of their body burden to eggs than fish species with relatively high egg Se EC10s. For example, the egg Se EC10 for mountain whitefish (*Prosopium williamsoni*) is >33 µg/g dry weight [44], among the highest for fish species tested. Also, egg Se concentrations for this species are often >20 µg/g dry weight in reference waters and typically approximately 8-fold greater than muscle Se concentrations [36,44]. In contrast, the species with the lowest EC10s in the SSD [30] also have the lowest ratios of egg Se concentrations to either whole-body or muscle Se concentrations (Supplemental Data, Figure S12). It is possible that species with higher ratios of egg/ovary to whole-body/muscle Se concentrations have a greater tolerance to Se; however, to our knowledge, this has not yet been demonstrated.

Lastly, the BCMOE's ambient aquatic life guidelines for Se include an alert concentration of 1 µg/L and a guideline of 2 µg/L [7]. These values are based on multiple lines of evidence, including food-web modeling and relationships between aquatic life impacts and corresponding water Se concentrations. The BCMOE did not derive waterborne Se guidelines that varied by site type but noted that the guideline was intended to protect the fish-inhabiting lentic environments, with a greater Se bioaccumulation potential. If the BCMOE's fish egg Se guideline of 11 µg/g dry weight was considered, which includes a safety factor of 2 in its derivation (British Columbia does not use the SSD for guideline derivation), the lotic and lentic back-calculated waterborne Se concentrations using our model are 1.1 and 0.9 µg/L, respectively. As previously discussed (see *Water Se screening guideline development* section), waterborne Se concentrations of this magnitude can be considered reference water body concentrations and are thus not useful as screening-level guidelines.

Important factors in deriving waterborne Se screening guidelines

Although many factors influence Se speciation, bioavailability, and bioaccumulation in aquatic systems, 3 that should currently be considered in development of waterborne Se screening thresholds are as follows: 1) differences between lotic and lentic sites, 2) the influence of exposure concentration on Se partitioning among compartments, and 3) the influence of sulfate on selenate bioavailability. Each of these factors was

incorporated into the waterborne Se screening guidelines recommended in the present study.

Regarding differences between lotic and lentic sites, use of colocated waterborne and particulate Se data from lotic and lentic sites still appears to be the most practical approach in accounting for this difference. There is clearly a continuum of Se bioconcentration potential between these 2 site types, but separating particulate Se data by site type facilitates development of waterborne Se screening guidelines that differ appropriately for lotic and lentic sites. However, more research is needed to refine these categories or even eliminate them as expanding research on Se speciation, fate, and biological interactions increases our ability to mechanistically predict Se bioconcentration potential across differing site conditions.

The influence of exposure concentration on Se partitioning among compartments is best addressed by considering relationships between Se concentration at each step in the food chain versus its corresponding exposure concentration, rather than simplifying these relationships to single ratios. Because Se enrichment factors and trophic transfer factors are inversely related to exposure concentration, selection of a single enrichment factor or trophic transfer factor for use in the multistep Se partitioning models is inappropriate. For example, selecting a high percentile enrichment factor or trophic transfer factor may be applicable only to background or reference site Se conditions, whereas selection of a low to even moderate percentile may underestimate Se exposure potential. Use of the regression-based approach quantitatively reduces that uncertainty.

Although both the BCMOE and the USEPA recognize the importance of sulfate on selenate bioavailability, neither derived nor recommended sulfate-dependent Se guidelines/criteria. We demonstrate that the influence of sulfate on the bioconcentration of selenate can be modeled quantitatively, as is done for hardness-dependent guidelines or other toxicity-modifying factors. A waterborne Se screening guideline, such as that derived in the present evaluation, would be applicable only to waters where Se enters a well-oxygenated, lotic receiving water as selenate and in which selenate is the dominant Se species at the interface between waterborne Se and particulate Se. Because of methodological challenges and high costs, it is difficult to evaluate the influence of sulfate on selenate bioconcentration and transfer up the food chain. However, because increasing sulfate concentrations reduce selenate bioconcentration in particulates at the base of food chains, it was possible to model selenate bioconcentration in particulates as a function of waterborne selenate and sulfate concentrations. As additional data on the influence of sulfate on selenate bioconcentration become available, this model can be further refined.

One observation with the recommended sulfate-dependent water Se screening guideline is that it increases at a proportionally greater rate than the waterborne sulfate concentration. For example, while holding the fish egg Se concentration constant, a 2-fold increase in the waterborne sulfate concentration results in a 2.6-fold increase in the sulfate-dependent water Se screening guideline. Alternatively, if the waterborne selenate concentration is held constant, a 2-fold increase in the waterborne sulfate concentration results in a predicted fish egg Se concentration that decreases by a factor of 0.33 (i.e., it decreases at a proportionately lesser rate). Thus, the sulfate-dependent water Se guideline responds more than proportionately to the waterborne sulfate concentration because the predicted fish egg Se concentration responds less than proportionately to the waterborne sulfate concentration. This observation of the model can be compared to empirical data that comprise one component of the model. For example, the slope of the relationship between the natural logarithms of the predicted fish egg Se concentrations and waterborne sulfate concentrations is –0.568 when the waterborne selenate concentration is held constant. This slope from the model falls within the range of empirical slopes (–0.836 to –0.424) when the natural logarithms of algae and macrophyte Se are regressed versus waterborne sulfate concentrations and waterborne selenate is held constant (Supplemental Data, Figure S13). This is perhaps not surprising because the algae and macrophyte data provide the foundation for the sulfate-dependent model, but this supports the idea that the relationship between the sulfate-dependent water Se screening guideline and waterborne sulfate concentrations is consistent with the empirical data.

Outlook and ongoing research needs

Methods for linking waterborne Se concentrations to fish tissue Se concentration range from conservative screening models to site-specific models. Screening models, such as those described in the present study, are intended to determine whether site-specific studies and models are necessary, while site-specific models currently require measurement of Se concentrations in at least one component of the aquatic food web at the site of interest. The ecosystem-scale Se model of Presser and Luoma [12], for example, requires site-specific measurement of Se in particulates at the base of the food web, whereas the Bayesian Monte Carlo model described in Brix et al. [2] requires site-specific measurement of Se in fish tissue.

Currently, models for predicting Se bioaccumulation potential in aquatic food webs as a function of site-specific biogeochemical characteristics are unavailable. The development of models for predicting Se enrichment at the base of aquatic food webs as a function of site-specific chemistry data, including Se speciation and factors that modify Se bioavailability, is the next step for advancing our ability to evaluate site-specific Se bioaccumulation potential. Such modeling at the base of the food web, coupled with an understanding of Se trophic transfer in different food webs (as described in Presser and Luoma [12]), would provide a more flexible and cost-effective tool for Se evaluations over a broad range of biogeochemical conditions.

Such models can be enhanced by considering the biokinetics of Se uptake and elimination at the base of the aquatic food web and through successive trophic levels. Selenium concentrations in surface waters are often temporally variable; thus, it is important to understand how Se concentrations in different food-web components respond to that variability and, ultimately, how those Se concentrations relate to the critical exposure period for fish species of interest at a given site. DeForest et al. [45], for example, conducted an evaluation of existing biokinetic data for Se and found that periphyton-based food webs respond differently to changes in waterborne Se concentrations compared with phytoplankton-based food webs, which is predominantly because of differences in Se uptake rates. More biokinetic Se studies are necessary for a broader range of primary producers and over a broader range of water chemistry conditions. These studies could also help in the development of predictive models of Se enrichment at the base of the food web under varying physicochemical conditions.

Supplemental Data—The Supplemental Data are available on the Wiley Online Library at DOI: 10.1002/etc.3793.

Acknowledgment—The present study was supported by member companies of the North American Metals Council–Selenium Working Group, Rio Tinto, and the National Mining Association.

Data availability—Data are available as Supplemental Data.

REFERENCES

1. Janz DM, DeForest DK, Brooks ML, Chapman PM, Gilron G, Hoff D, Hopkins WA, McIntyre DO, Mebane CA, Palace VP, Skorupa JP, Wayland M. 2010. Selenium toxicity to aquatic organisms. In Chapman PM, Adams WJ, Brooks ML, Delos CG, Luoma SN, Maher WA, Ohlendorf HM, Presser TS, Shaw DP, eds, *Ecological Assessment of Selenium in the Aquatic Environment*. SETAC, Pensacola, FL, USA, pp 141–231.
2. Brix KV, Toll JE, Tear LM, DeForest DK, Adams WJ. 2005. Setting site-specific water-quality standards by using tissue residue thresholds and bioaccumulation data. Part 2. Calculating site-specific selenium water-quality standards for protecting fish and birds. *Environ Toxicol Chem* 24:231–237.
3. Stewart R, Grosell M, Buchwalter D, Fisher N, Luoma S, Mathews T, Orr P, Wang W-X. 2010. Bioaccumulation and trophic transfer of selenium. In Chapman PM, Adams WJ, Brooks ML, Delos CG, Luoma SN, Maher WA, Ohlendorf HM, Presser TS, Shaw DP, eds, *Ecological Assessment of Selenium in the Aquatic Environment*. SETAC, Pensacola, FL, USA, pp 93–139.
4. Lillebo HP, Shaner S, Carlson D, Richard N, DuBowy P. 1988. Regulation of agricultural drainage to the San Joaquin River. Order W.Q. 85-1. State Water Resources Control Board, Sacramento, CA, USA.
5. Lemly AD. 1993. Guidelines for evaluating selenium data from aquatic monitoring and assessment studies. *Environ Monit Assess* 28:83–100.
6. DeForest DK, Brix KV, Adams WJ. 1999. Critical review of proposed residue-based selenium toxicity thresholds for freshwater fish. *Hum Ecol Risk Assess* 5:1187–1228.
7. British Columbia Ministry of Environment. 2014. Water quality guidelines for selenium. Technical report update. Fort St John, BC, Canada.
8. Chapman PM, Adams WJ, Brooks ML, Delos CG, Luoma SN, Maher WA, Ohlendorf HM, Presser TS, Shaw DP. 2010. *Ecological Assessment of Selenium in the Aquatic Environment*. CRC, Boca Raton, FL, USA.
9. US Environmental Protection Agency. 2016. Aquatic life ambient water quality criterion for selenium—Freshwater 2016. EPA 822-R-16-006. Washington, DC.
10. Australian and New Zealand Environment and Conservation Council and Agriculture and Resource Management Council of Australia and New Zealand. 2000. Australia and New Zealand guidelines for fresh and marine water quality. Canberra, Australia.
11. Department of Water Affairs and Forestry. 1996. South African water quality guidelines. Vol 7: Aquatic ecosystems. Pretoria, South Africa.
12. Presser TS, Luoma SN. 2010. A methodology for ecosystem-scale modeling of selenium. *Integr Environ Assess Manag* 6:685–710.
13. Lo BP, Elphick JR, Bailey HC, Baker JA, Kennedy CJ. 2015. The effect of sulphate on selenate bioaccumulation in two freshwater primary producers: A duckweed, *Lemna minor*, and a green alga, *Pseudokirchneriella subcapitata*. *Environ Toxicol Chem* 34:2841–2845.
14. Williams MJ, Ogle RS, Knight AW, Burau RG. 1994. Effects of sulfate on selenate uptake and toxicity in the green alga *Selenastrum capricornutum*. *Arch Environ Con Tox* 17:449–453.
15. Canadian Council of Ministers of the Environment. 2007. Canadian water quality guidelines for the protection of aquatic life: Summary table. Winnipeg, MB, Canada.
16. US Environmental Protection Agency. 2016. National recommended water quality criteria—Aquatic life criteria table. Washington, DC. [cited 2016 November 30]. Available from: https://www.epa.gov/wqc/national-recommended-water-quality-criteria-aquatic-life-criteria-table
17. Skorupa JP, Ohlendorf HM. 1991. Contaminants in drainage water and avian risk thresholds. In Dinar A, Zilberman D, eds, *The Economics and Management of Water and Drainage in Agriculture*. Kluwer Academic, Boston, MA, USA, pp 345–368.
18. Ohlendorf HM, Santolo GM. 1994. Kesterson Reservoir—Past, present, and future: An ecological risk assessment. In Frankenberger WT, Benson S, eds, *Selenium in the Environment*. Marcell Dekker, New York, NY, USA, pp 69–118.
19. Orr PL, Wiramanaden CIE, Paine MD, Franklin W, Fraser C. 2012. Food chain model based on field data to predict westslope cutthroat trout *(Oncorhynchus clarkii lewisi)* ovary selenium concentrations from water selenium concentrations in the Elk Valley, British Columbia. *Environ Toxicol Chem* 31:672–680.
20. Adams WJ, Brix KV, Cothern KA, Tear LM, Cardwell RD, Fairbrother A, Toll JE. 1998. Assessment of selenium food chain transfer and critical exposure factors for avian wildlife species: Need for site-specific data. In Little EE, Delonay AJ, Greenberg BM, eds, *Environmental Toxicology and Risk Assessment*, Vol 7. American Society for Testing and Materials, Philadelphia, PA, pp 312–342.
21. Toll JE, Tear LM, DeForest DK, Brix KV, Adams WJ. 2005. Setting site-specific water-quality standards by using tissue residue thresholds and bioaccumulation data. Part 1. *Methodology. Environ Toxicol Chem* 24:223–229.
22. Reinfelder JR, Fisher NS, Luoma SN, Nichols JW, Wang W-X. 1998. Trace element trophic transfer in aquatic organisms: A critique of the kinetic model approach. *Sci Total Environ* 219:117–135.
23. Wang W-X. 2002. Interactions of trace metals and different marine food chains. *Mar Ecol-Prog Ser* 243:295–309.
24. Luoma SN, Rainbow PS. 2005. Why is metal bioaccumulation so variable? Biodynamics as a unifying concept. *Environ Sci Technol* 39:1921–1931.
25. Besser JM, Canfield TJ, La Point TW. 1993. Bioaccumulation of organic and inorganic selenium in a laboratory food chain. *Environ Toxicol Chem* 12:57–72.
26. Guan R, Wang W-X. 2004. Dietary assimilation and elimination of Cd, Se, and Zn by *Daphnia magna* at different metal concentrations. *Environ Toxicol Chem* 23:2689–2698.
27. DeForest DK, Brix KV, Adams WJ. 2007. Assessing metal bioaccumulation in aquatic environments: The inverse relationship between bioaccumulation factors, trophic transfer factors and exposure concentration. *Aquat Toxicol* 84:236–246.
28. McGeer JC, Brix KV, Skeaff JM, DeForest DK, Brigham SI, Adams WJ, Green A. 2003. Inverse relationship between bioconcentration factor and exposure concentration for metals: Implications for hazard assessment of metals in the aquatic environment. *Environ Toxicol Chem* 22:1017–1037.
29. Orr PL, Guiguer KR, Russel CK. 2006. Food chain transfer of selenium in lentic and lotic habitats of a western Canadian watershed. *Ecotox Environ Safe* 63:175–188.
30. DeForest DK, Gilron G, Armstrong SA, Robertson EL. 2012. Species sensitivity distribution (SSD) evaluation for selenium in fish eggs: Considerations for development of a Canadian tissue-based guideline. *Integr Environ Assess Manag* 8:6–12.
31. Seiler RL, Skorupa JP, Naftz DL, Nolan BT. 2003. Irrigation-induced contamination of water, sediment, and biota in the western United States—Synthesis of data from the National Irrigation Water Quality Program. US Geological Survey Professional Paper 1655. Denver, CO.
32. R Core Team. 2013. *R: A Language and Environment for Statistical Computing*. Ver 3.0.1. R Foundation for Statistical Computing, Vienna, Austria.
33. Koenker R. 2013. *quantreg: Quantile Regression*. Ver 5.02. R Foundation for Statistical Computing, Vienna, Austria.
34. Skorupa JP. 1998. Selenium. National Irrigation Water Quality Program information report 3: Guidelines for interpretation of the biological effects of selected constituents in biota, water, and sediment. US Department of the Interior, Denver, CO.
35. Golder Associates. 2013. Statistical analysis of selenium bioaccumulation in fish. Prepared for the North American Metals Council–Selenium Working Group. Edmonton, Alberta, Canada.
36. deBruyn A, Hodaly A, Chapman P. 2008. Tissue selection criteria: Selection of tissue types for the development of a meaningful selenium tissue threshold in fish. Prepared for the North American Metals Council–Selenium Working Group, Washington, DC.
37. Hamilton SJ, Buhl KJ, Lamothe PJ. 2002. Selenium and other trace elements in water, sediment, aquatic plants, aquatic invertebrates, and fish from streams in southeastern Idaho near phosphate mining operations: June 2000. US Geological Survey, Yankton, SD, and Denver, CO.
38. Hamilton SJ, Buhl KJ. 2003. Selenium and other trace elements in water, sediment, aquatic plants, aquatic invertebrates, and fish from streams in southeastern Idaho near phosphate mining operations: September 2000. US Geological Survey. Yankton, SD.
39. Formation Environmental and HabiTech. 2012. Appendix A—Summary of exposure conditions: Surface water, sediment, tissue. Technical support document: Proposed site-specific selenium criterion, Sage and Crow Creeks, Idaho. Boulder, CO, and Laramie, WY, USA.
40. Lemly AD. 1985. Toxicology of selenium in a freshwater reservoir: Implications for environmental hazard evaluation and safety. *Ecotox Environ Safe* 10:314–338.

41. Wayland M, Crosley R. 2006. Selenium and other trace elements in aquatic insects in coal mine–affected streams in the rocky mountains of Alberta, Canada. *Arch Environ Con Tox* 50:511–522.
42. Morrissey CA, Bendell-Young LI, Elliott JE. 2005. Assessing trace-metal exposure to American dippers in mountain streams of southwestern British Columbia, Canada. *Environ Toxicol Chem* 24: 836–845.
43. Linville RG. 2006. *Effects of Excess Selenium on the Health and Reproduction of White Sturgeon (*Acipenser transmontanus*): Implications for San Francisco Bay-Delta.* University of California–Davis, Davis, CA, USA.
44. Nautilus Environmental. 2013. Evaluation of the effects of selenium on early lifestage development of mountain whitefish from the Elk Valley, BC. Burnaby, BC, Canada.
45. DeForest DK, Pargee S, Claytor C, Canton SP, Brix KV. 2016. Biokinetic food chain modeling of waterborne selenium pulses into aquatic food chains: Implications for water quality criteria. *Integr Environ Assess Manag* 12:230–246.
46. Woock SE, Garrett WR, Partin WE, Bryson WT. 1987. Decreased survival and teratogenesis during laboratory selenium exposures to bluegill, *Lepomis macrochirus*. *B Environ Contam Tox* 39:998–1005.
47. Doroshov S, Van Eenennaam J, Alexander C, Hallen E, Bailey H, Kroll K, Restrepo C. 1992. Development of water quality criteria for resident aquatic species of the San Joaquin River. Draft final report to the California State Water Resources Control Board for contract 7-197-250-0. University of California–Davis. Davis, CA, USA.
48. Coyle JJ, Buckler DR, Ingersoll CG, Fairchild JF, May TW. 1993. Effect of dietary selenium on the reproductive success of bluegills (*Lepomis macrochirus*). *Environ Toxicol Chem* 12:551–565.
49. Cleveland L, Little EE, Buckler DR, Wiedmeyer RH. 1993. Toxicity and bioaccumulation of waterborne and dietary selenium in juvenile bluegill (*Lepomis macrochirus*). *Aquat Toxicol* 27:265–280.
50. McIntyre DO, Pacheco MA, Garton MW, Wallschläger D, Delos CG. 2008. Effect of selenium on juvenile bluegill sunfish at reduced temperature. US Environmental Protection Agency, Washington, DC.
51. Hamilton SJ, Buhl KJ, Faerber NL, Wiedmeyer RH, Bullard FA. 1990. Toxicity of organic selenium in the diet to chinook salmon. *Environ Toxicol Chem* 9:347–358.
52. Ogle RS, Knight AW. 1989. Effects of elevated foodborne selenium on growth and reproduction of the fathead minnow (*Pimephales promelas*). *Arch Environ Con Tox* 18:795–803.
53. Teh SJ, Deng X, Deng D-F, Teh F-C, Hung SSO, Fan TW-M, Liu J, Higashi RM. 2004. Chronic effects of dietary selenium on juvenile Sacramento splittail (*Pogonichthys macrolepidotus*). *Environ Sci Technol* 38:6085–6093.
54. Tashjian DH, Teh SJ, Sogomonyan A, Hung SSO. 2006. Bioaccumulation and chronic toxicity of dietary L-selenomethionine in juvenile white sturgeon (*Acipenser transmontanus*). *Aquat Toxicol* 79:401–409.
55. Hardy RW, Oram LL, Möller G. 2010. Effects of dietary selenomethionine on cutthroat trout *(Oncorhynchus clarki bouvieri)* growth and reproductive performance over a life cycle. *Arch Environ Con Tox* 58:237–245.