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Assessing metal bioaccumulation in aquatic environments: The inverse relationship between bioaccumulation factors, trophic transfer factors and exposure concentration

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Abstract

Bioaccumulation potential in aquatic biota is typically expressed using ratios of chemical concentrations in organism tissue (typically whole body) relative to chemical exposure concentrations, such as bioconcentration factors (BCFs). Past reviews of metal BCFs for aquatic biota, which account for water-only exposures, have shown that BCFs are often highly variable between organisms and generally inversely related to exposure concentration. This paper further evaluates trends in metal bioaccumulation data by evaluating data for bioaccumulation factors (BAFs) and trophic transfer factors (TTFs). Bioaccumulation factor data were compiled from field studies that account for combined waterborne and dietary metal exposures. Trophic transfer factor data for metals were compiled from laboratory studies in which aquatic food chains were simulated. Natural aquatic food webs are rarely sufficiently understood to properly evaluate exact predator–prey relationships (i.e., TTFs). Results indicate that field BAFs, like laboratory BCFs, tend to be significantly ($p \leq 0.05$) inversely related to exposure concentration. Bioaccumulation factors are frequently 100–1000 times larger than BCFs for the same metal and species. This difference is attributed to both lower exposure levels in the field and inclusion of the dietary exposure route. Trophic transfer factors for the metals reviewed, including selenium and methyl mercury were also observed to be inversely related to exposure concentration, particularly at lower exposure concentrations. These inverse relationships have important implications for environmental regulations (e.g., hazard classification and tissue residue-based water quality criteria) and for the use of metal bioaccumulation data in site-specific environmental evaluations, such as ecological and human health risk assessments. Data presented indicate that for metals and metalloids, unlike organic substances, no one BAF or TTF can be used to express bioaccumulation and/or trophic transfer without consideration of the exposure concentration.

Keywords: Metals; Bioaccumulation; Trophic transfer; Hazard assessment; Risk assessment

1. Introduction

Current aquatic hazard identification procedures for chemical contaminants, such as those developed by the OECD, are based on persistence, bioaccumulation, and toxicity (PBT). The basic premise behind these procedures is that substances which are toxic present greater hazard when they are both persistent and bioaccumulative. Given the large number of chemicals currently in use worldwide, and the extremely large number of new chemicals developed every year, defining critical levels of persistence, bioaccumulation potential, and toxicity must be generalized for practicality. For metals, however, defining critical PBT levels is problematic because (1) metals are naturally persistent in the environment; (2) both essential and non-essential metals are naturally bioaccumulated and internally regulated using different strategies (e.g., active excretion, storage); and (3) the toxicity of metals is highly influenced by geochemical factors that influence metal bioavailability.

This paper focuses on the uncertainties in using the bioaccumulation component of PBT to regulate metals. Under the current PBT classification scheme, bioaccumulation potential is defined based on the ratio of a chemical's concentration in an organism to the chemical concentration in the water to which the organism was exposed (typically termed a bioconcentration fac-

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tor, or BCF) (Adams et al., 2000). The chemical concentration in the organism has traditionally been expressed on a whole body basis, likely because the PBT concept was originally developed with narcotic, lipophilic organic chemicals in mind. McGeer et al. (2003) compiled metal BCF data for a variety of aquatic organisms, on a whole body basis, and generally observed an inverse relationship between BCF and exposure concentration. They noted that BCFs are highest (indicating hazard) at low exposure concentrations (low potential for toxicity) and lowest (indicating no hazard) at higher exposure concentrations where toxicity is more likely.

The first objective of this paper was to build upon the predominantly BCF-based analysis of McGeer et al. (2003), which focused on aqueous metal exposures, by evaluating additional bioaccumulation data that either implicitly or explicitly considered dietary uptake of metals. We hypothesized that trends similar to those reported in McGeer et al. (2003), i.e., increased metal uptake at lower exposure concentrations, would also be observed when organisms were exposed to dietary metals (both essential and non-essential). The basis for this hypothesis is that organisms will actively uptake essential metals at low concentrations to meet metabolic needs, and that non-essential metals are often also regulated to varying degrees because the mechanisms for regulating essential metals are not metal-specific (Phillips and Rainbow, 1989). A recent study on the relative contribution of food and water to the bioaccumulation of 27 metals in caged *Hyalella azteca* found that concentrations of cadmium, copper, and selenium were the only metals for which concentrations in food had a significant effect on whole body concentration in the amphipods (Borgmann et al., 2007), which would also suggest that, for a majority of metals, bioaccumulation trends based on dietary metal exposures would be similar to those based on aqueous metal exposures (at least for *H. azteca*). Assuming that the above hypothesis proved correct (i.e., that increased metal uptake occurs at lower exposure, including dietary, concentrations), the second objective of this paper was to evaluate how these relationships between metal uptake and exposure concentration may influence metal hazard and risk assessments.

As research on the aquatic toxicology of metals increasingly focuses on mechanisms for linking aqueous metal concentrations to critical tissue concentrations (development of the biotic ligand model being one example), we emphasize that the analyses presented in this paper are based on empirical data and whole body tissue concentrations because this is the generalized approach typically dictated under PBT-based classification schemes. As noted in McGeer et al. (2003) and Adams and Chapman (2006), mechanistic chronic toxicity and bioavailability models for metals are under ongoing development, which may provide a validated bioaccumulation criterion for use in hazard classification.

2. Methods

Before describing the methods used to compile and evaluate field-based metal bioaccumulation data for metals, key terms are first defined.

Bioconcentration factor (*BCF*): The BCF is the ratio of a chemical concentration in an organism to the concentration in water, where the chemical concentration in the aquatic organism, usually determined in laboratory studies, results from exposure to waterborne chemical (Gobas and Morrison, 2000).

Bioaccumulation factor (*BAF*): The BAF is the ratio of a chemical concentration in an organism to the concentration in water, but differs from a BCF in that the chemical concentration in the aquatic organism results from all possible routes of exposure (e.g., dietary absorption, transport across the respiratory surface) (Gobas and Morrison, 2000). The BAFs compiled in this evaluation were all calculated based on field data.

Trophic transfer factor (*TTF*): The TTF is the ratio of a substance concentration in an organism's tissue to its concentration in the organism's food item. In most of these studies the test organism is only exposed to dietary metal, while in others the test organism is exposed to both dietary and aqueous metal (in some dietary selenium studies for example, the test organism is exposed to dietary and aqueous selenium, but most of the bioaccumulated selenium is from the diet). The TTF is a general term that may include biomagnification, whereby the substance concentration in an organism is greater than in its diet, and biodilution, whereby the substance concentration in an organism is less than in its diet.

2.1. Data sources

Field data for deriving metal BAFs were compiled based on reviews of peer-reviewed scientific literature and technical reports (e.g., USGS Water-Resources Investigations reports). The metals considered were arsenic, cadmium, chromium, copper, lead, mercury, selenium, and zinc. There is inherent uncertainty in using field studies to link chemical concentrations in organism tissues to surface water concentrations because the true chemical exposure history of the organisms are unknown. Even using tissue and water concentrations sampled at the same time from the same location are uncertain because the water concentration that day may not reflect the average exposure concentration to which the organism and its food items were exposed to in the days, weeks, or months leading up to the sampling time. In this paper, tissue and surface water samples were spatially co-located and tissue and surface water samples were most often collected on the same day, or generally within 2 weeks. Metal concentrations in surface water were generally reported on a dissolved basis, although for some studies only total recoverable metal concentrations were reported. The BAFs compiled for the review and analyses in this paper represent a random selection of studies identified in the scientific and grey (e.g., government agency) literature. The intent was to identify BAF data representing a broad range in metal concentrations that have been measured in field studies. We recognize that varying water quality and biological characteristics at the various field sites will influence the BAFs calculated between studies, but field-based BAFs are site-specific and we assume that site-to-site variability will not mask overall trends in metal BAF relationships. Trophic transfer data were

Table 1
Summary of sources for bioaccumulation factor (BAF) data

Metals	Species	Reference
As, Cu, Se, Zn	*Mytilus galloprovincialis*	Andreev and Simeonov (1990)
MeHg	*Dorosoma cepedianum*, *Lepomis macrochirus*, *Micropterus dolomieui*, *Morone americana*	Becker and Bigham (1995)
Se	Unidentified crustacea, unidentified invertebrates, numerous fish species	Butler et al. (1991, 1993, 1995)
As	Zooplankton, numerous fish species	Chen and Folt (2000)
Cd, Cr, Cu, Pb	*Dreissena polymorpha pallas*	Chevreuil et al. (1996)
MeHg	Small fish species	Cleckner et al. (1998)
Se	Unidentified bivalves, Belostomatidae, chironomids, coleoptera, corixids, crustacea, hemiptera, odonata	Dileanis et al. (1992)
Se	Macroinvertebrates	Kennecott, unpublished data
MeHg, total Hg	Zooplankton, *Micropterus salmoides*	Kuwabara et al. (2005)
Se	Unidentified corixids, odonata, chironomids, crustacea, hemiptera	Lambing et al. (1988, 1994)
Cd, Pb	*Crassostrea belcheri*, *Crassostrea iredalei*	Lim et al. (1998)
Se	Unidentified bivalves, caddisflies, hemiptera	Low and Mullins (1990)
Cu	*Ostrea edulis*, *Mytilus galloprovincialis*	Martincic et al. (1986, 1987)
Cd, Cr, Cu, Pb, Zn	*Amblema plicata*, *Fusconaia flava*, *Quadrula quadrula*	Mathis and Cummings (1973)
Se	Unidentified chironomids, coleoptera, corixids, diptera, odonata	Moore et al. (1990)
Se	Unidentified corixids, odonata	Ong et al. (1991)
Se	Unidentified corixids, crustacea	Peterson et al. (1991)
Cd, Cu	*Semelle solida*, *Tagelus dombeii*	Pinochet et al. (1995)
Cu, Pb, Zn	*Mytilus edulis*	Popham and D'Auria (1982)
Se	Unidentified corixids, odonata	Rinella and Schuler (1992)
Cd, Cr, Cu, Pb, Zn	*Cerastoderma edule*, *Tellinella pulchella*	Schuhmacher et al. (1995)
Se	Unidentified corixids, odonata, chironomids, crustacea	See et al. (1992)
Total Hg	*Corbicula fluminea*, *Crassostrea gigas*, *Mytilus californianus*	SFEI (2006)
MeHg	Numerous macroinvertebrate and fish species	Slotton et al. (2004)
Cd, Cu	Numerous macroinvertebrates, *Gasterosteus aculeatus*	Smith et al. (1996)
Se	Unidentified chironomids, corixids, other invertebrates	Sorensen and Schwarzbach (1991)
Se	Unidentified snails, chironomids, hemiptera, odonata, other invertebrates	Stephens et al. (1988)
Cd	*Mytilus edulis*	Talbot (1985)
Pb	*Mytilus edulis*	Talbot (1987)
Cd, Cu, Pb, Zn	Numerous macroinvertebrate species	Timmermans et al. (1989)
As, Cd, Cr, Cu, Pb, Se, Zn	*Mercenaria mercenaria*	Trocine and Trefry (1996)
Cu	*Lymnaea ovata*, *Physa fontinalis*, *Asellus aquaticus*, *Gammarus pulex*	Van Hattum et al. (1991)
Cu, Pb, Zn	*Astyanax mexicanus*, *Cichlasoma cyanoguttatum*, *Micropterus salmoides*, *Notropos lutrensis*, *Poecilia Formosa*	Villarreal-Trevino et al. (1986)

compiled from laboratory studies so that organism tissue concentrations could be unequivocally linked to concentrations in their diets. Gut contents were not depurated from the organisms prior to whole body tissue analysis in both field and laboratory studies.

Unless otherwise noted, all BAFs and TTFs are reported on a whole body basis and expressed based on dry weight tissue concentrations. If the tissue concentration in the original study was expressed on a wet weight basis, concentrations were converted to a dry weight basis assuming a moisture content of 75%. For non-detected metal concentrations, the full reported detection limit was used to calculate the BAF or TTF.

Sources of BAF data are summarized in Table 1; sources of TTF data are provided later in the paper.

2.2. Data analysis

The BAF and TTF data were used to evaluate a variety of bioaccumulation relationships. Given the wide range of concentrations considered in the BAF dataset, values were converted to log scale to aid visual comparisons.

3. Results

3.1. Bioaccumulation factors

Field BAFs were compiled for both saltwater and freshwater species (Figs. 1 and 2). A statistically significant ($p \leq 0.05$) inverse relationship was observed for seven of eight metals in saltwater and for six of nine metals (including methyl mercury) in freshwater; corresponding regression coefficients derived to describe these relationships are provided in (Table 2). Statistically significant ($p \leq 0.05$) inverse relationships were not observed for selenium ($p = 0.07$) in saltwater and arsenic ($p = 0.24$), cadmium ($p = 0.27$), and chromium ($p = 0.07$) in freshwater. Of these, selenium in saltwater and chromium in freshwater were just marginally insignificant, while for arsenic in freshwater the absence of a wide range of water concentrations hindered evaluation of whether an inverse relationship may exist and for cadmium in freshwater the highly variable BAFs between species at low aqueous cadmium concentrations precluded observation of an inverse relationship, although the BAFs at higher cadmium concentrations suggest an inverse relationship may exist.

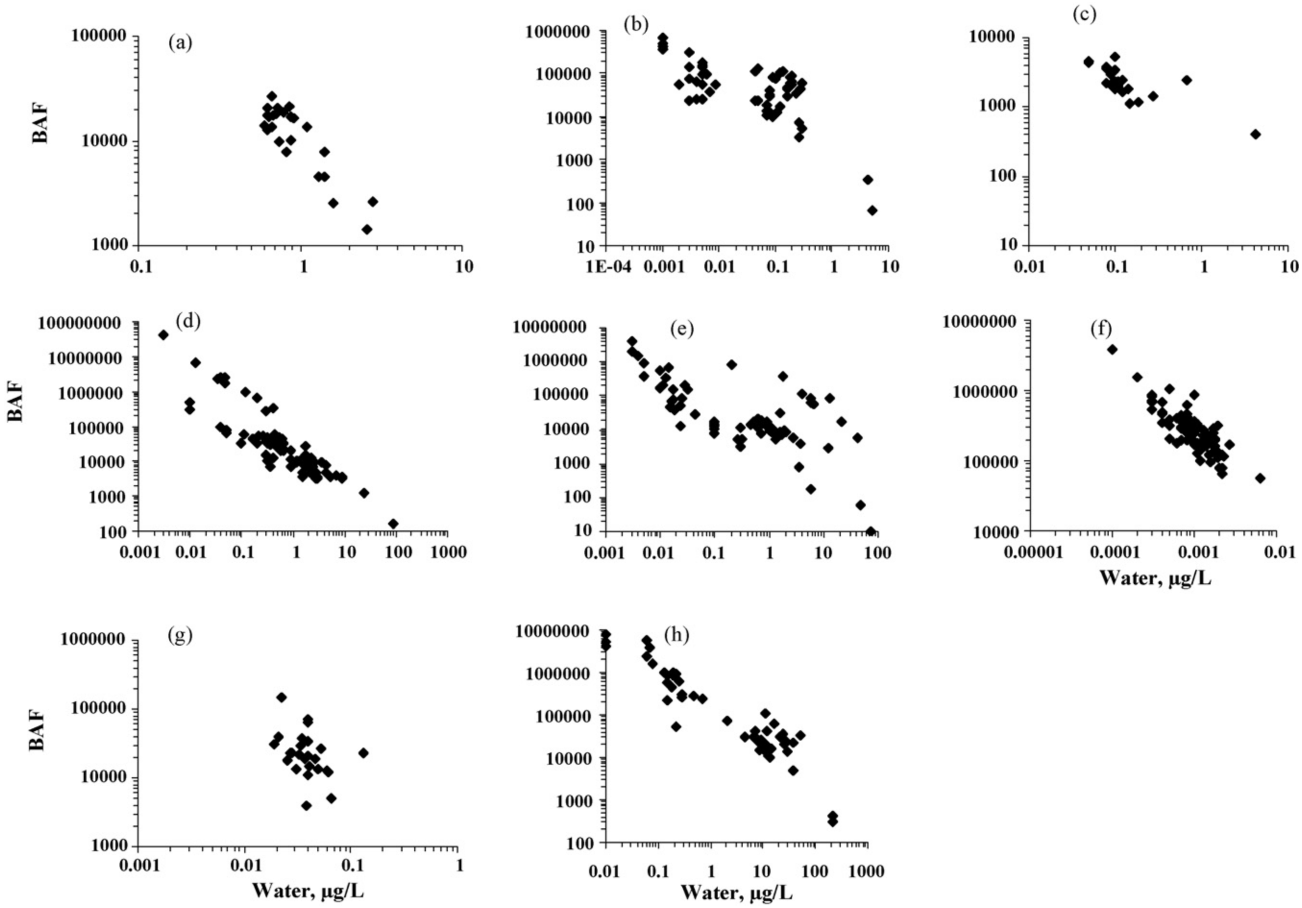

Fig. 1. Field saltwater BAFs for (a) arsenic, (b) cadmium, (c) chromium, (d) copper, (e) lead, (f) mercury, (g) selenium, and (h) zinc.

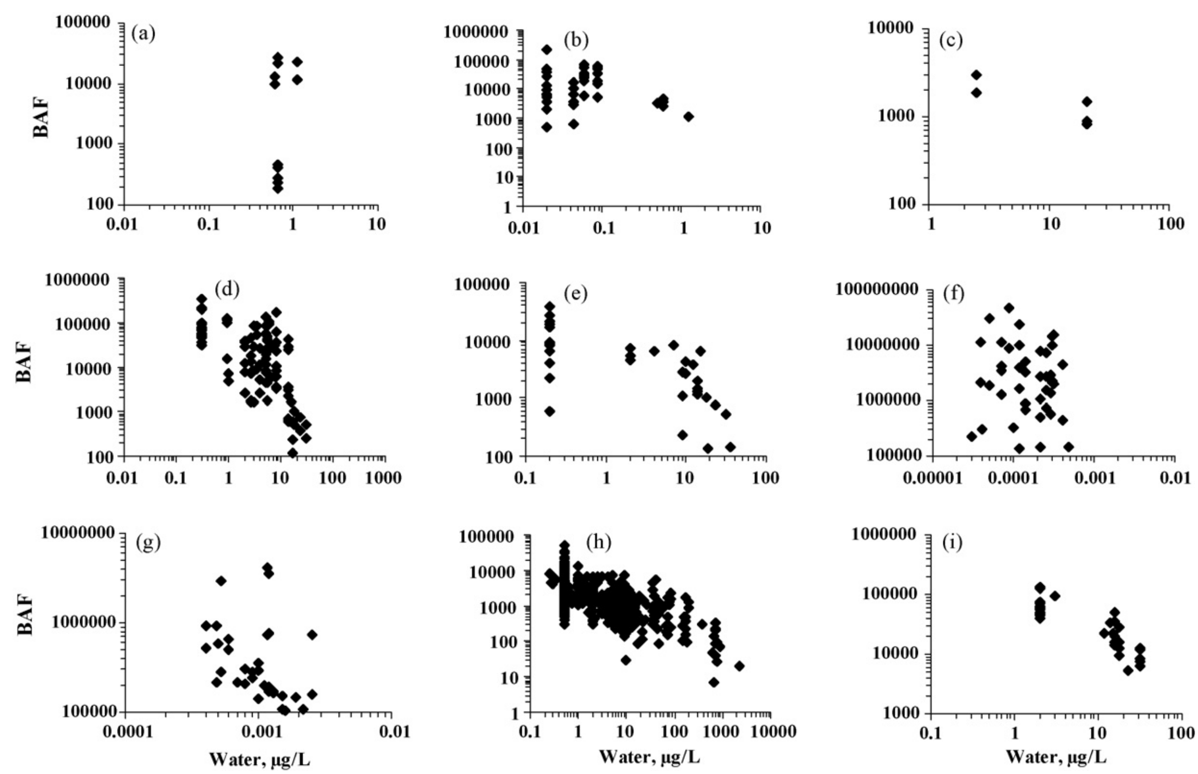

Fig. 2. Field freshwater BAFs for (a) arsenic, (b) cadmium, (c) chromium, (d) copper, (e) lead, (f) mercury (methyl), (g) mercury (total), (h) selenium, and (i) zinc.

Table 2
Regression coefficients (log:log scale) and summary statistics for empirical BAF data for freshwater and saltwater animals

Metal	Water type	Regression coefficients for log-linear relationship between BAF and exposure conc. (log:log scale)					Empirical BAFs		
		Slope	Intercept	R^2	*n*	*p*-Value	Minimum	Geometric mean	Maximum
As	FW	3.716	3.912	0.13	12	0.2	185	2,174	27,692
	SW	−1.531	3.968	0.77	25	<0.001	1,423	10,808	26,866
Cd	FW	−0.208	3.793	0.03	45	0.27	500	11,356	209,000
	SW	−0.691	3.536	0.61	59	<0.001	65	27,666	680,000
Cr	FW	−0.386	3.525	0.73	5	0.07	838	1,434	3,000
	SW	−0.428	2.994	0.67	27	<0.001	411	1,999	5,200
Cu	FW	−0.873	4.541	0.38	104	<0.001	121	11,460	333,333
	SW	−1.009	4.176	0.77	80	<0.001	157	26,016	40,666,667
Pb	FW	−0.474	3.659	0.47	36	<0.001	131	3,321	38,500
	SW	−0.723	3.969	0.56	84	<0.001	10	16,159	4,000,000
Hg (total)	FW	−0.985	2.532	0.35	41	<0.001	40,857	263,362	4,110,638
	SW	−0.893	2.722	0.70	74	<0.001	57,094	260,633	3,899,000
Hg (methyl)	FW	−0.206	5.571	0.01	43	<0.001	133,333	2,289,785	48,735,632
Se	FW	−0.443	3.384	0.45	451	<0.001	7	1,531	52,000
	SW	−0.692	3.356	0.14	25	0.07	3,947	21,634	145,455
Zn	FW	−0.669	5.038	0.75	34	<0.001	5,235	30,323	130,000
	SW	−0.886	5.189	0.89	55	<0.001	299	61,129	8,100,000

SW = saltwater; FW = freshwater.

For copper, multiple BAFs were available for several species, and for many of these exposure concentrations spanned over one order of magnitude. The relationship between copper BAFs and aqueous exposure concentration for these individual species were evaluated separately to ensure the metal-wide inverse relationship was not just an artifact of the data collected (e.g., BAFs for different species determined at different concentrations might create the perception of an inverse relationship between BAF and environmental metal concentrations). Statistically significant ($p \leq 0.05$) inverse relationships between copper BAFs and aqueous exposure concentration were observed for six of eight individual species or taxa, with the two exceptions being *Asellus* spp. (isopods) and *Lymnaea* spp. (snails) (Fig. 3). Of these two taxa, an inverse (i.e., negative slope), albeit not significant, relationship was observed for *Asellus*, and for *Lymnaea* the slope was essentially zero. For the latter, it is unclear if an inverse relationship was not observed due to uncertainties with the co-located tissue and water data, or whether *Lymnaea* tends to bioaccumulate and store copper at levels proportionate to its exposure concentration. For other metals, the absence of an inverse relationship between BAF and aqueous exposure concentration was likely to be most often due to an insufficient range in aqueous exposure concentration. In other cases, such as lead in the blue mussel *Mytilus edulis*, the lack of an inverse relationship may reflect this organism's ability to bioaccumulate and store lead at a rate proportional to its uptake.

Overall, the BAF data compiled demonstrate an inverse relationship typically exists for a variety of metals and metalloids. In some cases an inverse relationship may not have been observed because the aqueous exposure concentrations did not bracket a sufficient range, while in other cases it is possible that an organism may bioaccumulate a metal proportionate to its exposure concentration. For methyl mercury, for which a clear relationship between BAF and aqueous exposure concentration was not observed (Fig. 2), it is well known that upper levels of the food chain, and older organisms, tend to bioaccumulate more methyl mercury than organisms at lower levels in the food chain. Since the BAF dataset contains BAF data for invertebrates, small fish, and large fish, the large scatter in the relationship between BAF and exposure concentration for methyl mercury is driven more by species-related differences. Even across multiple trophic levels, total mercury BAFs ranged from about 57,000 to >4,000,000 and methyl mercury BAFs ranged from >100,000 to >48,000,000. For comparison, selenium (which is frequently reported to bioaccumulate substantially from abiotic media, including water) BAFs were observed to typically be in the range of 20–52,000 (Figs. 1 and 2). These values are no larger than those observed for all divalent metals.

The importance of the inverse relationship between BAF and exposure concentration in comparing BAFs between metals can be demonstrated using the regression data provided in Table 2. For example, at a metal concentration of 1 μg/L, which typically represents a "low" zinc concentration and a "high" mercury concentration, the highest BAFs are actually observed for zinc. Thus, a "high" BAF observed for a metal may be driven as much by the exposure concentration as other metal- and species-specific factors that influence metal bioaccumulation.

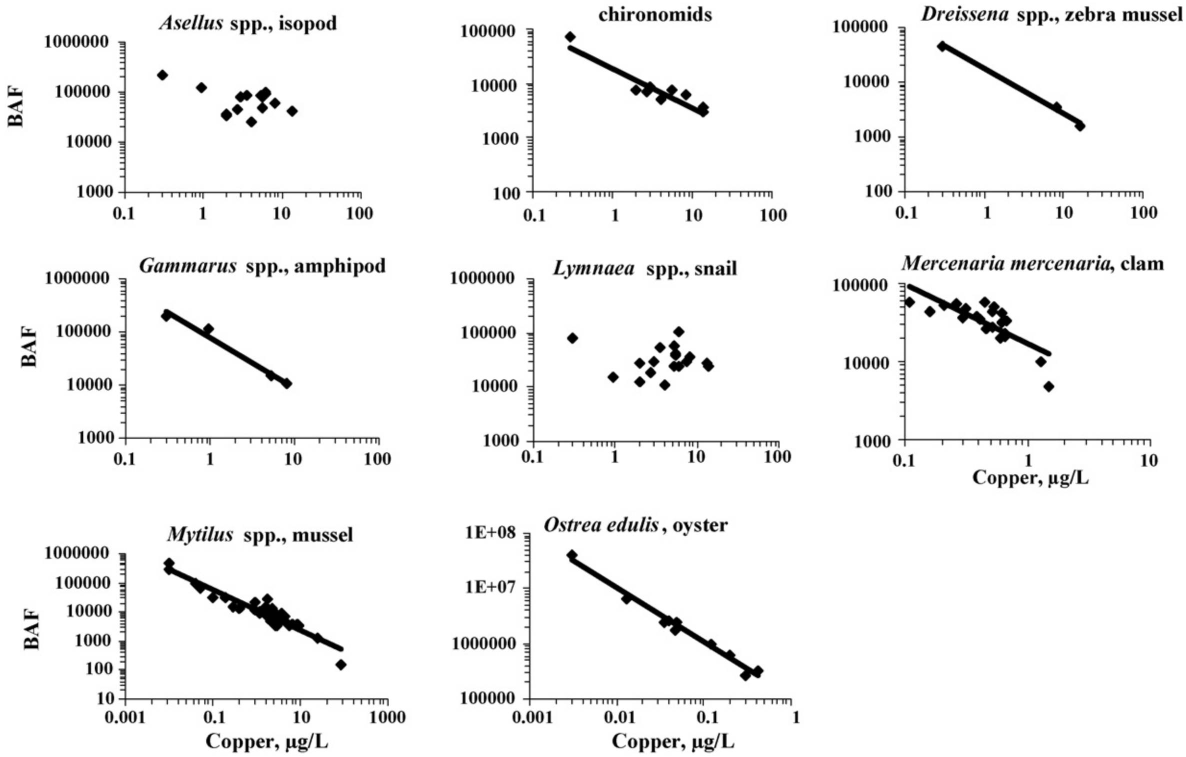

Fig. 3. Relationships between copper BAFs and exposure concentration for individual freshwater and saltwater species (for exposure concentrations spanning more than one order of magnitude). All inverse relationships statistically significant ($p \leq 0.05$) except for *Asellus* spp. and *Lymnaea* spp.

3.2. Trophic transfer factors

Trophic transfer factors from 12 laboratory studies were compiled for several divalent metals (cadmium, copper, lead, zinc), selenium, and methyl mercury. The trophic transfer of divalent metals based on whole body tissue residues, has not been extensively evaluated in the laboratory. Rainbow et al. (2006) fed the polychaete worm *Nereis virens* with *Nereis diversicolor* collected from metal-rich sediments in the southwestern UK. Trophic transfer factors for cadmium, copper, lead, and zinc were low (<1.6, dry wt. basis) and inverse relationships between TTF and dietary metal were observed for all metals. A similar pattern is observed for juvenile rainbow trout fed dietary cadmium, lead, and zinc based on data reported by Hansen et al. (2004); TTFs were <1.4 (dry wt. basis) and an inverse trend between TTF and dietary metal is observed. Guan and Wang (2004) evaluated the dietary assimilation and elimination of different cadmium, selenite, and zinc concentrations by *Daphnia magna* fed green algae. They found that the extent to which these metals were bioaccumulated by *D. magna* from their diets decreased with increasing dissolved metal concentration (i.e., the dissolved metal concentration to which the *D. magna* food items, green algae, were exposed). Reduction in the TTF with increasing exposure concentration appeared to be driven mostly by changes in metal assimilation efficiency, which decreased as the dietary metal concentration increased.

Selenium TTFs for three freshwater invertebrates: the cladoceran *D. magna* (Besser et al., 1993), the midge *Chironomus decorus* (Malchow et al., 1995), and the corixid *Trichocorixa reticulata* (Thomas et al., 1999) all showed a decreasing trend between TTF and dietary exposure concentration (Fig. 4a) with a TTF >1 only observed at the lowest (control) dietary concentrations. The higher TTFs at low dietary selenium concentrations likely reflect active regulation to meet essentiality requirements. Similarly, a decreasing trend between selenium TTF and dietary exposure concentration also was observed for bluegill (*Lepomis macrochirus*) and fathead minnow (*Pimephales promelas*) (Fig. 4b). The decreasing relationship for bluegill and fathead minnows was more subtle than that observed for invertebrates. For chinook salmon (*Oncorhynchus tshawytscha*), an inverse relationship between TTF and dietary concentration appears not to exist as the TTF at the highest dietary concentration is comparable to the control (TTF = 0.8). For all fish species, as for the invertebrates, TTFs only exceeded a value of 1.0 at the lowest (control) dietary concentrations.

Methyl mercury TTFs were identified for fathead minnows (*P. promelas*), rainbow trout (*Oncorhynchus mykiss*), and finescale dace (*Phoxinus neogaeus*). Like the selenium TTFs for most species, methyl mercury TTFs were inversely related to dietary exposure concentration (Fig. 4c). The inverse relationship was most pronounced at low dietary concentrations for the fathead minnow and finescale dace and became less pronounced in rainbow trout at increasing dietary concentrations. As would be expected since methyl mercury biomagnifies in aquatic food webs, methyl mercury TTFs were >1 at all dietary exposures evaluated (values ranged from 0.06 to 95 µg/g

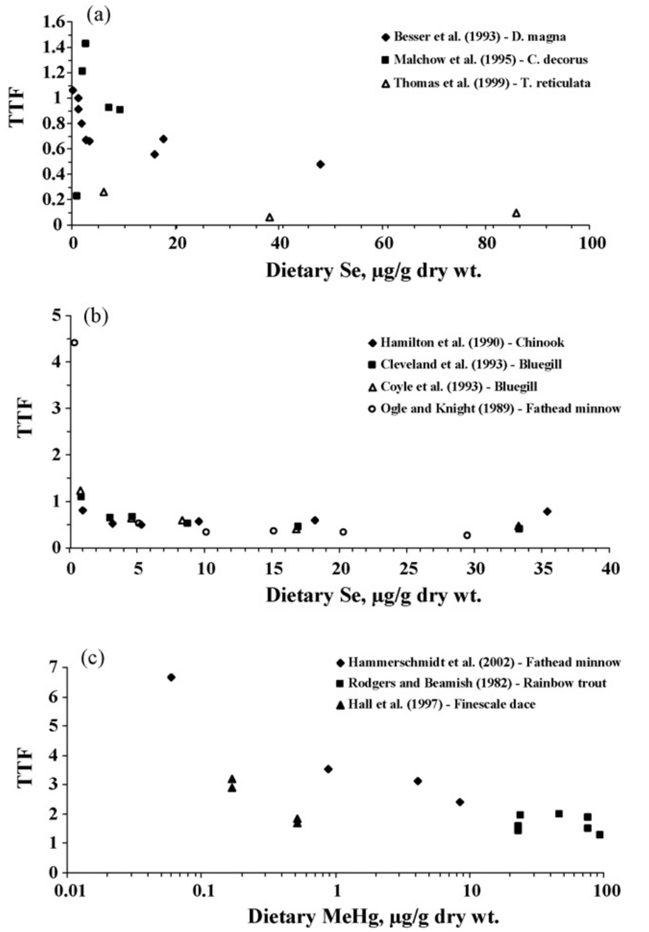

Fig. 4. Whole body selenium TTFs for (a) invertebrates, (b) fish, and methyl mercury (Cleveland et al., 1993; Coyle et al., 1993; Hall et al., 1997; Hamilton et al., 1990; Hammerschmidt et al., 2002; Ogle and Knight, 1989; Rodgers and Beamish, 1982)TTFs for fish (c).

dry wt.). The lower methyl mercury concentrations in the diet compared to selenium and other metals discussed above likely also contributed to the higher TTFs observed for methyl mercury.

4. Discussion

Statistically significant inverse relationships between animal BAFs were observed for a variety of metals in both saltwater and freshwater environments. This is consistent with the analysis conducted by McGeer et al. (2003) using predominantly laboratory-derived BCFs. There are multiple mechanisms potentially responsible for an inverse relationship between accumulation factor and exposure concentration, which are metal- and species-specific. These mechanisms include active regulation (e.g., White and Rainbow, 1982), the influence of natural background concentrations (e.g., Borgmann and Norwood, 1995), and saturable uptake kinetics (e.g., Simkiss and Taylor, 1989) at elevated concentrations. Regardless of the mechanistic basis, the empirical data overwhelmingly support the presence of an inverse relationship for most metals and species evaluated. Those cases where an inverse relationship is not observed may be a function of the data collected (for example, data from a sufficient range of exposure concentrations were not available or aqueous grab samples in field data sets may not fully characterize exposure). For other cases, an inverse relationship may not exist because the organism accumulates metal at a rate proportional to the aqueous exposure concentration due to reduced metal elimination strategies. This would be relevant to organisms that store excess metal with minimal excretion, such as oysters storing copper in granular forms (Brown, 1982). From the perspective of hazard or risk characterization, this latter case may appear to be of particular interest because it tends to result in consistently high accumulation factors over a wide range of exposure concentrations. However, metals stored as granules tend to have reduced bioavailability to consumer organisms (e.g., Nott, 1998; Wallace and Lopez, 1997). Thus, accumulation factors based on such species may provide an indication of total metal present in the organism, but are not directly indicative of a metal's hazard potential unless bioavailability is appropriately considered.

4.1. Comparison of field BAFs and laboratory BCFs

In theory, comparison of the relationships between BAFs and BCFs and their respective exposure concentrations should provide an indication of how dietary metal influences this relationship. Adequate direct comparisons are difficult because BAFs and BCFs are available from separate studies not designed to answer this question and factors such as varying water quality between sites and laboratory waters further complicate comparisons. Due to differences in how aquatic biota regulate/store metals, BAF and BCF comparisons between the same species, or at least within the same taxa, are probably most relevant. This limits the number of comparisons that can be made between BAFs and BCFs. The blue mussel (*Mytilus* sp.) has been extensively evaluated in both field and laboratory studies—BAFs and BCFs for copper, lead, and zinc are provided in Fig. 5. As shown, BAFs and BCFs for both copper and zinc are inversely related to exposure concentration and the regression relationships suggest that BAFs may be higher than BCFs, although the differences are not significantly different if one compares the BCFs and BAFs associated with the same range in water exposure concentrations. For lead, *Mytilus* BAFs are somewhat inversely related to exposure concentration, but the BCF regression slope is near zero. Taken together, this suggests that the whole body lead concentration for this species is increasing in response to lead in the water especially at concentrations greater that 5 µg/L, a level which is much higher than normally found in aquatic environments. Comparisons with other species are not presented here, but our evaluation shows that BAFs tend to be considerably larger than BCFs. A casual review of McGeer et al. (2003), which summarizes data on BCFs for several species, shows these values to typically to be in the range of 100—1000; whereas, BAFs for most species presented here and in our review are typically 1000–500,000. We point out that these higher BAF values do not necessarily point to greater hazard. Frequently, this reflects natural conditions where exposure is low in both diet and water exposures.

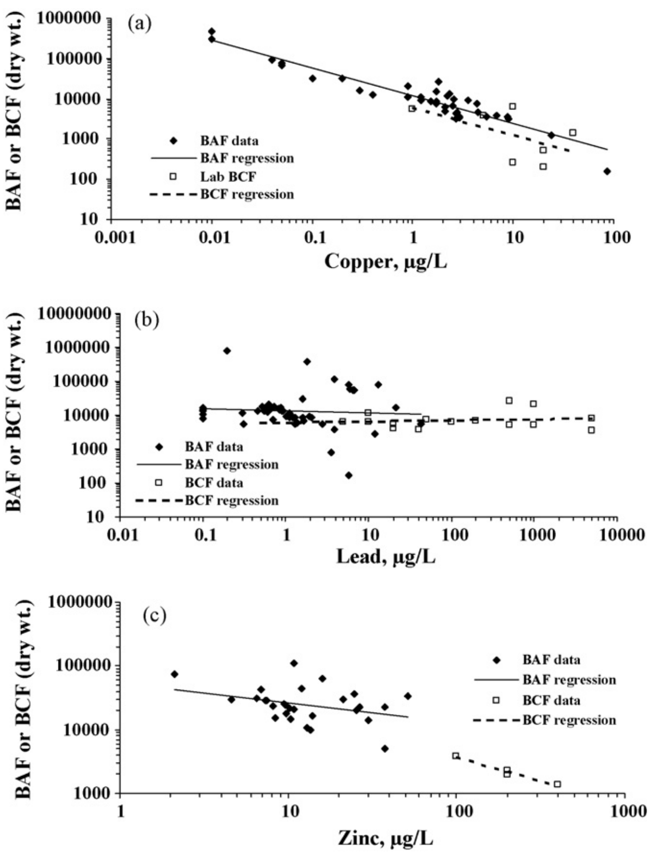

Fig. 5. Comparison of field BAFs and laboratory BCFs for (a) copper, (b) lead, and (c) zinc in blue mussel (*Mytilus* spp.).

4.2. Metal bioaccumulation factors and risk assessment

It is common in environmental assessments to focus on surface water and sediments to characterize exposure, rather than directly measure tissue metal burdens. Most often, this is because compliance with environmental guidelines is normally based on the exposure media (i.e., water or sediment concentrations), but the cost and logistics of tissue sampling can also be an issue. As a result, it is often necessary to relate tissue chemistry to sediment or surface water using accumulation factors. The accumulation factors selected for such an assessment are generally point estimates that do not consider the associated exposure concentration at the site or the exposure conditions from which the accumulation factor was derived. The point estimates of accumulation factors may reflect measures of central tendency of the available data (e.g., mean, median) or upper bound measures (e.g., maximum, 95th percentile).

To evaluate the potential influence of the BAF used to estimate tissue metal concentrations in an environmental assessment, a simple bioaccumulation model was constructed by predicting tissue concentrations using metal concentration in the exposure media and various BAFs. It was assumed that this tissue concentration represented a dietary exposure concentration to aquatic-dependent wildlife, which could then be compared to dietary wildlife thresholds (in this example dietary toxicity thresholds for select metals were identified from Sample et al. (1996) and Heinz et al. (1989)). For this analysis, exposure concentrations were assumed based on chronic USEPA ambient water quality criteria (AWQC) adjusted to a hardness of 100 mg/L (as $CaCO_3$). Chronic criteria were used to represent a site with moderately elevated metal concentrations (i.e., the types of sites typically targeted for environmental assessments). To model bioaccumulation of representative metals from surface water, tissue concentrations were estimated using the minimum, geometric mean and maximum freshwater BAFs for each metal (Eq. (1)), as well as the estimated BAF for the assumed water concentrations using the log-linear inverse relationship data from Fig. 2 (Eq. (2)):

$$\text{estimated tissue conc.} = \text{AWQC} \times \text{BAF} \times 0.001 \qquad (1)$$

$$\text{estimated tissue conc.} = 10^{(m[\log \text{AWQC}]+b)} \times 0.001 \qquad (2)$$

where estimated tissue conc. = µg/g dry wt; AWQC = µg/L; m = slope (based on regression of log BAF versus log water conc.); b = intercept (based on regression of log BAF versus log water conc.).

The estimated tissue concentrations using the BAF from the log-linear regression relationship only exceeded the selected wildlife dietary toxicity threshold for methyl mercury, with the estimate selenium tissue concentration falling just below the wildlife dietary toxicity threshold (Table 3). This is not unreasonable in this generic example as both methyl mercury and selenium have low wildlife toxicity thresholds relative to the BAFs. The ratio of the BAF based on the log-linear regression to the wildlife toxicity threshold further bears this out, for mercury in particular, which has a BAF-to-threshold ratio of approximately 105,000 (Table 3). Site-specific exposures and BAFs become critically important for both methyl mercury and selenium. In contrast, use of the maximum BAF resulted in tissue concentrations exceeding wildlife toxicity thresholds for all metals (except cadmium) based on exposure concentrations equal to the chronic AWQC. These results clearly demonstrate that identification of the most relevant BAF for a given exposure scenario is critical, and not taking concentration dependence into account can lead to overestimation of hazard/risk.

4.3. Metal bioaccumulation factors and hazard assessment

The significance of the inverse relationship between accumulation factors and exposure concentration, and the inability of these factors to be predictive of hazard potential, has been previously addressed by McGeer et al. (2003). Our analysis confirms these same issues apply to BAF data that could be used in hazard and risk assessments. The limited data we evaluated for divalent metals, selenium, and mercury also suggest these exposure levels must be considered when using TTFs to predict metal accumulation across species.

While use of a fixed BAF does not appear to be a viable approach for assessing metal bioaccumulation hazard, we believe using a simple bioaccumulation model such as the one presented above could achieve this objective and allow for assessment of bioaccumulation of metals and metal compounds. From a hazard perspective, the primary concern with bioac-

Table 3
Estimated tissue concentrations based on mean, maximum, and estimated BAFs using the linear inverse regression at a hypothetical site with water concentrations equivalent to their respective chronic AWQC

Metal	Wildlife threshold (mg/kg dry wt.)[a]	Chronic freshwater AWQC[b] (μg/L)	BAF (*n*)	Basis for BAF	BAF (dry wt.)	Estimated tissue concentration at chronic AWQC (mg/kg dry wt.)	Ratio of BAF to wildlife threshold
Cadmium	56	0.25	45	Minimum	500	0.13	9
				geometric mean	11,356	2.8	203
				Maximum	209,000	52	3,732
				Regression[c]	8,284	2.1	148
Copper	97	9	104	Minimum	121	1.1	1
				Geometric mean	11,460	**103**	118
				Maximum	333,333	**3,000**	3,436
				Regression	5,104	46	53
Lead	32	2.5	36	Minimum	131	0.33	4
				Geometric mean	3,321	8.3	104
				Maximum	38,500	**96**	1,203
				Regression	2,954	7.4	92
Mercury (methyl)	125	0.077[d]	43	Minimum[e]	2,727,273	**210**	2,181,818
				Geometric mean[e]	5,751,359	**443**	4,601,088
				Maximum[e]	23,333,333	**1,797**	18,666,667
				Regression[e]	131,317	**10**	105,054
Selenium	6.3	5	451	Minimum	7	0.04	1
				Geometric mean	1,531	**7.7**	243
				Maximum	52,000	**260**	8,254
				Regression	1,187	5.9	188
Zinc	680	120	34	Minimum	5,235	628	8
				Geometric mean	30,323	**3,638**	45
				Maximum	130,000	**15,600**	191
				Regression	4,436	532	7

Bold and underlined estimated tissue concentrations exceed the wildlife dietary toxicity threshold.

[a] Wildlife thresholds are calculated as the geometric mean of dietary NOAEL and LOAEL values reported in Sample et al. (1996) and Heinz et al. (1989) for selenium.

[b] AWQC from USEPA (2006). Criteria for cadmium, copper, lead, and zinc normalized to a total hardness of 100 mg/L.

[c] Estimated BAF assuming site water concentration is equal to chronic AWQC, using log-linear regression.

[d] The USEPA's chronic aquatic life criterion for total mercury is 0.77 μg/L. This value was multiplied by 0.10 to estimate the associated methyl mercury concentration in surface water (USEPA (1997b) noted that the fraction of total mercury present as methyl mercury in surface waters is most commonly about 10%).

[e] Because methyl mercury BAFs are highly dependent on species, age, and size, the BAF statistics shown here are for large piscivorous fish and large suckers.

cumulation of metals/metalloids appears not to be for direct effects to the organism but whether they bioaccumulate to levels harmful to predators. The above analysis demonstrates potential implications of using a selected BAF for wildlife risk assessment and provides a potential solution to this problem. Using such an approach, metals with predicted tissue concentrations associated with water/sediment criteria/guidelines exceeding dietary thresholds for wildlife could be identified as having an increased hazard potential in aquatic systems due to bioaccumulation. To be accurate, such an approach would need to be site-specific.

Based on the above analysis, methyl mercury and selenium (under some exposure conditions) were identified as possibly posing hazard to wildlife via secondary poisoning at water concentrations equal to chronic AWQC, while the divalent metals cadmium, copper, lead, and zinc were predicted to pose no hazard to wildlife at water concentrations equal to AWQC. The results for mercury and selenium are consistent with the scientific literature, in which it has been clearly demonstrated that these chemicals can cause secondary poisoning to wildlife at water concentrations not toxic to prey items (USEPA, 1997a; Maier and Knight, 1994). We reiterate the point that bioaccumulation of both substances is highly site specific. In contrast, we are not aware of any examples for the other metals where effects have been observed on wildlife via exposure from an aquatic food chain, but where no effects are observed on the food chain itself from direct exposure. In other words, to the best of our knowledge, when dietary exposure results in effects on aquatic dependent wildlife, the concentrations are always high enough to cause direct effects on aquatic life as well. Thus, in most cases, hazard and risk assessment of these metals should be driven by their direct toxicity to aquatic biota, not by using BAF as an indicator of potential for secondary poisoning.

5. Conclusions

Overall, the following conclusions were reached from this analysis: (1) the inverse relationship between laboratory-based BCFs and water concentrations is also observed for field-based BAFs, as well as TTFs; (2) the inverse relationship for BAFs suggests that any single BAF considered by itself is not indicative of the hazard posed by that metal because the highest factors are associated with the lowest exposure concentrations (i.e., the least toxic or hazardous); and (3) accounting for the inverse relationship in environmental assessments through the use of bioaccumulation regression models reduces the likelihood that bioaccumulation-based risks are not severely over- or under-estimated.

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