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Technical Issues Affecting the Implementation of US Environmental Protection Agency's Proposed Fish Tissue–Based Aquatic Criterion for Selenium

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ABSTRACT

The US Environmental Protection Agency is developing a national water quality criterion for selenium that is based on concentrations of the element in fish tissue. Although this approach offers advantages over the current water-based regulations, it also presents new challenges with respect to implementation. A comprehensive protocol that answers the "what, where, and when" is essential with the new tissue-based approach in order to ensure proper acquisition of data that apply to the criterion. Dischargers will need to understand selenium transport, cycling, and bioaccumulation in order to effectively monitor for the criterion and, if necessary, develop site-specific standards. This paper discusses 11 key issues that affect the implementation of a tissue-based criterion, ranging from the selection of fish species to the importance of hydrological units in the sampling design. It also outlines a strategy that incorporates both water column and tissue-based approaches. A national generic safety-net water criterion could be combined with a fish tissue–based criterion for site-specific implementation. For the majority of waters nationwide, National Pollution Discharge Elimination System permitting and other activities associated with the Clean Water Act could continue without the increased expense of sampling and interpreting biological materials. Dischargers would do biotic sampling intermittently (not a routine monitoring burden) on fish tissue relative to the fish tissue criterion. Only when the fish tissue criterion is exceeded would a full site-specific analysis including development of intermedia translation factors be necessary.

Keywords: Selenium water quality criteria Federal regulatory guidelines National Pollution Discharge Elimination System permits US Environmental Protection Agency fish tissue criterion

Brief Communication

INTRODUCTION

Selenium occurs naturally in aquatic ecosystems and is a necessary micronutrient in the diet of fish and birds at low (0.1–0.5 μg/g dry weight) concentrations (Gatlin and Wilson 1984; Hodson and Hilton 1983; Klasing 1998). However, at concentrations only 7 to 30 times those required (i.e., >3 μg/g dry weight), selenium becomes a potent poison (Lemly 1993a; Hamilton 2004). A variety of waste materials from industry, agriculture, mining, and petrochemical operations can increase the amount of selenium in aquatic habitats (Lemly 2004). Once waterborne concentrations become elevated, selenium can bioaccumulate in the food chain and reach levels that are toxic to fish and wildlife (Hamilton 2004). Real-world selenium studies show that impacts may be rapid and severe, with teratogenic deformities and reproductive failure affecting entire fish communities and local populations of aquatic birds (Ohlendorf et al. 1988; Skorupa 1998; Lemly 2002a). Few environmental contaminants have the potential to impact aquatic resources on such a broad scale, and even fewer exhibit the complex aquatic cycling pathways and range of toxic effects that are characteristic of selenium (Lemly and Smith 1987; Lemly 2004). Not surprisingly, selenium is a substance of considerable interest to water quality regulators.

The core regulatory guidelines for aquatic selenium pollution in the United States are the Aquatic Life Water Quality Criteria derived by the US Environmental Protection Agency (USEPA) pursuant to the Clean Water Act (CWA) of 1977 (as amended). Because selenium is highly bioaccumulative and its toxicity to fish and birds occurs primarily via dietary exposure, it is the long-term chronic criterion that is virtually always the controlling standard from a risk management perspective. The USEPA last promulgated an updated national chronic criterion for selenium in 1987, some 20 y ago, setting the criterion at 5 μg Se/L on an acid-soluble basis (USEPA 1987). Since that time, serious weaknesses in the national criterion have been revealed. For example, several reviewers of more recent selenium literature suggested that the criterion should be 2 μg/L or less (DuBowy 1989; Peterson and Nebeker 1992; Swift 2002). A key turning point came in 1997 when the USEPA published a proposed set of water quality criteria for aquatic pollutants known as the California Toxics Rule, aka CTR (USEPA 1997). Pursuant to the Endangered Species Act (ESA) of 1973 (as amended), and prior to the USEPA promulgating the CTR, the USEPA was required to consult with the US Fish and Wildlife Service and the National Marine Fisheries Service (Services) and obtain the Services' concurrence that none of the proposed criteria would jeopardize any ESA-listed species. Upon review, the Services found that the 5 μg/L chronic criterion for selenium would likely jeopardize 15 ESA-listed species. To avoid a final "Jeopardy Opinion" from the Services, and the associated legal ramifications, the USEPA agreed to reevaluate their CWA criteria guidance for selenium by 2002 (FWS and NMFS 2000).

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Reevaluating the selenium criteria guidance in the context of an ESA consultation with the Services raised new technical challenges for the USEPA. To address the highly bioaccumulative nature of selenium, and concordant with expert consensus (USEPA 1998; Hamilton 2002; Sappington 2002; Reiley et al. 2003), the USEPA moved away from a water-based chronic criterion and began to develop a fish tissue–based concentration limit. In March 2002, the USEPA completed the draft update document for selenium (USEPA 2002) which was peer reviewed and revised over the course of the next 2 y (USEPA 2004a), and then released in the Federal Register soliciting public comment in December 2004 (USEPA 2004b). One important shortcoming evident in the Federal Register notice as well as the final draft criteria document is a lack of implementation guidance for the proposed chronic criterion. Several of the peer reviews emphasized the complexity of the implementation issues and recommended that the final criteria document include guidance on implementation (Canton et al. 2002). The Federal Register notice and 2004 draft document refer to historical publications that discuss procedures for implementing water-based criteria (i.e., Stephan et al. 1985; USEPA 1987), but do not provide guidance to "fit" the new tissue-based criterion to the real world. This paper bridges that gap by identifying the key factors that will affect the implementation of the USEPA's proposed tissue-based criterion for selenium.

Technical issues

Fish species selection—When selecting species to monitor for regulation of selenium discharges, it is important not only to consider the chemical sensitivity, but also to be mindful of the candidate species' life history aspects, which contribute to their exposure and vulnerability. For example, the type of diet (e.g., detritivore, omnivore, insectivore, piscivore, planktivore) may greatly influence the intake of selenium and thus result in different tissue concentrations among the species available for sampling (Lemly 1985). Species with long life cycles and low reproductive rates are often more vulnerable to increases in mortality than species with short life cycles and high reproductive rates (Matthews 1998; Meyers et al. 1999). The selection of species will thus affect assessment of cumulative impacts from reduced reproduction (i.e., the compounded effect of eliminating potential reproductive individuals from subsequent generations). These characteristics are particularly important when assessing the potential adverse effects of selenium to threatened and endangered aquatic species. For the initial monitoring effort it would be prudent to sample multiple trophic levels and different life stages (juvenile and adult) in order to ensure that the range of tissue selenium concentrations present in the aquatic system is identified. This range-finding would be useful for selecting species and life stages for sampling in subsequent monitoring efforts.

Age of the fish—The USEPA's proposed tissue-based criterion of 7.91 μg/g is founded on the whole-body concentration of selenium in juvenile bluegill associated with winter mortality. The controlling study for the criterion indicated a steep rise in selenium-related mortality following the onset of cold water temperature, and characterized the condition as Winter Stress Syndrome (Lemly 1993b). Cold water temperature caused young bluegill to reduce their food intake sharply and, consequently, their selenium intake. However, loss of lipids and lower body weights created an offsetting rise in selenium concentrations. The result was that a whole-body tissue concentration of selenium approaching 5.8 μg/g—although considerably lower than the proposed criterion value and innocuous in summer—became a grave risk in winter conditions. The USEPA draft document acknowledged the potential for summer selenium concentrations to become toxic in cold weather and recommended different summer and fall screening values of whole-body selenium to act as a trigger point for identifying risks of subsequent mortality. However, the draft document proposes to monitor adult fish as a check on whether exposure during those seasons may exceed the proposed criterion value in winter. Using adult fish is not appropriate for 2 reasons. First, the dietary habits, and therefore exposure to selenium, are very different between the adults and juveniles of many fish species. This means that tissue concentrations in adults will not necessarily reflect those in juveniles. Second, the threat of Winter Stress Syndrome is much greater for young fish. Adults of species such as bluegill continue to feed even in cold weather and do not exhibit lipid depletion and reduced body weight to the same degree as juveniles (Lemly and Esch 1984; Lemly 1996). Selenium-related winter mortality would be expected in juvenile fish but not adult fish (Lemly 1993b). Therefore, for many species the tissue concentrations of selenium in adult fish will not represent responses of juvenile fish to Winter Stress Syndrome.

Survivor bias—When dealing with a mortality endpoint and the sampling of surviving fish, it is difficult to get an accurate measure of tissue selenium due to "survivor-bias" (Seiler et al. 2003), which is a skewing of the random pool of individuals (and thus selenium concentrations) from which to sample by eliminating those who have died. The criterion value would be expected to kill at least 20% of juvenile fish (USEPA's expressed level of acceptable mortality), thereby biasing the pool of surviving fish available for tissue monitoring (i.e., introducing survivor-bias). To address survivor-bias, the USEPA draft document suggested monitoring adult fish tissues because their survival will not be affected by the criterion value concentration (USEPAb 2004b). However, as discussed previously, using adult fish would introduce age-related bias into the risk assessment.

Fishless waters—Implementing a fish tissue–based chronic criterion is problematic for fishless waters. This may seem to be a trivial issue because if there are no fish, why the concern? However, hydrological linkages between fishless waters and other aquatic systems that support fish make them inseparable with respect to selenium transport, bioaccumulation, and exposure (Lemly 1999). Thus, it is essential to apply the criterion to fishless waters in order to assess overall risks to aquatic life. The USEPA suggests the possibility of applying the criterion to invertebrate tissue where invertebrate samples are obtained in place of fish samples (USEPA 2004b). However, in fishless waters invertebrates would not be eaten by fish, but rather, would become food for aquatic-dependent wildlife, especially aquatic birds (Lemly and Smith 1987). Although the intent of the proposed criterion was not to protect wildlife (USEPA 2004b), more work may be needed to determine the effect of proposed selenium thresholds on wildlife that feed on aquatic invertebrates.

Sample locations—In order to accurately assess selenium risks, the locations where samples are to be collected need to be well defined in the context of selenium exposure. For example, selenium discharges can create a delta or zone of highly contaminated sediments and food sources that may result in higher local concentrations in fish than in samples taken from outlying areas (Lemly 1985). Monitoring exclusively in this zone would not yield a representative assessment of tissue concentrations for the entire aquatic system under consideration. Conversely, avoiding these areas would bias the monitoring results in favor of low tissue concentrations. Locations with organic-rich sediments may accumulate selenium to a greater extent than inorganic sediments (Lemly and Smith 1987), resulting in higher food-chain bioaccumulation and exposure of fish in those areas. There can also be substantial differences in exposure between lotic and lentic habitats (Lemly and Smith 1987; Lemly 2002b). The major habitat types, sediment characteristics, and flow conditions must all be considered when the sampling protocol is designed.

Appropriate tissue—The proposed chronic criterion value of 7.91 μg/g selenium on a whole-body fish tissue basis was developed from the USEPA's interpretation of an overwintering survival endpoint (Lemly 1993b). However, reproductive impacts manifested through the selenium accumulated in ovaries and eggs are normally considered to be the most sensitive fish and wildlife biological effects endpoints for selenium (USEPA 2004a). Therefore, it is necessary to evaluate what the proposed criterion would imply for gravid ovaries and eggs of fish. A regression to relate selenium in bluegill ovaries to selenium in bluegill whole-body tissue was presented in the Draft Criteria Document (USEPA 2004a:appendix H) to translate fish exposure data from studies of fish ovaries to a whole-body tissue basis so all species chronic values can be reported as whole-body tissue equivalents. The use of eggs and ovaries may be necessary in situations where winter stress is not pertinent to water bodies, such as in climatologically mild regions or for coldwater species of fish (Moller 2002; Mebane 2005). In these situations, the ovary or egg endpoint will be necessary because the temperature-related stress response and the concomitant loss of body weight and apparent rise in whole-body tissue concentrations of selenium would not be expected to occur. Thus, it is necessary to clearly articulate what tissue is appropriate for monitoring to ensure that the species and community of fish under consideration are being appropriately sampled to identify risks to reproductive endpoints.

Site-specific bioaccumulation factors—A tissue-based criterion will be problematic for the development of a permit limit for new discharges regulated under the National Pollution Discharge Elimination System (NPDES). The USEPA notes that "where translation from the tissue benchmark to a water concentration is needed, a bioaccumulation factor (BAF), which may vary substantially from site to site, would need to be established" (USEPA 2004b). Difficult technical obstacles exist for determining representative BAFs required for site-specific selenium standards. First, it is essential to know the maximum fish tissue selenium concentrations in order to derive a protective water concentration. This necessitates a rigorous, structured sampling program (see sections on averages and minimum datasets). Second, the BAF is not a fixed number that can be applied universally, even to a single body of water. This value is usually dependent upon the concentration of selenium in the water column—sometimes proportional to concentration and sometimes inversely proportional (Lemly 1985, 1997a; McGeer et al. 2003)—and varies with the temporal, spatial, and biogeochemical factors affecting water column and food-chain concentrations (Lemly and Smith 1987; Presser and Ohlendorf 1987). In anticipation of the USEPA's tissue criterion, attempts have been made to develop statistical models that estimate safe water concentrations using bioaccumulation and tissue residue data (Toll et al. 2005). The models did not perform acceptably for lotic habitats (Brix et al. 2005), which is a serious limitation because most NPDES permits are for discharges into lotic waters. Therefore, the BAF issue has not been satisfactorily addressed in the context of the CWA. More effort will be needed to develop accurate, site-specific BAFs that will allow the proposed fish tissue criterion to be translated into acceptable water quality–based limits.

Averages and exceedances—The implementation guidance for the USEPA's current water-based selenium regulations allows the criterion for chronic exposure to be exceeded periodically (once every 3 y, on average) as long as the 4-d average concentration does not exceed the criterion value (USEPA 1987). During exceedances, the permissible ambient (ecosystem-wide) concentration can be up to 4 times the chronic criterion value. This approach, which is based on a generic model for contaminant exposure-response, was rationalized by Stephan et al. (1985) as being the USEPA's best judgment of ecosystem recovery time for certain waterborne pollutants. Conversely, Lemly (1998) pointed out that because of bioaccumulation in aquatic food chains and exposure of fish and wildlife through the diet, averaging periods and excursions above the criterion value should not be allowed for selenium. The USEPA's proposed tissue-based approach will address the flaws associated with water sampling because a tissue measure will effectively integrate waterborne and food-chain exposure. Nevertheless, the flaws pertaining to averages and exceedances may still occur if the basic toxicological premise underlying Lemly's 1998 critique is not accounted for. In the only other instance where the USEPA has developed a fish tissue criterion (mercury), averaging of measurements is permitted (USEPA 2006).

Regardless of whether selenium is measured in water or fish tissue, the numbers used to assess compliance with a criterion or to conduct a risk–hazard–impact assessment must be the maximum concentrations found. In the real world, maximum concentrations are the driving force behind selenium bioaccumulation and toxic effects, not averages. This is an important principle and it is consistent with the toxicity profile for selenium that has emerged from 3 decades of laboratory studies and field case histories of selenium pollution in the United States and elsewhere (Skorupa 1998; Lemly 2002a; Hamilton 2004; Holm et al. 2005; Muscatello et al. 2006). To illustrate the principle consider this hypothetical example: The criterion (toxic threshold) is 200, one-half of the fish sampled contain 300 and the other half contain 50. If simple averaging were used, the result would be 175, which is well within acceptable limits for the criterion, yet one-half of the fish exceed the toxic threshold by 50%. This approach would constitute a fatal flaw, literally, if applied to selenium because exceeding the tissue toxicity threshold by 50% can result in up to 60% teratogenic deformities and mortality (Woock et al. 1987; Cleveland et al. 1993; Coyle et al. 1993; Lemly 1993b, 1993c, 1997a; Holm et al. 2005; Muscatello et al. 2006). Averaging will bias monitoring data by generating a low number and incorrectly suggesting that toxic hazard is lower than it actually is. There should be no provision for spatial or temporal averaging of concentrations, nor averaging among the various fish species that may be sampled. Similarly, there should be no provision to allow tissue concentrations to periodically exceed the criterion value. The concentration–toxicity curve for selenium is very steep, and a small exceedance could cause an exponential increase in death of young fish (Lemly 1997b; Holm et al. 2003). Moreover, the USEPA's criterion value (7.91 μg/g) represents greater than a reduction of 20% in the response observed in controls (EC20) toxicity level (USEPA 2004a). Therefore, finding even a single fish exceeding the criterion value implies substantial impacts and should trigger additional monitoring, particularly if the initial sample size is small.

Dilution or mixing zones—EPA guidance for water-based pollutants designates dilution areas and mixing zones as locations that are exempt from the chronic criterion (Stephan et al. 1985). This approach presents ecological problems when applied in lentic and wetland systems where the "dilution area" may constitute the entire body of open water, making it impossible to reasonably designate a finite mixing zone. In lotic and riverine habitats the mixing zone may vary seasonally, extending for vastly greater distances during dry periods. Because of the tendency of selenium to bioaccumulate in food organisms the mixing zone can become an area of extremely high exposure for fish and wildlife (Lemly 2002b). In these situations, inclusion of dilution and mixing zones would be needed for accuracy when determining site-specific BAFs, developing NPDES permit limits, and evaluating compliance with the chronic criterion. In the only other instance where USEPA has developed a fish tissue criterion (mercury), dischargers may exclude mixing zones if they implement the criterion as a water-based limit calculated using BAFs (USEPA 2006). However, no specific guidance is given on where to sample fish, thus it is possible for a dilution area or mixing zone to be completely excluded from the monitoring protocol.

Hydrological units—It is important to understand the hydrological unit principle and why it should be used to shape the selenium sampling protocol (Lemly 1999). With regard to selenium hazard assessment, a hydrological unit is not based on the US Geological Survey standard of delineation, i.e., the 2,150 cataloging units (Seaber at al. 1987). Rather, a unit consists of the area affected by selenium input sufficient to elevate waterborne selenium concentrations above reference levels typical for the location. This means that a unit extends from source to attenuation or removal, and it may or may not follow well-defined watershed boundaries (Lemly 1999). Individual units may be very small or quite large depending on the concentration of selenium input, biogeochemical cycling, and climate. The hydrological pathways that transport selenium across the landscape, as well as the presence of different habitat types (wetlands, streams, rivers, lakes, impoundments) within many watershed basins have important implications for the sampling regime. Hydrological connections provide a mechanism for selenium discharges to permeate a wide range of habitat types and environmental conditions. These conditions will temper the cycling and biological uptake of selenium (Lemly and Smith 1987). Thus, what may appear to be safe concentrations in water and fish tissue sampled from an area of low bioaccumulation may not accurately reflect what is occurring in nearby, hydrologically connected habitats where bioaccumulation is greater. Failure to include all of the interconnected parts of the hydrological unit in the sampling protocol can result in an incomplete estimate of selenium concentrations and associated risk (Lemly 2002b).

Minimum datasets—Successfully identifying the maximum tissue concentrations of selenium, which is key to environmentally sound risk analysis (Lemly 2002b), depends on taking a sufficient number of samples, but the standard for sufficiency for a given fish species, location, or time depends on a host of factors related to selenium cycling and bioaccumulation as well as demographics of the fish population. These factors confound efforts to prescribe a minimum dataset for broad application, but some initial guidelines can be formulated from existing research and datasets (e.g., Lemly 1985; Schmitt and Brumbaugh 1990; Lemly 1997a; Seiler and Skorupa 2001; Seiler et al. 2003). A reasonable target with respect to monitoring for ecotoxicological applications such as the USEPA criterion would be to capture the upper 95th percentile concentration (Meador 2006). As a general rule, the larger the sample pool the better. It is highly desirable to attain the 95th percentile since the USEPA's criterion is an EC20 rather than a lowest-observed-effect concentration or no-observed-effect concentration (USEPA 2004a). Large datasets will also strengthen the statistical power of model analyses that use BAFs to estimate safe waterborne concentrations (Toll et al. 2005).

Suggested approach

Many of the technical difficulties associated with implementing the new criterion could be avoided if a mixed strategy were employed. A national generic safety-net water criterion of 2 μg/L, as has been recommended (DuBowy 1989; Peterson and Nebeker 1992; Swift 2002), could be combined with the tissue criterion for site-specific implementation. The majority of waters nationwide fall below this safety-net concentration (e.g., Apodaca et al. 2006; USGS 2007), thus NPDES permitting and other CWA activities could continue without increased expense of biotic sampling and translation of those sample results back to a water basis. Dischargers could be required to do biotic sampling intermittently (not a routine monitoring burden) on fish tissue to assess compliance with the criterion. Only when the fish tissue criterion was exceeded would a full site-specific analysis including development of intermedia translation factors be necessary. Exceedance of the water criterion would trigger additional biological monitoring to determine if the tissue criterion was also exceeded (Fig. 1). The tissue-based criterion would be used in the CWA Section 303(d) process to list impaired waters and to develop a Selenium Management Plan (SMP), which could involve using BAFs to derive a water-based concentration limit, establishing total maximum daily loads, and prescribing waste load reduction goals. Other advantages of a mixed strategy are to allow collection of data which may alleviate uncertainties, both with tissue criteria values and difficulties implementing the criteria.

A mixed strategy would have to be developed more fully but we believe the concept has merit, and that the literature contains useful information for the USEPA to draw upon. For example, Lemly (2002b:chapter 7) presents a procedure for deriving site-specific chronic criteria for selenium. The method uses water and tissue concentrations, diagnostic residues, and biological effects to set local criteria for hydrological units. Hamilton (2002) reported that a mixed strategy was being employed for mercury criteria in Australia and

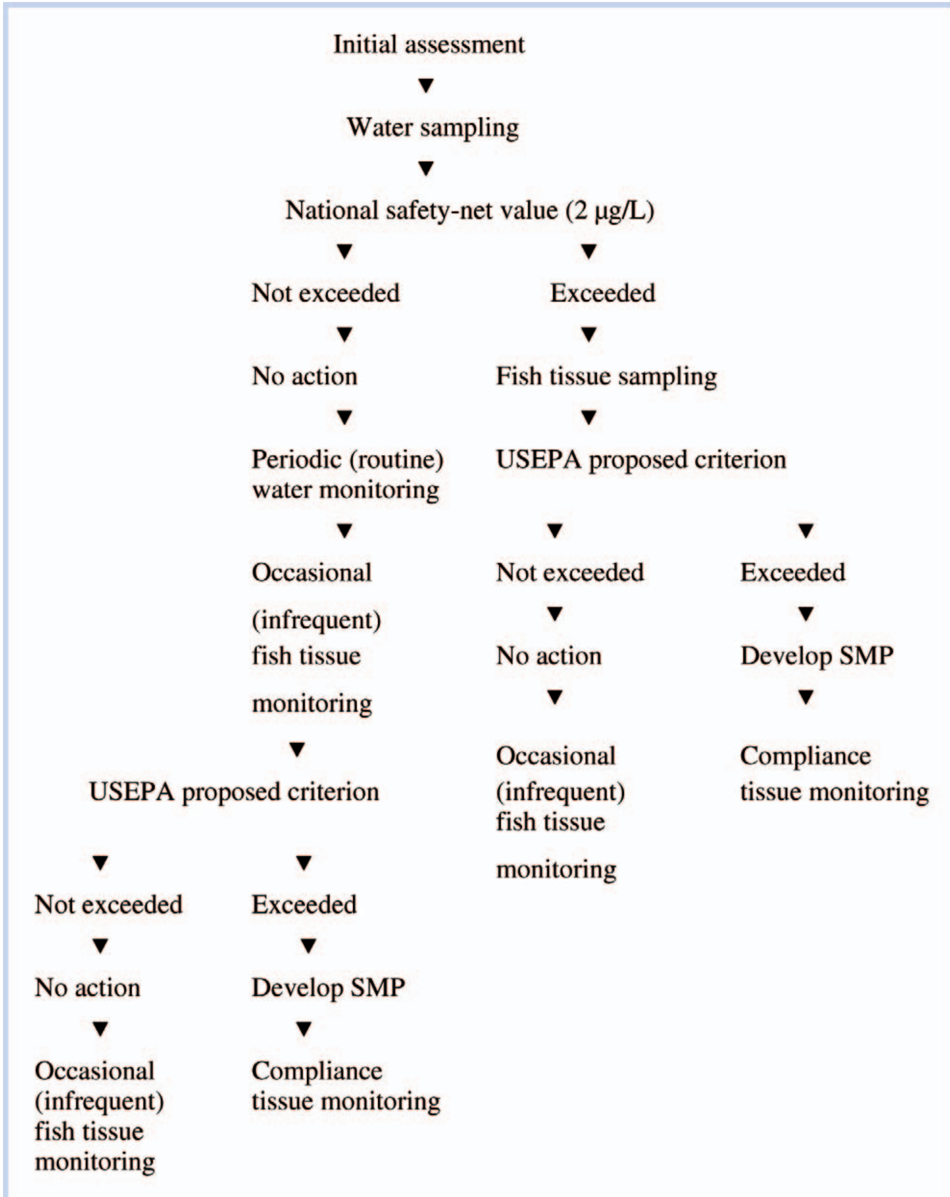

Figure 1. Mixed strategy for implementing a tissue-based aquatic criterion for selenium.

Canada. Because mercury, like selenium, is a highly bioaccumulative pollutant, valuable information may also be gained from the Australian and Canadian experiences. However, we caution against the adaptation of USEPA protocols for implementing fish tissue criteria for mercury (USEPA 2006). This guidance is targeted at protecting public health and would not be transferable to selenium. For example, the fish monitoring protocol recommends compositing samples (which would have the effect of averaging), using skinless fillets in the analysis (which would yield lower values than whole-body measure), and averaging concentrations across locations and trophic levels (which would underestimate toxic hazard). The BAFs are calculated by averaging, and are weighted by human fish consumption parameters, with no intent to ascertain threats to the fish community itself.

CONCLUSIONS

A clear, scientifically sound implementation protocol for the new tissue-based selenium criterion is needed for 3 reasons. First, it would provide an appropriate monitoring design, as the success of the criterion depends on accurate, representative sampling of target populations and receiving waters. Second, the regulated community needs technically correct procedures in order to comply with a more complex monitoring effort than was needed for water-based criteria. For example, in the past a simple grab-sample of ambient waters was sufficient to run a check for compliance with the criterion but now it will also be necessary to sample tissue following a methodology that accounts for several biological and environmental factors. Finally, the protocol would provide crucial technical support for those carrying out provisions of the CWA, such as NPDES permit writers who must have reliable guidance on data collection, modeling, monitoring, and other keys to tracking and controlling selenium discharges. These 3 issues necessitate a comprehensive, detailed guidance document to support the new criterion. In order to facilitate practical implementation, we recommend that the USEPA give serious consideration to a protocol that incorporates both water column and tissue-based approaches.

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