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Remediation and Bioremediation of Selenium-Contaminated Waters

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Abstract

Selenium (Se) is a trace element that occurs naturally in soils, water, biota, and food. It is nutritionally required, but Se in excess is toxic to aquatic-associated wildlife such as fish and birds. Exposure to Se primarily occurs through the diet, not by direct exposure to water; therefore it is vital to account for the "biogeochemistry" of Se—the complex paths by which it moves from contaminated water up the foodweb. As a consequence, waterborne Se is an inappropriate focus for remediation, and this entry illustrates how such a focus can inadvertently misguide major remediation efforts. In fact, there is no fixed "target concentration" for remediation to achieve, as universally accepted water or tissue threshold concentrations for the protection of wildlife are likely to remain controversial for some time to come. This can stymie the traditional remediation strategies of containment, removal, or treatment. One approach to Se remediation that functions under changing regulatory limits is mitigation of wildlife impacts through "alternative" and "compensation" habitats. Also, strategies that take advantage of the natural biogeochemistry, such as algal volatilization of Se, may provide cost-effective management of contaminated water; however, this approach must be combined with foodweb interruption to prevent Se bioaccumulation.

1. General and Historical Background

The trace element selenium (atomic symbol Se) occurs naturally in soils, water, and biota, including food. It is nutritionally required due to various Se-bearing proteins that

incorporate selenocysteine, now recognized as the 21st essential amino acid. In addition to being a required nutrient, Se in excess is toxic to biota (1). The first recorded cases of Se toxicity were penned by Marco Polo during his travels in western China, based on symptoms he observed in livestock (2). In the 1930s, the element Se was attributed to these symptoms, by then known variously as “alkali disease” and “blind staggers,” which afflicted livestock in the western United States (3). However, in certain cases, such symptoms could have been due to other factors (4).

For Se, the margin between nutritional requirement and toxicity is unusually narrow and depends on the individual species and circumstance (3). This fact creates a quandary when setting environmentally protective Se criteria. Health benefits of consuming vegetables high in Se are widely touted (5), as cases of human toxicity and crop damage from Se are rare (6). Livestock face both deficiency and toxicity (7), while toxicity is the primary concern with fish, birds, and in particular various aquatic-associated wildlife (8). The latter issue roared to the news headlines as the aquatic bird disaster at California's Kesterson Reservoir surfaced in the early 1980s (9)—a harbinger of widespread Se problems such as fish mortality and deformities in Belews Lake (10) and numerous other cases internationally (8).

A closer look at the affected biota and circumstances surrounding Se toxicity reveals *water* as the transmitting medium of highest concern (8). Natural water processes (precipitation) and human activities relocate the naturally occurring Se in soils, rocks, and groundwater into collected water bodies that harbor and attract wildlife (8), which might also be used for growing forage and watering livestock (6). Human mobilization of Se mainly consists of irrigation, mine drainage, and surface discharge of groundwater; the latter includes petroleum processing, agriculture, and urban uses. We focus here on the factors important in Se-contaminated water; for the reader interested in the physiology and biochemistry of Se wildlife toxicity, there is an excellent recent review on this complex topic by Spallholz and Hoffman (11).

2. Understanding the Problem: Fundamentals of Selenium Biogeochemistry

In order to solve a problem, it is essential to first understand the nature of the problem. In the case of Se toxicosis of wildlife, exposure to Se primarily occurs through the diet, not by direct exposure to water (12). Therefore, it is vital to understand the “biogeochemistry” of Se: how it moves from contaminated water and sediment to work its way up the foodweb. Figure 1 illustrates the various paths comprising the biogeochemistry of Se. This figure illustrates the first four fundamental facts about wildlife exposure to Se:

1. The paths of Se are numerous and interrelated.
2. Most of the paths eventually lead up the foodweb.
3. Se changes chemical form from an inorganic salt to various organic forms.
4. Some of the organic forms lead to volatilization, a natural path for Se to leave an

aquatic system altogether.

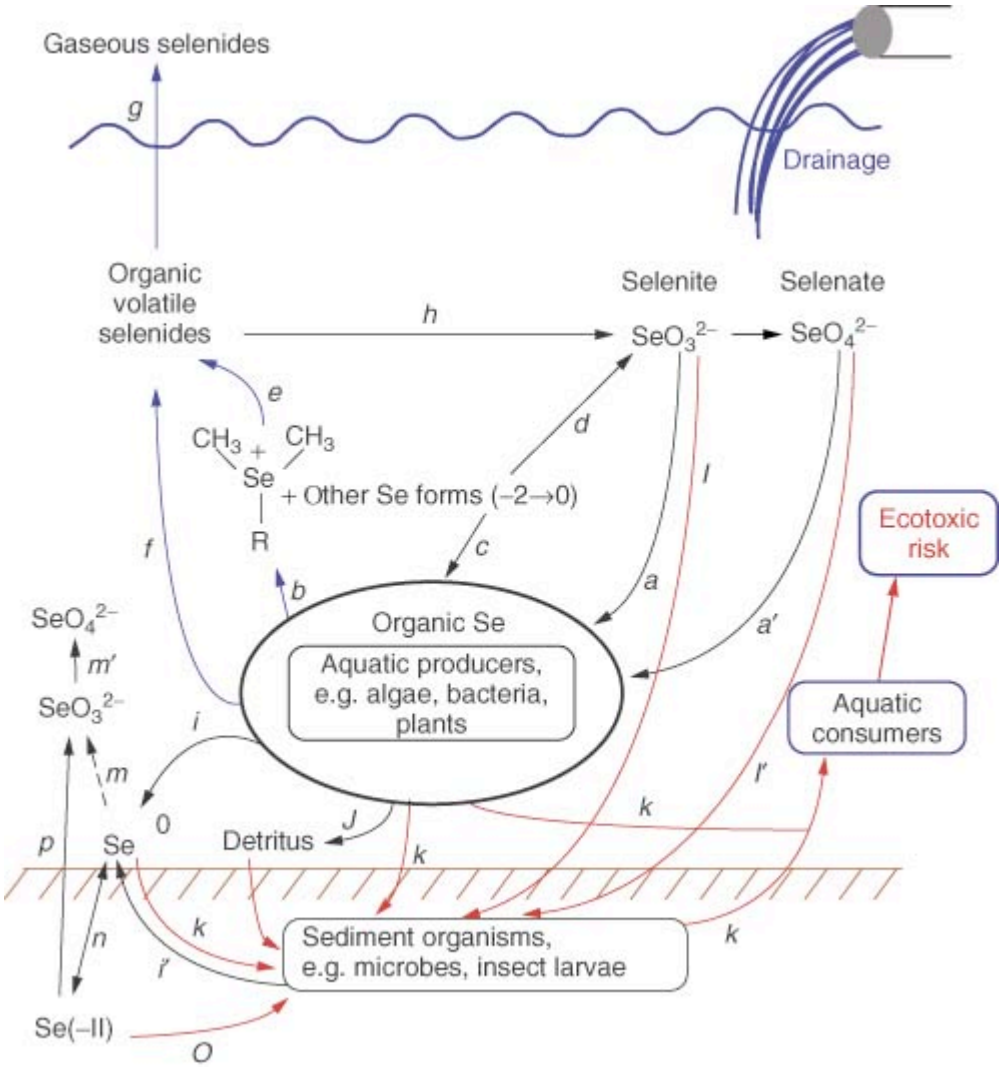


Figure 1. Biogeochemical cycling of Se in aquatic ecosystem. This scheme is modified from Reference 13. Arrows indicate processes that can lead to risk from foodweb accumulation of Se ("ecotoxic" risk). Other arrows trace the Se volatilization process by which Se can be lost from the aquatic system.

a, Uptake and transformation of Se oxyanions by aquatic primary and secondary producers; much of the biotransformation pathway is yet to be defined.

b, Release of selenonium and other organic Se metabolites by aquatic producers.

c, Uptake of organic Se compounds by aquatic producers.

d, Abiotic oxidation of organic Se compounds to Se oxyanions.

e, Release of alkylselenides from selenonium or other alkylated Se precursors through abiotic reaction.

f, Release of alkylselenides from selenonium or other alkylated Se precursors through aquatic producers.

g, Volatilization of alkyselenides into the atmosphere.

h, Oxidation of alkylselenides to Se oxyanions.

i, Formation of red amorphous Se element by aquatic and sediment producers.

j, Detrital formation from aquatic producers.

k, Se bioaccumulation into the foodchain with potential ecotoxic consequences; the toxic form(s) are yet to be defined.

l, Assimilation of waterborne selenium oxyanions into sediment biota.

m, Oxidation of sediment Se(0) to oxyanions.

n, Reduction of sediment Se(0) to Se(–II) or vice versa.

o, Assimilation of sediment Se(–II) into sediment biota.

p, Oxidation of sediment Se(–II) to selenite.

A fifth important fact, not evident in Fig. 1, is that Se "bioaccumulates" as it heads up the foodweb, generally becoming more concentrated in tissues. This concept may be familiar to many readers, as this phenomenon is discussed widely in the general press and health advisories regarding foodweb bioaccumulation of mercury, pesticides, polychlorinated biphenyls (PCBs), and many other contaminants. Table 1 (13) is an example from the scientific literature that documents this process for Se. Note the very large variation in bioaccumulation factors (BCFs), illustrating the effects of site-specific biogeochemistry in different aquatic systems. A comprehensive study illustrating the complex pathways of bioaccumulation has recently been published (14).

Table 1. Excerpted Data from Reference 13 Illustrating Bioaccumulation of Se and the Great Variance in the Bioconcentration Factors of Se

	Salinity, ppth	Waterborne Se, μg/L	Body Burden, μg/g	BCF
Algae	76	13.5	11.8	873
Algae	14	7.9	14.8	1877
Algae	41	7.0	16.0	2281
Algae	136	13.1	9.7	745
Algae	80	13.3	22.6	1695
Algae	10	6.0	1.5	247
Algae	47	5.2	11.7	2273
Algae	66	5.1	9.9	1954
Algae	54	8.7	8.6	978
Average	58.2	8.8	11.8	1436
S.D.	38.1	3.5	5.8	738
Midge larvae	12	5.2	18.1	3489
Benthic composite	76	5.8	7.4	1288
Water column composite	76	5.8	12.3	2130
Corixid	39	7.5	8.9	1187
Corixid	90	8.8	6.8	778
Artemia + corixid	126	11.5	11.7	1012
Artemia	108	12.2	9.0	740
Corixid	108	12.2	10.1	834
Artemia	47	5.6	9.8	1749
Artemia	66	4.9	16.6	3372
Artemia	54	9.5	10.9	1145
Midge larvae	8.3	2.5	42.3	16886
Artemia	67	506.0	19.6	39
Average	67.5	46.0	14.1	2665
S.D.	35.8	138.3	9.4	4389

3. Setting the Remediation Target: The Aquatic Life Criterion

For remediation to occur, there must first be measurable targets to achieve. The complexity of Se biogeochemistry makes setting such a target extremely difficult to accomplish or even define. An earlier United States Environmental Protection Agency (U.S. EPA) Water Quality Criterion for Se, which set a maximum of 5 µg/L total Se, provided a clear target (15). The setting of this temporary value spawned numerous attempts to apply the three traditional strategies of remediation, which are containment, removal, and treatment (16).

However, such a criterion for remediation remains very much a "moving target" due to the complexity of the biogeochemistry. In particular, there are currently attempts to develop a more scientifically sound U.S. EPA Aquatic Life Criterion for Se (17), for which there is now a draft (18). What follows is a very abbreviated description of the issues surrounding the setting of an Aquatic Life Criterion. For a more complete understanding, the reader should refer to the cited literature.

Several independent studies submitted to the U.S. EPA estimated that the 5-µg/L Se toxicity threshold was either too high or too low; interestingly, arguments for higher or lower thresholds appear to be closely associated with whether researchers were affiliated with the corporate-service (arguing that 5 µg/L is too low) or public-service (arguing that 5 µg/L is too high) scientific communities. A slightly different formulation of this observation has also been presented elsewhere (19). Additionally, the proposed California Toxics Rule of 5 µg/L Se in water (20) was judged by the U.S. Fish and Wildlife Service to be too high (21), jeopardizing 15 species protected by the Endangered Species Act (22), the majority of which were aquatic-dependent wildlife that do not actually live *in* the water. The lesson here is that, because of the biogeochemistry of Se, it is important to include consideration of the entire aquatic-based foodweb, not just aquatic organisms.

Recently, the U.S. EPA contracted the Great Lakes Environmental Center to derive chronic Se criteria on a fish-tissue basis, rather than the traditional water concentration basis. The results of this analysis were considered to have limited applicability because it produced a threshold (7.9 µg Se/g tissue) based on an LC_{20} (lethal concentration at which 20% mortality is expected), which is not adequately protective of the species of concern (22). Several other flaws were found in the study, which essentially compiled experimental data from 17 published studies (18). One of these (23) is qualitatively distinct because it incorporated a "winter-stress" design accounting for the increase in toxicity of dietary Se to birds, fish, and mammals under low temperatures. Adjustment of these data for the chronic-level protection needed (<5% mortality) would result in a limit of less than 5.8 µg/g on a whole-body fish-tissue basis.

Situations under which fish tissues would be contaminated to that level, however, may

still be unacceptable to birds eating lower on the trophic level (e.g., invertebrate eaters) (24). Bioconcentration factors relating the transfer of Se from aquatic invertebrates to whole-body tissue of invertebrate-eating fish have been documented in the field to vary from 0.67 to 1.36, when whole-body fish tissue contains 5–10 µg/g Se. Applying those bioconcentration factors to the recently proposed fish-tissue criterion for Se of 7.9 µg/g (18) suggests that the corresponding Se content of aquatic invertebrates would range from 5.8 to 11.8 µg/g, which would result in an EC_{20}–EC_{85} range (effects concentrations, a range for which 20–85% of a population would show toxic effects) in mallard ducks (and other birds) (23) that also utilize the invertebrates as food (24).

4. Strategies for Remediation

From the preceding section, it is evident that there is no universally accepted water or tissue threshold concentrations for the protection of wildlife, and the issues are likely to remain unsettled for some time. Therefore, there is no fixed "target concentration" for remediation to achieve. This section on remediation strategies is written with this situation in mind.

Most of the past remediation attempts are mentioned only briefly here, as they were based on the obsolete 5- µg/L water criterion and do not incorporate the vital concepts of Se biogeochemistry. The following description attempts to discuss Se remediation efforts on the basis of the three strategies of remediation, which are containment, removal, and treatment (16). However, as the reader will see for Se, sometimes none of these three textbook categories are appropriate.

4.1. Containment

The first strategy of remediation, containment, remains difficult to achieve in many cases, as much Se contamination occurs in open lentic systems such as reservoirs, in lotic systems such as streams and rivers, and in receiving waters such as deltas and estuaries. To this end, the U.S. EPA has embarked on regulation based on the total maximum daily load (TMDL) for lotic and receiving waters, under Section 303(d) of the Clean Water Act (25). More recently, the best waterfowl protective estimates (based on empirical data from multiple environments) argue for reduction of the old 5- µg/L limits (8), forcing TMDL limits downward. This is an illustration of the "moving target" frustrating remediation efforts. Clearly, any remediation strategy must be compatible with changing regulations, but despite this clarion call, no traditional remediation strategies have met the challenge. For further description of TMDLs in general, the reader is referred to Reference 25.

The TMDL for Se basically collapses the complexity of Se biogeochemistry into a single box and a single total Se value, which may be protective of a given receiving water. However, this approach does not engender remediation solutions, since it does not account for Se biogeochemistry, provides little guidance for lotic system remediation, and provides no guidance at all for remediation of lentic systems.

In the United States, the lotic Se containment strategy is already implemented under TMDL regulations in California and elsewhere (25). For example, selected agricultural drainage from California's San Joaquin Valley is metered into the San Joaquin River, the allowed volume of which is based on quick turnaround time of Se analysis of the discharged water. For descriptions and issues surrounding the implementation of Se TMDL, the reader is referred further to Reference 25.

Simple containment of lotic Se into terminal water evaporation basins can avoid restrictive TMDL regulations since there is no discharge of water into lotic systems, and this strategy therefore has been suggested as worthy of further investigation (26). However, those authors also recognized that this simply obligates the discharger to use another remediation strategy, since the real problem is foodweb accumulation from either lotic or lentic systems. Therefore, while such total containment might "remediate" the downstream lotic systems, there is still a need to remediate the contaminated lentic system that has been created in the process.

4.2. Removal

The strategy of removal of Se is highly vulnerable to changing threshold regulations, as discussed earlier. Moreover, simple removal of Se is not necessarily "remediation" because the focus is put on waterborne concentrations of Se. In spite of this, several removal strategies have been tested in the past and tests of several more are currently underway. For the few that are in the peer-reviewed published literature, the reader is referred to a recent compilation (27). Removal of Se has been categorically difficult because of its typically low starting concentrations in the parts-per-billion range, its chemical similarity to sulfur (which can be present at more than a millionfold higher concentrations), the very high volumes of water for agricultural drainage, and especially because of the ever-lowering threshold target, as discussed previously.

4.3. Treatment

The third textbook strategy, treatment of Se, has much overlap with removal approaches, and again the focus here has been on reducing waterborne concentrations of Se, which does not equate to remediation (28).

An instructive example of a combined removal–treatment strategy is the algal–bacterial process (29) using the dissimilatory reduction capability of bacteria to reduce selenate into elemental Se, which has been developed to the point of large-scale trials. Although it can remove approximately 80% of the total Se from agricultural drainage water, the microbial and algal action on Se increased the levels of the more bioavailable selenite and organic Se, resulting in 2–4 times greater Se concentrations in the test invertebrates (30).

The lesson here is that waterborne Se is an inappropriate focus for remediation, one of the key reasons for drafting a new U.S. EPA Aquatic Life Criterion for Se based on tissue concentrations (18). The algal–bacterial process is also an example of how TMDL regulations, while written to be protective of wildlife in receiving waters, can inadvertently misguide major remediation efforts because it is based on waterborne Se.

4.4. Management for Mitigation

For Se remediation, there is a fourth strategy, that of management. This strategy is embodied by, for example, a California Waste Discharge Requirement under the Porter-Cologne Water Quality Act, which specified "a program of management actions to reduce, avoid, and mitigate for adverse environmental impacts to wildlife" (26). Management with this goal is a bona fide remediation strategy.

Along this vein, there has been much effort, especially in the western United States, to manage impacts on wildlife using a watershed-based approach, some of which are briefly outlined in Reference 6. In that paper, Engberg and co-workers have stated that "of the five management options presented, mitigation (option 5) may be the only long-term solution to managing selenium." A key tool in such mitigation is the construction of "alternative" and "compensation" habitats to build wetland habitats with clean water for impacted species (6, 26). Note that this remediation strategy does not draw upon any of the textbook "remediation" strategies of containment, removal, or treatment.

The mitigation approach also has its downsides. In the western United States, water supply issues can complicate the Se remediation in water drainage. For example, the cost of land and water is a major obstacle to building and maintaining compensation and alternative habitats. In general, the management approach may require extraordinary effort: it has been stated that "managers must translate the complexities of selenium chemistry and biochemistry into cogent management and regulatory approaches, all the while understanding and blending financial, economic, and social constraints into the solutions" (6). Furthermore, since Se is a semi metal (or metalloid) with a wide variety of forms in many types of samples, it is exceptionally challenging to analyze.

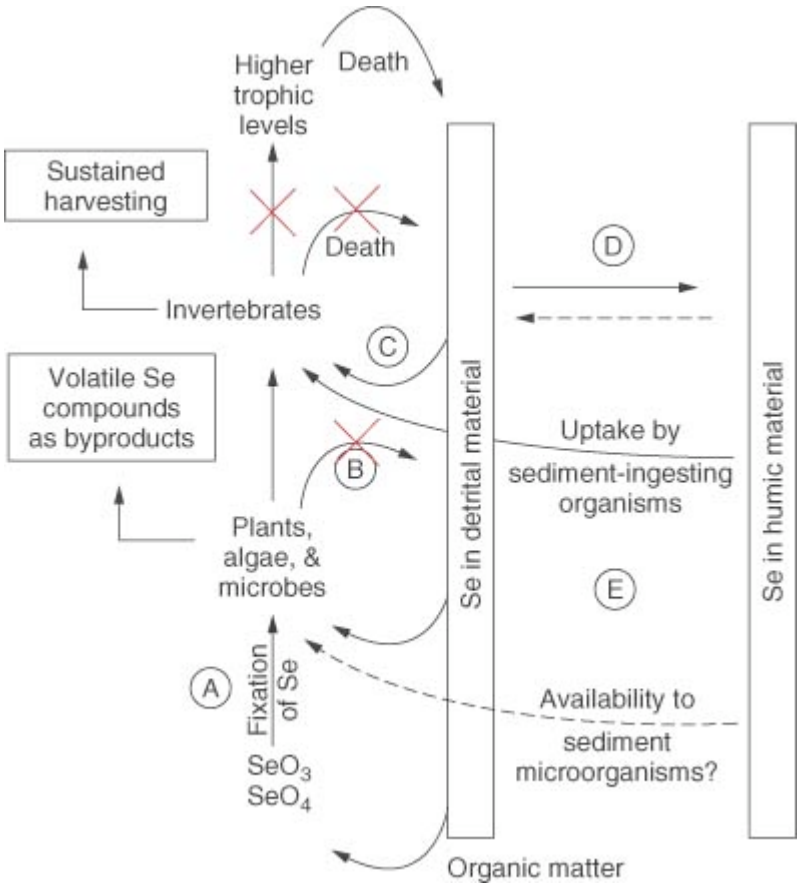
4.5. Natural Bioremediation of Se

As mentioned above, the containment of waterborne Se into terminal water basins eliminates downstream impacts but creates a contaminated lentic system. Use of the mitigation approach is probably needed (26), but combinations of water treatment and management of the terminal basins have also been investigated. Only a few of these take advantage of the natural biogeochemistry to focus on removal of Se from the natural foodweb accumulation. Such an approach differs from the above strategies, which have an underlying assumption that the contaminated system presents only a problem that must be "fixed" somehow (containment, removal, treatment) or bypassed (management–mitigation). Rarely is consideration given that the aquatic system may be mostly functioning "right," and only a few particular processes that are awry need to be addressed.

"Biovolatilization" of Se is one such approach, since it takes advantage of the natural biogeochemistry to remove Se. The problem with biovolatilization of any type, as Fig. 1 outlines, is that the process also draws Se into the biota, and consequently up the foodweb. This tendency has been particularly troublesome in attempts to utilize aquatic vascular plants to volatilize Se. For example, vascular plants volatilize a relatively small amount of Se while sequestering it in bioavailable foodweb materials such as the shoots and roots. Although the shoots could be harvested and disposed, the Se is mostly contained in the below-ground portions of the plants (31), which are not practical for harvesting.

Fan and Higashi (28, 32) have described natural algal Se volatilization as part of an alternative remediation process in terminal basins. The overall remediation process, outlined in Fig. 2, combines volatilization of Se with interrupting the foodweb accumulation of Se (33). In this strategy, photosynthetic algae function to volatilize Se while serving as food to macroinvertebrates (brine shrimp). The brine shrimp graze the algae, ideally preventing algal accumulation and participation in the detrital cycle. In turn, the brine shrimp are harvested as a marketable product, thereby intercepting the foodweb-accumulated Se before it can impact fish and birds. Both volatilization and harvesting of brine shrimp result in a net removal of Se from the aquatic system. The resulting blockages to the foodweb accumulation of Se are shown by the X's in Fig. 2.

Figure 2. Bioremediation in drainage basins by reducing Se ecotoxic risk through invertebrate harvest and Se volatilization. In this "biogeochemical reflux" scheme, the drainage inorganic Se forms are initially biologically fixed by aquatic algae and microbes Ⓐ. The fixed Se does not directly head up the foodchain in the water column, as is often portrayed. Instead, a major fraction enters into organic matter, taking a detour through detritus (recently dead organic matter) and sediment Ⓑ, then reentering the foodchain at several trophic levels Ⓒ. Over longer periods, part of the detrital material is converted to recalcitrant humic material Ⓓ, locking up the Se until sediment-ingesting organisms reintroduce them to the foodchain Ⓔ. Through sustained harvesting (upper left box) of water-column invertebrates that consume algae and microbes, the bioavailable Se is removed from water, plus detrital formation resulting from the death of water-column organisms is also blocked. Both types of blockages are shown by the three X's. In turn, this would help minimize the sediment–detritus foodchain pathway for Se. In the meanwhile, additional Se can be removed by manipulating the algae/microbe community for optimal Se volatilization (lower left box).



A full-scale evaporation basin system has been monitored for over three years in this regard (33). The terminal evaporation basins are brine, which allows the flourishing of brine shrimp that has commercial value—in fact, at California's Tulare Lake Drainage District, there has been successful commercial harvesting of brine shrimp for over five years (34). The efficacy of the approach is clear: (1) the algae that volatilize Se and feed the brine shrimp grow naturally in these basins; (2) the brine shrimp also grow naturally in these basins to a high density; (3) much of the scheme utilizes water management that is familiar to drainage operators; and (4) costs of encouraging and managing brine shrimp growth is offset by marketing harvested materials.

Comparisons with analogous systems where brine shrimp harvesting is not implemented has demonstrated that, under active brine shrimp harvesting (33):

- Waterborne Se concentrations are not increased.
- Se volatilization—a removal process of Se—appears to be enhanced.
- Concentrations of Se in algae and macroinvertebrates—a risk indicator for fish and bird toxicity—are not increased.
- Benthic macroinvertebrate biomass—a key foodweb accumulation indicator—is greatly decreased.

- Total Se as well as organic Se deposited to the sediment (available for biogeochemical refluxing as in Fig. 2) is decreased >90%.

Research is continuing to determine how this strategy can be applied to other systems and to achieve long-term sustainability. At present, the combined strategy of impounding Se-contaminated water in basins, applying the algal volatilization–foodweb interruption process, and constructing compensation/alternative habitats illustrates the advantages of an integrated management approach. In fact, such watershed-scale management appears to be required in order to remediate a contaminant with complex biogeochemistry such as Se.

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