Bioaccumulation of Selenium from Natural Geologic Sources in Western States and Its Potential Consequences

THERESA S. PRESSER*
National Research Program
US Geological Survey
345 Middlefield Road (MS 435)
Menlo Park, California 94025, USA

MARC A. SYLVESTER
National Water Quality Assessment Program
US Geological Survey
345 Middlefield Road (MS 470)
Menlo Park, California 94025, USA

WALTON H. LOW
US Geological Survey, District Office
230 Collins Road
Boise, Idaho 83702, USA

ABSTRACT / Ecological impacts of water-quality problems have developed in the western United States resulting from the disposal of seleniferous agricultural wastewater in wetland areas. Overt effects of selenium toxicosis occurred at five areas where deformities of wild aquatic birds were similar to those first observed at Kesterson National Wildlife Refuge in the west-central San Joaquin Valley of California. These areas are: Tulare Lake Bed Area, California, Middle Green River Basin, Utah, Kendrick Reclamation Project Area, Wyoming, Sun River Basin, Montana, and Stillwater Wildlife Management Area, Nevada. Potential for ecological damage is indicated at six more sites in Oregon, Colorado, the Colorado/Kansas border, and South Dakota out of 16 areas in 11 states where biological tissue data were collected. This conclusion is based on the fact that selenium bioaccumulated in bird livers to median levels that had exceeded or were in the range associated with adverse reproductive effects. Selenium concentrations in samples of fish and bird eggs support these conclusions at a majority of these areas. Reason for concern is also given for the lower Colorado River Valley, although this is not exclusively a conclusion from these reconnaissance data. Biogeochemical conditions and the extent of selenium contamination of water, bottom sediment, and biota from which this assessment was made are given here. In a companion paper, the biogeochemical pathway postulated for selenium contamination to take place from natural geologic sources to aquatic wildlife is defined.

In the San Joaquin Valley of California, seleniferous irrigation drainage water has become a new source of environmental pollution (Presser and Ohlendorf 1987). Waterlogging of soils by intensive irrigation may be remediated by installation of subsurface drains 2.5–3.5 m (8–10 ft) below ground surface. These drains collect what is essentially a soil leachate that contains natural trace elements, including selenium (Se), that have been concentrated by physical weathering in the process of soil formation and evapotranspiration. The enrichment of Se, and some part of the extensive salinization of soil that takes place in the arid climate of the valley, are thought to have their origin in Cretaceous marine sedimentary rocks, which historically have been shown to be seleniferous in the western United States (Trelease and Beath 1949). Current practices of agricultural wastewater management allow the transfer and storage of subsurface drainage to wetland areas. This leads to extended exposure of the ecological community to Se. The wetlands receiving irrigation drainage water, in the course of being used as wildlife habitat, are then operated as evaporation systems to concentrate this agricultural wastewater further, to the form of salts, for eventual disposal. The future means of disposal of the end product is now an issue of concern and controversy in the San Joaquin Valley of California.

First discovered in 1983, high rates of embryonic deformity and death in wild aquatic birds occurred at Kesterson National Wildlife Refuge (NWR), the terminus of the subsurface drains, and was attributed to

KEY WORDS: Selenium; Avian teratogenicity; Bioaccumulation; Environmental contamination; Western United States

*Author to whom correspondence should be addressed.

Se toxicosis (Ohlendorf and others 1986). Selenium was found to be one of the elements that is mobile and able to bioaccumulate in the food chain. Its concentration in the diets of birds and consequently in their eggs, exceeded levels known to cause deformity (Presser and Ohlendorf 1987). In this same study, Se concentrations in livers of birds have been shown to be indicators of toxicity.

Approximately 200 wildlife refuges and management areas receive water from more than 400 US Department of the Interior (USDOI) water projects in the western United States [US Bureau of Reclamation (USBR 1981)]. The majority of these projects consist of agricultural irrigation–drainage facilities constructed by the USBR. In response to concern expressed by the US Congress over Kesterson NWR, the USDOI started a program in 1985 to identify the nature and extent of irrigation-induced water-quality problems that might exist in other parts of the western United States in settings similar to that surrounding Kesterson NWR, i.e., Cretaceous marine sedimentary basins (Sylvester and others 1988, Feltz and others 1990, Harris 1991). Outcrops of Cretaceous marine sedimentary rocks in the western United States comprise a total area of approximately 300,000 square miles from Texas north to Canada, east to Iowa, and west to Utah (Trelease and Beath 1949). However, this survey did not include the coastal states of Washington, Oregon, and California nor the state of Arizona. Twenty-eight areas in 17 states underwent or are to undergo reconnaissance investigations including, in some cases, a search for waterfowl deformities. In lieu of the more time-dependent and time-consuming, field-intensive search for eggs and deformities, concentrations of Se in water, bottom sediment, and biota were used to indicate potential adverse ecological effects and Se in bird livers was used as evidence of toxicity. Historically, only one of the 28 water project sites selected for study had been previously investigated for the presence of Se (Crist 1974). At the Kendrick Reclamation Project in Wyoming in 1974, it was shown that Se movement was accelerated by irrigation, and irrigation drainage water contained up to 1200 μg/liter Se.

In this paper, we identify areas of possible Se contamination in the western United States from reports of the USDOI on reconnaissance investigations of water, bottom sediment, and biota where wetland habitats are receiving agricultural drainage. Most of these areas have similar climatic, geologic, soil, and hydrologic conditions to that of the west-central San Joaquin Valley. These are the first data available on the extent of the Se problem. These data show that bioaccumulation of Se is occurring in most of the wetland areas studied. We also describe the complex chemistry associated with the biological cycle of Se, an element that is both an essential micronutrient in animals and a priority pollutant. Based on the extent of Se contamination found, we propose that there is a fundamental problem in the disposal of agricultural drainage water, particularly to wetland areas, where Se can bioaccumulate to toxic levels in wildlife. This presents a danger to aquatic birds on both the Pacific and Central Flyways. Under federal law, the resulting death of aquatic birds may violate the Migratory Bird Treaty Act, which in recent years has acted as an environmental safeguard (Margolin 1979). The geologic source area and the mobilization and transport mechanisms for the contamination at Kesterson NWR will be presented in detail in the following companion paper. The setting and conditions will be compared to those existing in the USDOI reconnaissance areas.

Entrance of Selenium into the Food Chain and the Chemistry of Uptake

Introduction of Se into the food chain was first demonstrated in Kesterson NWR in 1983 (Presser and Ohlendorf 1987). Selenium in the entrance pond, as soluble selenate, was at a concentration of 350 μg/liter in that year. Selenium in this form is the highest oxidation state, +6. The water was classified as a sodium-sulfate water with a sulfate concentration of 5550 mg/liter. Concentrations of these soluble constituents were expected to progressively increase with evaporative concentration as water flowed from one pond to another, in a series of 12 interconnected ponds. In the terminal evaporation pond at Kesterson NWR, the concentration of Se in water was low (14 μg/liter), even though the sulfate concentration was extremely high (11,500 mg/liter), indicating concentration of inorganic constituents. The Se was found to have been taken into an organic state in an algal mat that covered the almost dry pond. The concentration of Se in the algal mat approached a level of 13 μg/g dry weight, in May 1983 and 24 μg/g in August 1983, a level that is toxic to aquatic birds. Although this sampling was limited, it did demonstrate that selenium lost from solution in the ponds could enter the food chain through uptake by biota and that organic processes were probably more effective in removing Se than the inorganic processes in surface water.

Extensive analyses of organisms consumed by aquatic birds at Kesterson NWR reflected biomagnification of Se in the food chain (Ohlendorf and others 1986, Presser and Ohlendorf 1987). The mean concentrations in filamentous algae, rooted plants, and plankton, all postulated to initiate entrance into the food chain, were 35–85 μg/g Se dry weight. Mean concentrations in higher food-chain organisms, insects and fish, were further enriched at 22–175 μg/g Se, dry weight. These means were approximately 12–130 times those found at Volta Wildlife Management Area (WMA), the control site in the San Joaquin Valley, where Se concentrations for biota were <3 μg/g dry weight. As the area continued to be impacted, 246 μg/g Se dry weight was eventually reached in the algae, 108 μg/g in plankton, 273 μg/g in macrophytes, 293 μg/g in aquatic insects, and 247 μg/g in mosquitofish, the only species of fish remaining in the ponds (Presser and Ohlendorf 1987, Saiki and Lowe 1987). In the wild aquatic birds, the geometric means for livers of adult birds ranged from 20 to 127 μg/g Se and in eggs from 7 to 70 μg/g Se dry weight, depending on species. For comparison, mean concentrations of bird livers (dry weight) from the Volta WMA ranged from 4.4 to 8.8 μg/g Se. Mean concentrations in eggs (dry weight) from Volta WMA were generally <2 μg/g, with no abnormalities found.

The guidelines given for the toxicity of Se do not specify its chemical form. The initial uptake of Se into the food chain involves a reductive incorporation (Cutter 1982), supposedly supported by the rate of development of anaerobic conditions. In the Kesterson NWR ponds, although Se was mainly in the selenate form (+6 Se), up to 30% of the total Se in the latter ponds was in the selenite form (+4 Se). In recent experiments with algae, it was found that organic Se, in the −2 state, with an amino fraction was produced even though the original dosing in the water was with selenite (Se+4) or selenate (Se+6) (USFWS 1990a, Maier and others 1993). This reduced form of Se is presumably as the selenoproteins, selenocysteine and selenomethionine, in which Se is directly substituted for sulfur. This demonstrates that, when introduced into algae, the metabolic fate of accumulated Se is independent of the initial Se species.

From marine studies in the North and South Pacific Oceans (Cutter and Bruland 1984), organic selenide in surface water (<300 m in depth) makes up 80% of the total dissolved Se and the downward flux of particulate Se, found primarily in the −2 oxidation state, decreases with depth. The reduced-state organic selenide maximum, supposedly consisting of selenoamino acids, coincides with the maxima of primary productivity, suggesting entrance into food-chain organisms. This relationship shows that bioaccumulation could be currently taking place in ocean water.

From the freshwater study of Besser and others (1993), selenomethionine at waterborne concentrations of <1 μg/liter have been shown to be bioconcentrated by a factor of 50,000 in algae and 350,000 in daphnids, greatly exceeding those measured with other Se compounds. Selenium concentrations in tissue residues for these food-chain organisms ranged from 5 to 12 μg/g Se dry weight, which is in the range of that which is toxic in the diet of fish and birds (Heinz and others 1987). Similar concentrations of Se were found in biogenic debris from the presentday Atlantic Ocean, which contained from 6.6 (zooplanton fecal pellets) to 8 (surface biogenic particulates) μg/g Se dry weight (Fowler and Knauer 1986).

Biological Cycling of Selenium

The bioaccumulative property of Se may be an essential function in the cellular metabolism of this element. Termed "bioreactive," it shows a nutrient-type distribution in marine environments (Bruland 1983). These bioreactive elements are termed as such, since they are proposed to be mainly involved in biological cycles of the sea (Broecker and Peng 1982). Lipman and Waksman (1923) and Shrift (1964) proposed a biological Se cycle similar to the cycle for sulfur, in which they determined the reductive half and left only a portion of the oxidizing half unsubstantiated. The characteristics that these cyclable elements have in common are that they exist as a gas in at least one stage of transformation and that they undergo a change in oxidation state (Konetzka 1977). Both these conditions are met by Se in biological systems.

Evidence for the reductive half of the cycle has been updated in recent research initiated to elucidate possible bacterial processes in bottom sediment from San Joaquin Valley evaporation ponds. Bacterial mineralization involving the use of inorganic elements as energy sources was found to take place anaerobically in cultures. Selenate (Se+6) was reduced to elemental Se (Se0) by using the selenate as an electron acceptor for bacterial respiration (Oremland and others 1989, Macy and others 1989). The laboratory cultures turned red with precipitation of elemental Se by bacteria thought to be ubiquitous in nature (Oremland and others 1991). The microbes easily reduced amounts of Se at nutrient levels (2860 mg/liter), greater than that occurring in any drainage water. An inorganic process however, has yet to be successfully proven for the reduction of selenate to elemental Se under field conditions. The organic reduction technique is a possible way of sequestering Se in sediments in the San Joaquin Valley. Fungi that aerobically reduce selenate to alkylselenide gas have also recently been cultured (Frankenberger and Karlson 1989), but the conversion rate has not been found to be cost effective as a means of removing Se.

Evidence for the reverse cycle of oxidation of metallic selenides (e.g., CuSe, Se−2) and elemental Se to selenate by microorganisms has also been researched since the experiments of Lipman and Waksman in 1923 (Geering and others 1968, Torma and Habashi 1972).

The oxidation–reduction processes further include a recycling of Se in soluble–insoluble phases similar to those involved in the recycling of sulfur by *Thiobacillus ferrooxidans*. A large population of sulfate-reducing bacteria has been found in ponds used for concentrating metals from solution by introducing microorganisms. There was removal of metals (U, Mo, Se) but no decrease of soluble sulfate in the water flowing through the system (Brierley and Brierley 1981). Reduced forms of sulfur may be oxidized by the aerobic thiobacilli in oxidizing regions, returning the sulfur to the soluble sulfate species. If the case is similar for Se cycles, equilibrium conditions may be attained in pond ecosystems with a rapid turnover time in the biomass, mainly involving the protein form of Se. Including areas such as wetlands in the cycle adds the dimension of higher food-chain bioaccumulation in fish and birds and increased risk of deformities and toxicity.

Reconnaissance Areas

With Kesterson NWR as a prototype, reconnaissance areas in the western United States were generally selected based on six factors, whose significance will be expanded on in the companion paper, being present: (1) a basin of saline marine sedimentary origin that includes soils derived from Cretaceous deposits; (2) oxidized, alkaline soils that promote the formation of selenate, the mobile form of Se; (3) an arid to semiarid climate with evaporation much greater than precipitation leading to salinization of soils; (4) irrigated agriculture served by USDOI-supported irrigation–drainage facilities to leach salts; (5) saline groundwater aquifers resulting mainly from alluvial clay layers that impede downward movement of irrigation water and that cause waterlogging of the crop root zone; and (6) drainage by natural gradient or through buried tile drain networks to USDOI managed migratory-bird refuges, wetland areas, or other areas in receipt of USDOI waters. Names and locations of areas of study by the USDOI are shown in Figure 1. Type (reconnaissance or detailed) and status (potential, underway, or completed) of the studies are also given. Because of early reports of bird deformities at Tulare Lake Bed Area and the expertise of the US Geological Survey in California hydrologic systems, the Tulare Lake Bed Area was studied by the US Geological Survey, not the USDOI. The study, however, conformed to the USDOI protocol and is included here. Data from 15 completed reports (Sorenson and Schwarzbach 1991, Roddy and others 1991, Peterson and others 1988, 1991, Ong and others 1991, Greene and others 1990, Low and Mullins 1990, Setmire and others 1990, Hoffman and others 1990, Stephens and others 1988, Schroeder and others 1988, Radtke and others 1988, Wells and others 1988, Lambing and others 1988, Knapton and others 1987) and data from five other reports that are not yet published (Butler and others 1993a,b, Dileanis and others 1993, Rinella and Schuler 1993, Mueller and others 1993), but where reconnaissance investigations are complete, are discussed in the following sections.

Although these 20 areas underwent reconnaissance as part of the USDOI program, not all biological media (i.e., food-chain items such as plankton and benthos) were sampled at all sites. The following discussion is based on all analytical results for Se in five basic categories in each area: water, bottom sediment, fish, bird livers, and bird eggs. For fish and birds, results for species were combined for each area to allow a comparison among areas.

Of the 20 areas, 13 contain a total of 20 national wildlife refuges (NWR) (Table 1), many of considerable size, up to 750 sq km (288 sq mi) in area. Ten areas are the home or breeding ground of designated endangered species of birds or fish. Before these studies were instigated, declines in or die-offs of birds or fish took place at six areas. Internal drainage basins present at eight sites are of particular concern because they compound the effect of evaporation in the arid to semiarid climate. For example, the Harney Basin is the largest closed basin in Oregon at 13,725 sq km (5300 sq mi). The basin drains to the Malheur NWR, one of the largest inland wetlands in the United States and the largest studied here. The extent of areas in the refuges devoted to evaporation ponds, where concentrating effects take place, is also a factor for consideration. The Tulare Lake Bed Area now contains 2700 ha (6680 acres) of evaporation ponds, whose acreage could be increased fivefold in a planned expansion in the next ten years.

The first factor was not always present at the 20 areas studied. Source rock designations given in Table 2 show that drainage from Cretaceous marine sediments is implicated explicitly in 12 and implicitly

Table 1. National wildlife refuges located in the 20 USDOI study areas

Montana
Benton Lake NWR
Texas
Laguna Atascosa NWR
New Mexico
Bosque del Apache NWR
Utah
Ouray NWR
Wyoming
Bowdoin NWR
Idaho
Lake Walcott and Minidoka NWR
Oregon
Malheur NWR
Nevada
Stillwater NWR
California/Arizona
Havasu NWR,
Cibola NWR
Imperial NWR
California
Lower Klamath NWR
Tule Lake NWR
Salton Sea NWR
Pixley NWR
Kern NWR
Sacramento NWR
Delevan NWR
Colusa NWR
Sutter NWR

Table 2. Source rock designations

Cretaceous marine sedimentary rocks
Pierre shale
Belle Fourche Reclamation Project, South Dakota
Angostura Reclamation Project, South Dakota
Pierre shale and Carlile shale
Middle Arkansas River Basin, Colorado/Kansas
Niobrara, Carlile, Greenhorn, and Belle Fourche shales
Sun River River Basin, Montana
Cody shale
Kendrick Reclamation Project, Wyoming
Riverton Reclamation Project, Wyoming
Mancos shale or equivalent
Middle Green River Basin, Utah
Gunnison River Basin/Grande Valley Project, Colorado
Pine River Area, Colorado
Downstream areas from Colorado River
Salton Sea Area, California
Lower Colorado River Valley, California/Arizona
Downstream areas from Rio Grande River
Middle Rio Grande River Basin, New Mexico
Lower Rio Grande Valley, Texas
Bearpaw shale
Milk River Basin, Montana
Panoche formation and Moreno shale
Sacramento River Complex, California
Tulare Lake Bed Area, California
Permian marine sedimentary rocks
American Falls Reservoir, Idaho
Volcanic rocks
Salton Sea Area, California
Malheur National Wildlife Refuge, Oregon
Stillwater Wildlife Management Area, Nevada
Klamath Basin Refuge Complex, California/Oregon

(downstream areas) in four sites for a total of 16 of the 20 sites studied. Contributions of Se from ash or crystalline volcanic rocks at the other areas are unknown, due to lack of historical data. Other environmental risks considered in the selection of study areas were suspected elevated arsenic concentrations at Malheur NWR and Stillwater WMA and pesticide use in the vicinity of Bosque del Apache, Laguna Atascosa, and Lower Klamath NWRs and the Lower Colorado River Valley.

Methods

Selenium in water samples was analyzed in the National Water Quality Laboratory of the US Geological Survey, Water Resources Division, Arvada, Colorado, using the methods of Fishman and Friedman (1989). Selenium in bottom material was determined in the laboratory of the US Geological Survey, Geologic Division, Lakewood, Colorado, using the methods of Severson and others (1987). Selenium in biota was analyzed in US Fish and Wildlife Service Analytical Control Facility, Laurel, Maryland, or those laboratories under contract to it by the methods described in USFWS (1985). All of the analyses for Se utilized hydride generation atomic absorption spectrophotometry.

Criteria for Selenium Toxicity

Selenium was the constituent of concern most commonly found at elevated concentrations in water, bottom sediment, and biota at the 20 reconnaissance areas (Figure 2). It has been designated as a priority pollutant by the EPA, and it has the greatest potential for toxicological effects in most of the study areas based on comparison to federal regulatory standards and thresholds for ecological damage. Statistical methods used in presentation of the data in Figure 2 are given in Tukey (1977). The data in Figure 2 are referenced to: the US Environmental Protection Agency (USEPA) drinking-water maximum contamination limit (MCL) of 10 μg/liter (USEPA 1988); the USEPA water-quality criteria for protection of freshwater aquatic life of 5 μg/liter (chronic, four-day aver-

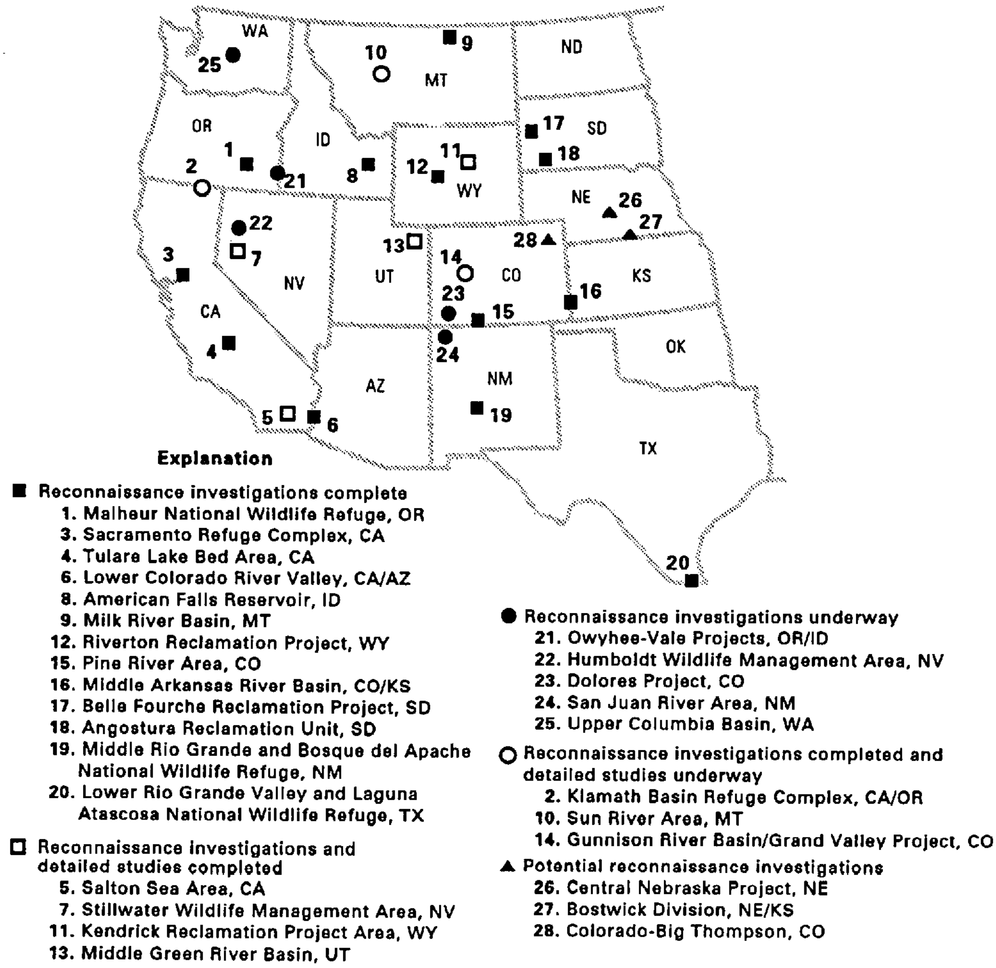

Figure 1. Locations and names of the US Department of the Interior study areas.

age) and 20 μg/liter (acute, 1-h average) (USEPA 1987); the US Geological Survey upper limit of the expected 95% baseline range for soils of the western United States of 1.4 μg/g dry weight (Shacklette and Boerngen 1984); the National Contaminant Biomonitoring Program of the US Fish and Wildlife Service (USFWS) 85th percentile baseline for fish of 2.8 μg/g dry weight (Lowe and others 1985) and the threshold concentration for adverse reproductive effects in fish of 8 μg/g dry weight (Baumann and May 1984); the USFWS concentration range (due to species difference) in livers of birds associated with adverse reproductive effects of 9–41 μg/g dry weight (Heinz and others 1987) and of 30 μg/g dry weight as a threshold for embryo deformity (USFWS 1990b); and the USFWS concentration range in eggs associated with deformities and reduced survival of hatchlings of 10–19 μg/g dry weight (Heinz and others 1987). More recent research has resulted in four further guidelines. A value of <2.3 μg/liter Se is the suggested limit for the protection of aquatic life in water (Skorupa and Ohlendorf 1991). A value of 0.5 μg/g Se for bottom sediments is flagged as an alert to possible contamination (SJVDP 1990), although no standardized sampling procedures or toxic waste standards have been set. The 10/30 guideline (Skorupa and others 1990) suggested for concentrations of Se in bird livers concludes that concentrations <10 μg/g are usually not associated with biological risk and those >30 μg/g usually result in deformity (100% teratogenesis was detected in assessed populations). The same rule is suggested for concentrations of Se in eggs and results in the corresponding 3/20 guideline.

Selenium in Water from the USDOI Reconnaissance Areas

Selenium concentrations varied from site to site in surface collective agricultural drains or streams used

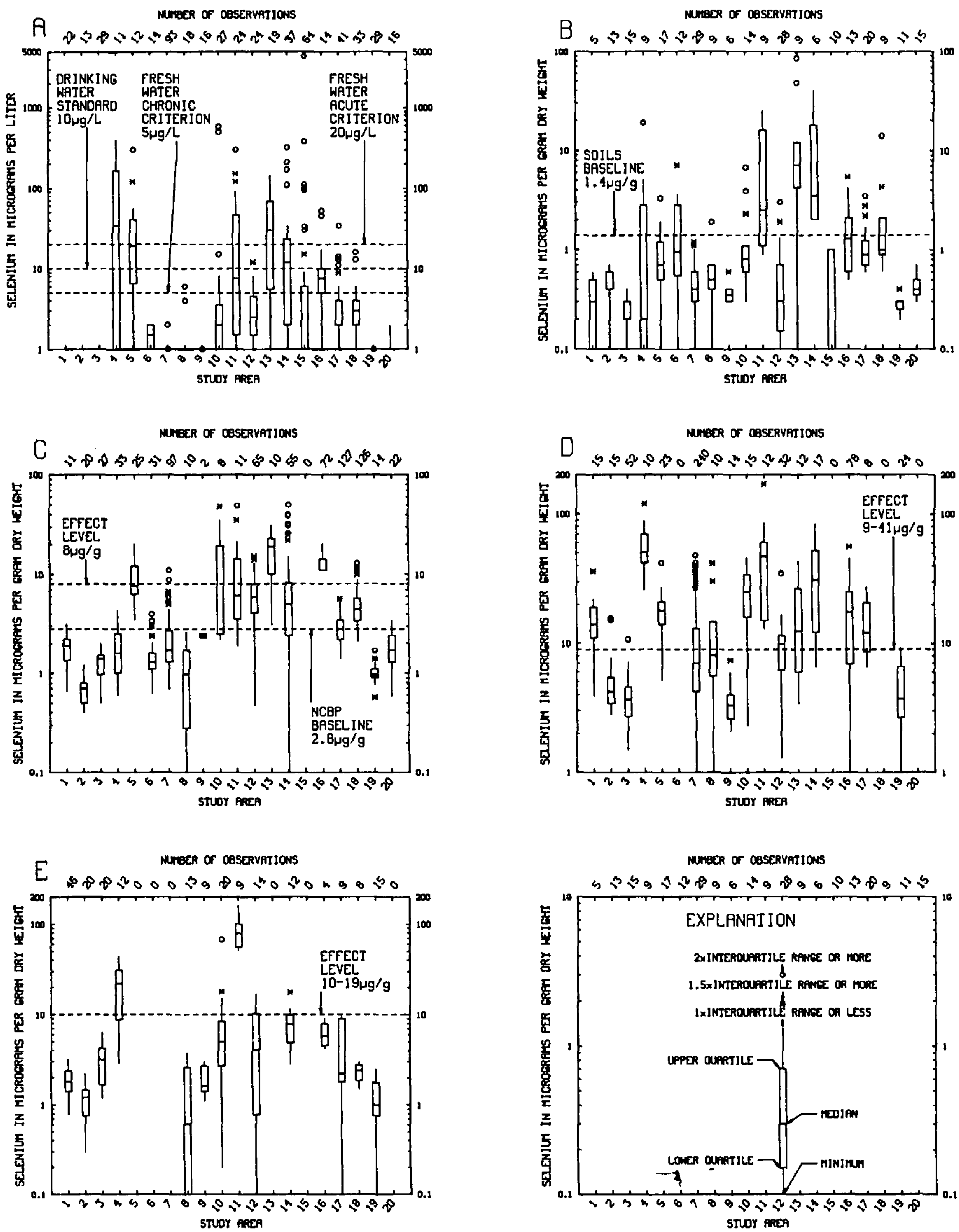

Figure 2. Comparison of selenium concentrations in: **(A)** water (box plots not shown when all observations are less than the reporting limit), **(B)** bottom material, **(C)** fish, **(D)** bird livers, and **(E)** bird eggs. Explanation of statistical representation is given in the lower right corner. Numbers on *x*-axis correspond to Figure 1 names of study areas.

for drainage [e.g., Kendrick Reclamation Project Area (RPA), 27–300 μg/liter; Middle Green River Basin (RB), 4–140 μg/liter; Tulare Lake Bed Area, 36–390 μg/liter; Stillwater Wildlife Management Area (WMA), <1–1 μg/liter; and Salton Sea Area, 7–300 μg/liter]. The two highest Se concentrations observed for the reconnaissance areas studied are 4400 μg/liter in an abandoned well in the Pine RB and 580 μg/liter in a seep in the Sun RB.

Median selenium concentrations in waters, including irrigation supply, drainwater, and refuge-receiving sites such as ponds, to present an overall picture (Figure 2A), are generally low (<10 μg/liter). These levels, however, are not less than the suggested limit for the protection of aquatic life now under consideration by the USFWS (2.3 μg/liter). Two of the areas with high Se levels are the Tulare Lake Bed Area and the Middle Green RB, both of which have median Se concentrations that exceed 20 μg/liter. In addition, median concentrations in the Salton Sea Area and the Gunnison RB/Grand Valley Project exceed 10 μg/liter.

A rather more complicated case shows that time of sampling may be an important factor in assessment of contamination. Assessment may be affected by not only time of year in regards to irrigation but also by a particular year, depending on rainfall amounts or precedence of a drought. Two sites on Beaver Creek in the Milk River Basin contained high Se concentrations in 1985 (Everett Pitt, unpublished data, 1985, Northern Montana College, Havre, Montana). The value for the water at the site upstream of the associated Bowdoin National Wildlife Refuge was 100 μg/liter while the site downstream was 70 μg/liter Se. Although the hydrology of the area is complex, these high values were obtained after four years of drought (1980–1984). In the wetter than normal year that followed (i.e., 1986, two times normal), in which the study reported on here was done, low Se values for the area were obtained. From historical data (Miller and others 1980), water from wells and saline seeps in the Milk River basin, which may be indicative of deeper Cretaceous sources of Se with possible connections to the refuge, showed elevated concentrations of Se (up to 188 μg/liter). The source of this Se will be discussed in the following companion paper.

Selenium in Bottom Sediment from USDOI Reconnaissance Areas

Median concentrations of Se (3.5–7.1 μg/g) in bottom sediments are above the upper limit of the expected 95% baseline range for Se in soils of the western United States (Figure 2B) at three sites: Kendrick RPA, Middle Green RB, and Gunnison RB/Grand Valley Project. Greater than 25% of the values for these sites are above 10 μg/g, with a maximum of 85 μg/g at Gunnison RB/Grand Valley Project. Significant values also occur in other areas. At least 25% of the Se concentrations in bottom sediment exceed the upper baseline range at Tulare Lake Bed, lower Colorado River Valley, Middle Arkansas RB, and Angostura Reclamation Unit. Although the majority of medians for the 20 sites are less than the baseline level, it should be noted that these statistical representations may not have the detail to identify all potential processes of significance and a large variance in sampled material was reported. Limits for bottom sediments above which Se toxicity may be probable (0.5 μg/g) are now being considered and may be much lower, as mentioned before. The organic portion of the bottom sediment, which may include benthos and detritus as parts of the food chain expected to be elevated in Se, may well be the source of the elevated Se concentrations in sediment at the USDOI areas. The importance of this organic material and whether it was included in all samples collected here will be considered later in light of the values for Se obtained for the Kesterson NWR system.

Selenium in Biota from USDOI Reconnaissance Areas

Selenium concentrations in the majority of fish sampled exceed the 85th percentile baseline of the National Contaminant Biomonitoring Program (Figure 2C). At least 25% of the Se concentrations exceed the threshold for adverse reproductive effects in fish in seven of 19 sites where fish samples were collected. They are: Sun RB, Kendrick RPA, Gunnison RB/Grand Valley Project, Salton Sea Area, Riverton RPA, Middle Green RB, and Middle Arkansas RB. Maximum levels of 50 μg/g were seen at the first three of these areas.

Most concentrations of Se in bird livers collected in the study areas are within or greater than the levels associated with adverse reproductive effects in birds (Figure 2D). It is further noted that there are only four areas where nearly all Se concentrations in livers are below the level associated with biological risk. Medians were greater than or in the range of adverse effects at 10 of 16 areas, namely: Tulare Lake Bed Area, Kendrick RPA, Gunnison RB/Grand Valley Project, Sun RB, Malheur NWR, Salton Sea Area, Riverton RPA, Middle Green RB, Middle Arkansas RB, and Belle Fouche RPA. Selenium concentrations in bird livers from the two areas of greatest contamination range from 26 to 120 μg/g dry weight with a median greater than 50 in the Tulare Lake Bed Area, and from 13 to 170 μg/g, with a median of 47 in the Kendrick RPA. The first three areas have median Se concentrations greater than the concentration (30 μg/g Se) at which 100% teratogenicity occurs. Deformed birds were actually observed at greater than background levels of teratogenicity at five sites—Tulare Lake Bed Area, Sun RB, Kendrick RPA, Middle Green RB, and Stillwater WMA. The types of deformities are consistent with Se toxicosis (Presser and Ohlendorf 1987). The evaporation ponds in the Tulare Basin are the ones most comprehensively studied. Deformed aquatic bird embryos from five different species were found at four of the five ponds; the maximum rate of embryonic deformities approached 38% (Skorupa and others 1993). Stillwater WMA, although not having a median concentration in bird livers in the range of adverse effects, has greater than 25% of those values in the effect range (Figure 2D). Deformities also were identified at Stillwater WMA, and "hot spots" were noted as evidenced by a skewed data set at values greater than 30 μg/g for this area. Thus, based on our assessment of bird liver and deformity data, 11 of 16 areas, including Stillwater WMA, are contaminated to a Se level that is associated with adverse reproductive effects. Seven of these areas match those where at least 25% of the Se concentrations in fish exceed the threshold for adverse reproductive effects.

At seven of 14 areas where bird eggs were collected (Figure 2E), medians for Se in eggs are above 3 μg/g, which is generally regarded as the no-risk background limit. These are: Tulare Lake Bed Area, Kendrick RPA, Sun RB, Riverton RPA, Gunnison RB/Grand Valley Project, Middle Arkansas RB, and Sacramento River Complex. Concentrations of Se in at least one egg equal or exceed the range of adverse effects at the first five of these areas, and deformities were observed in the first three of these areas. Concentrations of Se are greatest at the first two of these areas, Tulare Lake Bed Area (range 3–44; median 22 μg/g dry weight) and Kendrick RPA (range 51–160, median 79 μg/g dry weight), where deformities occurred. All of the seven areas, except for the Sacramento Refuge Complex, are in common with those identified as contaminated based on fish and bird liver data.

Complexities of the data from three areas warrant further discussion: (1) Although Stillwater WMA is located in the Carson Sink, recently exploited for containment of drainage and the site of previous extensive bird die-offs, contamination of the area by Se is difficult to understand. Selenium concentrations in water and bed sediment were low. Nearly all Se concentrations in fish were less than those associated with adverse reproductive effects. Yet, reproductive deformities in birds were observed in this area and Se concentrations in livers from many of the birds sampled (Figure 2D) were in the range associated with adverse reproductive effects. A better understanding of food-chain pathways and migratory patterns of birds sampled at Stillwater WMA may help explain this situation. (2) Although concentrations of Se were low at the Bowdoin NWR in the Milk RB in a year of twice-normal rainfall, the refuge overlies Cretaceous shale, and historical data on wells and saline seeps suggests penetration to a Se source that may affect the refuge at times of hydrologic stress. (3) Although not identified as an area of Se contamination in the USDOI reconnaissance studies or in our assessment of data from these studies, the lower Colorado River Valley might be an area of Se concern based on more recent biota and well-water data (Kepner and others 1993). Because of the relevance of data from this area in providing a regional perspective on factors involved in causing Se contamination, additional information from this area is included in the discussion that follows on the Colorado River as a source of Se.

Regional Sources of Selenium

Either Cretaceous marine sedimentary rocks or volcanic rocks are present in the 11 contaminated study areas. The impact of Cretaceous marine sedimentary rocks in the nine areas where this type of rock is a postulated direct or indirect source of Se is the main topic of the following companion paper. Additional comments are given here on their impact on the Colorado River.

If taken in a regional context, the Colorado River may be implicated as a system-wide source of Se contamination due to Cretaceous marine sedimentary rock exposures in its drainage. The river is of high salinity and is influenced by the Cretaceous Mancos Shale (Radtke and others 1988). Half of its salt load is added mainly from erosion of marine sediments and saline springs and approximately one third from irrigation-return flows, all being possible sources of Se. Shallow wells in the upstream reaches of the Colorado and Uncompahgre River Valleys, located in the extensive alluvium and residuum of the Cretaceous Mancos Shale, contained water with Se concentrations of up to 1300 μg/liter (W. G. Wright, written communication, USGS, Grand Junction, Colorado, 1992).

Two of the USDOI reconnaissance study areas associated with the Colorado River are the Salton Sea Area in the Imperial and Coachella Valleys of California and the lower Colorado River Valley located along a 200-mile stretch of river forming the borders of Nevada, Arizona, and California. The Salton Sea Area has been shown to be contaminated with Se based on results in this paper, and results from the lower Colorado River Valley USDOI study (Radtke and others 1988) led to a recommendation of continued monitoring for Se in this area.

The geology of the Salton Sea Area is characterized in Table 2 as complex in nature, receiving a combination of material derived both from Cretaceous marine and volcanic sources. The Imperial and Coachella Valleys and the depression that is the Salton Sea are made up in part by deposition of rocks of Cretaceous age (Norris and Webb 1990). Currently, both valleys are irrigated with Colorado River water, which ultimately drains to the below sea-level, hydrologically closed Salton Sea. During most years, annual evaporation losses approach or sometime exceed a balance with annual inflow, which can concentrate salts.

In the second area, the lower Colorado River Valley, biota data collected in 1988, but not yet published, show contamination associated with the Colorado River (Kepner and others 1993). The USDOI study data were inconclusive, showing whole body carp samples that were greater than baseline levels by approximately a factor of two (mean 6.0 μg/g Se, dry weight) and samples of whole body double-crested cormorants showing a mean of 5.0 μg/g Se dry weight. This latter mean may be significant but is difficult to compare to concentrations in wildfowl liver and egg samples on which toxicity levels are based. The most compelling evidence, however, is from levels of Se bioaccumulated in the Yuma clapper rail, a federally listed endangered species whose main habitat is along the lower Colorado River (Kepner and others 1993). Mean Se concentrations in rail livers (25.3 μg/g dry weight) and eggs (12.5 μg/g dry weight) are comparable to concentrations found in aquatic birds at Kesterson NWR and both exceed the threshold for embryo deformity. Crayfish (4.2 μg/g Se, mean concentration, dry weight), the main component in the diet of rails, are thought to be the source of Se.

The major geologic source of the Se in the lower Colorado River (Kepner and others 1993) appears to be the sediments eroded from Se-bearing Cretaceous formations upstream and deposited in connected backwaters and above dams of the river. Selenium concentrations are greatest in fine-grained bottom sediment containing a large portion of organic matter and not main channel sand deposits. Crayfish feed on this detritus and other benthic species that have already bioconcentrated the contaminants in the detritus. These results emphasize the possible long-term effects of Se cycling in Colorado river water and especially Se loading of sediments.

Importance of Defining Food-Chain Pathways

At specific areas known to be impacted by Se, e.g., Tulare Lake Bed evaporation ponds, concentrations in some food-chain organisms sampled were elevated to levels that exceeded those at Kesterson NWR (water boatman, up to 140 μg/g Se). Because of the reconnaissance nature of the studies, food-chain components were not always collected. Because of this unsystematic collection and the diverse nature of the food-chain materials that were collected, an analysis of these data is not possible here. In retrospect, even though this degree of detail was not required in a reconnaissance study, this was a limitation because these studies indicate that bioaccumulation is occurring, but do not define the food-chain pathways for this accumulation. Notably, the Stillwater WMA, Sun RB, and Riverton RPA have low concentrations of Se in water (generally <3 μg/liter) and bottom sediment (<1.4 μg/g dry weight) but large concentrations of Se in fish and/or bird livers (up to 48 μg/g dry weight). The importance of measuring bioaccumulation should be emphasized because water is not always a good indicator of Se contamination in a biologically active marsh, as seen at Kesterson NWR, where Se has already been taken up in food-chain organisms.

In the reconnaissance studies reported on here, the delivery of the seleniferous waters to the ponds and streams where bioaccumulation is taking place and the degree to which these ecosystems have built up decayed matter may differ. Drainage water began to replace previous input into Kesterson NWR ponds in 1978 and up to a 64% rate of deformity and death in embryos and hatchlings occurred in wild aquatic birds in the 1983 nesting season. The degree of development of organic-rich sediments (i.e., detrital layers), which is known to accelerate the entrance of Se biologically into the detrital food chain, and anaerobic and reducing conditions for geochemically sequestering it in bottom sediments, must be taken into account for determining the potential for Se bioaccumulation. This may well be illustrated by the differing amounts of Se found in the collective drain at Kesterson NWR and the Kesterson NWR ponds themselves. Data from ponds in 1983 showed sediment levels in the range of 5–10 μg/g Se but detritus (decomposing organic matter) in the range of 40–130 μg/g dry weight. Bottom material from the main collective drain at Kesterson NWR, which was described as rich in organics, was as high as 210 μg/g Se. In an attempt to separate out sediment and detritus in this drain in another sample, 92 and 308 μg/g Se, respectively, dry weight, were found. The drain water, although relatively fast flowing, contained a limited food chain and a relatively large amount of soil and debris blown in from the surrounding agricultural fields. Obviously, however, sediment loading of Se was taking place. These high concentrations in bottom material were found normally in the top 2 cm or, in the case of the drain, the 25-cm interval. In the dynamic wetland system of Kesterson NWR, where Se may have been constantly recycled, the food chain and organic debris were the dominant repositories and eventual outlet for Se. Reconnaissance sampling of sediment as an indicator of contamination did not differentiate soil, benthos, or detritus. Thus, significant media, both food-chain items and detritus, in which Se could have been present and was at Kesterson NWR were not fully assessed in the ecosystems studied.

Ecological damage by the same biogeochemical mechanism as reported here for wetlands receiving agricultural drainage may also occur in waterbodies receiving fly ash from the burning of sulfurous coal. A coal of approximately 5 μg/g Se can generate a Se concentration in fly ash (Kaakinen and others 1975) that is sufficient to cause deformities and/or death of fish in receiving reservoirs (Lemly 1985, Cumbie and Van Horn 1978). Bioaccumulation factors of up to 4000 have been observed in such reservoirs. In one reservoir studied, 16 of the original species of fish were eliminated. Studies by Lemly (1993) have shown sublethal effects of Se in fish including mouth, spine, and fin deformities that are analogous to beak, leg, and feet deformities caused by Se in birds. In the USDOI reconnaissance studies, deformities of fish were not looked for nor was the number or diversity monitored. These studies of Se contamination illustrate the importance of understanding the Se cycle from sources such as the weathering of exposed Cretaceous marine sedimentary rocks in the western United States and the burning of sulfurous coals in power plants to food-chain pathways that result in Se toxicity.

Summary

As we have shown, the definition of Se contamination must be formulated on an ecosystem level and these ecosystems have now been assessed for the USDOI reconnaissance areas. Contamination has proven to be prevalent in the western United States where wetland habitats are receiving agricultural drainage. Selenium was seen in detectable amounts in samples of water, sediment, or biota from all 20 reconnaissance areas investigated. Median levels of selenium bioaccumulated in bird livers exceeded or were in the range associated with adverse reproductive effects in ten of 16 reconnaissance study areas where bird liver samples were collected. These areas include seven for which 25% of the Se concentrations exceeded thresholds for adverse reproductive effects in fish and six areas for which medians of Se in eggs were greater than the no-effect level. From these data and the deformities found at five areas, 11 areas of 16 where fish and bird samples were collected have been assessed as being contaminated. With the areas where deformities were found listed first, these areas are: Tulare Lake Bed Area, California; Stillwater WMA, Nevada; Sun RB, Montana; Kendrick RPA, Wyoming; Middle Green RB, Utah; Malheur NWR, Oregon; Salton Sea Area, California; Riverton RPA, Wyoming; Gunnison RB/Grand Valley Project, Colorado; Middle Arkansas RB, Colorado/Kansas; and Belle Fourche RPA, South Dakota. In addition, the Colorado River Basin gives reason for concern in view of its regional contamination.

Concluding Perspectives

This study of USDOI water projects and wetland areas in the western United States is not exhaustive. However, possible predictions based on the many phases of this work may discover or prevent further damage in areas where maintaining high agricultural production and retaining major environmental resources are at issue. This study has documented that agricultural drainage is a source of Se in the western United States and that its introduction into wetlands is a danger to the life of the ecosystem. Selenium in these wetlands, which provide food and habitat to migratory birds, along with resident birds, poses a threat to the health of the populations of aquatic birds on the Pacific and Central Flyways. Death of aquatic birds by Se exposure from such wetlands could therefore lead to possible violations of the Migratory Bird Treaty Act.

In the companion paper, we describe the contamination pathway for the type locality, Kesterson NWR, as an example of the biogeochemical processes probably occurring in most other reconnaissance areas found to be affected. We summarize this "rock-to-duck" cycle and name it the Kesterson effect.

Acknowledgments

We wish to thank the members of the US Geological Survey, US Fish and Wildlife Service, and the US Bureau of Reclamation who participated in the US Department of Interior reconnaissance studies and prepared the reports.

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