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ABSTRACT

Selenium (1) can be beneficial or toxic to plants and animals (including humans) depending on its concentration. It occurs in low crustal abundance in most geological materials but is found in higher concentrations in Cretaceous and early Tertiary age sedimentary rocks, tuffaceous sediments, roll front deposits, and in association with sulfide minerals in metaliferous deposits. In arid regions soils developed from such parent rocks can contain relatively high concentrations of selenium. Higher concentrations of selenium can occur in ecosystems impacted by human actives such as irrigation projects, air pollution, mining, or long term use of soil amendments (e.g. fly ash) containing elevated levels of selenium. Plant uptake and incorporation of selenium into tissue varies widely between species and ecotypes within plant species. Selenium may be essential to some plant species, particularly those that accumulate it in higher concentrations. The element is essential for animals but the range between deficiency and toxicity is relatively narrow. Selenium is frequently deficient in animal diets in higher moisture environments. In arid environments the higher dietary selenium intake from forage and water sources rarely leads to mortalities from acute selenium toxicity. To evaluate the impact of Se on land use several factors should be considered: 1) careful analytical definition of the total and available selenium content of earthen materials on the site; 2) identification of the new depositional environments for these materials; 3) description of post-disturbance planned and potential land uses; 4) an understanding of the components and interactions of the ecosystems.

INTRODUCTION

Selenium (Se) is a unique and interesting element from geological, biological, and natural resource perspectives. It is widely but unevenly distributed in the earth's crust being found in higher concentrations in sedimentary, roll front, and tuffaceous earthen materials. The element is deficient in many animal diets in high moisture environments, rarely concentrated sufficiently in forages to be acutely toxic, and its essentiality in plants remains an issue. The selenium pathway in natural systems provides an important

^(*) Margaret Condron of Parker, CO was a major contributor to this paper and deserves the credit, but none of the blame, for completion of the effort.

⁽¹⁾ Selenium is an element that resembles sulfur and is related to tellurium. It was first described in 1817 by Jons Jacob Berzelius in sulfuric acid residue. Properties include atomic number 34, atomic weight 78.96, natural solid at room temperature, non-metal chalcogen group, relative abundance in solar system is 1.793 log, earth's crustal abundance -1.3 log, density is 4.79, melting point 217 C, boiling point 685 C, minimum oxidation shell and minimum common oxidation number is -2, parallel chain structure, black color, hardness 2 mohs, vigorous reaction with air/heat to form SeO2, no reaction with 6M HCl, found as 6 isotopes, and exists as a selenium hydride SeH2 (Chemicool, 2000; CS.UBC, 2000; and Bayerus, 2000).

classroom or educational example of the biogeochemical relationships that exist within an ecosystems. Se is released from parent rocks or materials through weathering and modified through pedogenesis, Subsequently it is taken up by plants from the soil or other plant growth materials and incorporated into tissue. Plant tissue is then consumed by herbivores utilizing this Se to meet their metabolic requirements Much of the early understanding of Se dates back to work done in arid and semi-arid naturally occurring ecosystems by Beath, Rosenfeld, Gilbert, et. al. at the University of Wyoming, Laramie, and researchers from South Dakota State University from the early 1900's through the 1950's (Beath, 1982). Land disturbed by human activities can produce abnormally high concentrations of Se in water, soils, and plants, and though we have excellent tools for evaluating Se and creating resource inventories and models for planning, many procedural problems remain.

CONFUSED ABOUT SELENIUM?

Studies of Se and its impacts and potential for management often take on the aura of sociopolitical theory instead of well designed and executed natural science research. Tate (1997) suggests that within the realm of "selenium management" one can find a series of conflicts associated with societal needs, soil science realities, and environmental management compromises. Many individuals professing great knowledge and carrying on work related to Se often have vested interests in other issues - politics, regulatory positions and/or advancement, returns from financial investments, funding for research programs, misplaced environmental concerns, etc.

The Food and Drug Administration (FDA) regulates supplementary additions to livestock feeds in the United States and the changes in the standards for Se supplements is an example of the confusion that surrounds Se. In 1974 the FDA approved a dietary supplementation of 0.1 to 0.2 ppm of Se for domestic animals and in 1987 the level was increased to 0.3 ppm for all domesticated animals. A stay order of the 1987 increase was issued in 1993 and levels were reduced to 0.1 ppm for all species except turkeys (0.2 ppm) and growing swine (0.3 ppm). The latter action was taken in spite of overwhelming data supporting supplementation up to 0.5 ppm) in animal feeds and it appears the lower levels were approved in response to problems with Se toxicity at the Kesterson reservoir in California. Additionally, Se is the only essential nutrient not classified as generally regarded as safe (GRAS) by the agency. Confusion becomes widespread on the part of the general public and livestock producers wonder how they are to compete in world markets (Oldfield, 1995a)

Ecosystems are not simple linear systems where cause and effect relationships can be quickly ascertained. A classic example of these difficulties involved in Se research can be found in some of the

early Wyoming work from Chalk Bluff where: Se was thought to have caused large scale livestock toxicities. Beath (1982) attributed a large loss of sheep (approximately 1800 head) on this site to the presence of excessive levels of Se in vegetation. He used the term "blind staggers" to describe the regional livestock losses attributed to the intake of forage containing excessive levels of Se. The Chalk Bluff site has confounding factors such as the presence of locoweeds (Oxytropis lambertii) and high sulfur contents in water and forage. Raisbeck, et. al. (1995) suggested that many of these early Wyoming livestock losses should be attributed to sulfur-induced polio encepihalomalacia, malignant catarrhal fever and/or starvation. Adding to the confusion, however, are the results from more recent plant sampling at the Chalk Bluff site; Se concentrations in plant tissue from Astragalus bisulcatus ranged from 7,000 to > 10,000 ppm and as many as seven or eight other plant species with relatively wide distribution on the site had > 400 ppm throughout much of the spring and summer (Munshower and Prodgers, 1990 and Fisher, unpublished data.) The established upper level of Se concentration for sustained forage consumption by livestock is 5 ppm (National Academy of Sciences, 1976); based on this standard it is hard not to conclude that Se may either have been the cause of the livestock mortality or played a complementary role. Resource managers and administrators frequently want black and white answers, simplistic approaches to these problems, and Se is the antithesis of those desires.

DISTRIBUTION OF SELENIUM

Se does not occur in true deposits and is not directly from the earth. It is generally found in sulfide ores in many metaliferous deposits having significant, often mineable, concentrations of metals. Mineral forms in these ores would include pyrite, clausthalite, naumannite, tienammite and in selenosulfur (EPA, 1998.) When not found associated directly with pyrite, it can be found with silver, copper, lead, and nickel minerals (ATSDR, 1989.) Distribution processes for Se include volcanic activity, combustion of fossil fuels, soil leaching, ground-water transport, metabolic uptake and release by plants and animals, sorption and desorption, chemical or bacterial reduction and oxidation, and mineral formation (Juracek and Ziegler, 1998.)

Most geologic materials contain relatively low concentrations of Se, however, some formations are noted for their higher Se levels - the Cretaceous and older Tertiary sediments in the Great Plains, as an example. Secondary Se deposition in roll front zones, associated with uranium - molybdenum - sulfur, are of significance in some areas and have the potential to create problems in certain mining and mineral extraction operations. Siegel (1979) gave a world wide Se content average of 0.5 ppm for shales and <0.01 ppm for sandstones. Naftz and Rice (1989) reported total Se values of 0.5 to 2.0ppm for Early

Tertiary sandstones associated with coal seams in the Powder River Basin of Wyoming. Se is strongly enriched in coal where it is found as an organoselenium compound, a chelated species, or as an adsorbed element (EPA, 1998). Coal deposits in the form of unmineable stringer beds, and related carbonaceous shales have consistently had elevated levels of Se in the Great Plains and are not considered suitable materials for minesoil construction (Boon and Smith, 1985.)

Anthropogenic activities have a role in the distribution and abundance of Se. The increasing use of fly ash as a soil amendment, with some sources of ash having elevated Se levels, often has implications (positive or negative) for Se content in plants and soils. Similar issues and concerns are found with the land application of activated sewage sludge on agricultural lands in areas located near urban centers. Se compounds are released into the air during the combustion of coal and petroleum fuels and during the smelting and refining of other metals. From 1987 to 1993 (Toxic Release Inventory), Se releases to the land (99%) and water totaled over 1 million pounds. The largest component was from the copper smelting industries with the largest release amounts occurring in Utah. Selenium dioxide (SeO4) is the primary form of Se in industrial settings (EPA, 1998.) Irrigated agriculture has been a focal point for Se concerns in recent years. The economic and societal benefits from irrigation cannot be denied, but the environmental costs associated with this increased productivity, only partially related to Se, have been widespread and significant. The Kesterson irrigation project and related reservoir (National Wildlife Refuge, U. S. Fish and Wildlife Service) developed as an important Se related environmental issue in the early 1980's where it was found to form congenital deformities and mortalities in birds.

Distribution of rocks and soils containing elevated quantities of Se were studied extensively during the 1930 to 1950 period. Lakin and Byers (1941) concluded that soils derived from Cretaceous shales in South Dakota were likely to contain elevated Se content. The exposure of Upper Cretaceous sedimentary rocks in the 17 western states is about 311,000 square miles (Seiler, 1997).

SELENIUM - SAMPLING, SAMPLE PREPARATION, ANALYSIS, AND DATA INTREPRETATION

The literature on Se contains anomalies and contradictions in terms of analytical procedures and interpretation of Se data and studies. Others have categorically rejected any data or papers that were not published within the last ten or fifteen years (as a result much of what is known about Se and its potential role in ecosystems would have to be discarded.) Some authorities believe the analytical methodology and equipment utilized during the early Se studies did not produce the accuracy and reproducibility required of scientific studies. Results from a Se study on the Navajo mine in New Mexico indicated little difference between the fluorometric method for Se analysis and the continuous hydride generation atomic absorption

spectometry (HGAA) procedure used in most commercial analytical laboratories. Palmer (1995) provided comparative Se data between the fluorometric procedure as modified by Koh and Benson and the HGAA procedure and found that the latter provided only slightly higher results.

In studying Se in terrestrial environments, it is often most important to determine the amount of Se that is <u>available for plant uptake</u> from various soil horizons and pedons. Total Se analysis will not provide this type of information. Three extracts are currently, or have been, in general use to evaluate "available selenium" - water, 0.1 M CaC12, and ammonium bicarbonate DTPA. Several factors are important in selecting an extractant: the character of the earthen material being extracted; the availability of an interpretative framework in which to evaluate the resulting analytical data; and the reproducibility of the data obtained from use of the extractant.

The general lack of standardization in soil or earthen material sampling protocol, sample storage, preparation, subsampling for analysis, and type of extract (for available Se) have added to lack of consistency in analytical data and its interpretation. Se volatilizes from soils during drying and the Se forms convert to volatile compounds such as dimethyl selenide, dimethyl diselenide, and others by microorganisms. Similar volatilization of Se compounds in plant tissue occurs and plant tissue sample storage in coolers after collection in the field is essential. Upon return to the laboratory plant samples should be frozen to reduce Se losses. These volatilization losses are a significant issue in terms of generating accurate and reliable analytical data. Lack of quality assurance and quality control programs in Se analytical programs, even in the more current studies, emphasizes the question of data quality and interpretation. Round robin studies including Se as a parameter provide more than adequate support for these conclusions (Severson and Fisher, 1985 & 1987). The Wyoming State Subcommittee on Selenium in Soils, Vegetation, Overburden, and Wildlife and the University of Wyoming have tried to overcome some of these problems through development and publication of documents setting forth standard operating procedures for soil and overburden sampling and analysis (Spackman, et. al., 1994) and vegetation sampling (Steward, 1994.)

SELENIUM AND THE SOIL RESOURCE

Spoils, waste rock and tailings are not the equivalent of soil (2). Many characteristics of the soil

(2) Soil is the collection of natural bodies on the earth's surface, in places modified or even made by man of earthy materials, containing living matter and supporting or being capable of supporting plant out-of-doors (Soil Survey Staff, 1975); Fanning and Fanning (1989) adds comments on humus contents, likening soils to organisms, all loose or unconsolidated material overlying indurated bedrock medium for plant growth, an organized natural body.

system are living, capable of nutrient cycling, and, to a large extent, biological in nature - all factors generally missing in the spoil-like complex of materials. Other key differences include the impacts of weathering and related pedogenic processes The differences between soil and spoil-like materials is striking and pivotal to the understanding of Se and terrestrial ecosystems. To a lesser or greater extent soils are oxidized and in equilibrium with the atmosphere and lower substrates. Fanning and Fanning (1989) identify the presence of oxidative processes as one of the characteristics of soil systems. Although their concept is built on the sulfur cycle, the Se cycle is similar in many respects and relevant.

The oxidized forms of Se, selenate and selenite, are the more biologically active forms and are the Se ion species most involved in Se uptake by higher plants. Se ion species may show some diurnal, seasonal, and annual shifts but they are relatively constant. Spoil materials when exposed to near surface environments and weathering and pedogenic processes will undergo major shifts in Se ion species, surface area and related increase in solubility or reactivity. Extractable Se analysis utilized for the characterization of such materials give only instantaneous evaluations of potential toxicities.

Sampling, characterization, and interpretation of Se characterization data from naturally occurring soils would appear to be a relatively simple matter. As usual with Se, this is not the case. Pedogenic processes, varying in intensity across the landscape, leach some of the soil Se to lower portions of the profile - as with calcium carbonate and calcium sulfate (Munn, 1995 and Fisher, et. al., 1987) Additionally, plant species which incorporate relatively large amounts of Se into their plant tissue continue to recharge the soil system with Se through deposition of and incorporation of plant tissue into soil surface horizons. In soils with well developed profiles - the Argids are an example - it is common to find a bimodal distribution of Se. The first concentration occurring in the upper portion of the argillic horizon with a secondary zone of deposition in the parent material (Munn, 1995). On sites known to have potential Se toxicities, we have encountered many profiles without B horizons or Entisols and the upper zone of Se concentration was not present (Fisher and Munshower, unpublished data and Munshower and Prodgers, 1990). Detailed sampling and characterization data from historically important Se sites in Wyoming indicate the presence of a narrow zone (2 to 6 cm in width) of Se concentration frequently found below the zone of calcium carbonate enrichment in many profiles. The importance of this concentration of extractable Se remains relatively unknown. Standard or routine types of soil descriptions and sampling will often fail to identify such a narrow strata or horizon particularly in the C horizon (parent material), thus missing a potentially important component of the soil Se characterization and evaluation process.

The source of soil Se must be determined on a site specific basis. In natural systems, the source of the Se can be the parent rock where soils are developing in situ, upslope soils and parent rock being a

source where erosional and depositional processes are active, or percolating surface waters or downslope subsurface water movement through surrounding soils, parent material or rock. Se in the spoil-like complex of materials is simpler - it is a product of the Se content in the geologic material from which these materials were extracted. Soil amendments including fertilizers, fly ash, activated sewage sludge that are applied to the surface of soils or incorporated into surface horizons can significantly contribute to Se concentrations in the soil system. Soils found in proximity to coal fired generating units or smelters will reflect elevated Se levels if Se is a component of the airborne discharge from the facility.

The National Academy of Sciences (1983) classified soils and related Se issues into three groups. First, the soils that are the toxic seleniferous soils which are generally alkaline in reaction, well oxidized, and with the Se primarily in the selenate form. The second group of soils are seleniferous but nontoxic because of the limited solubility of the dominant Se ion species (high total Se content but low availability). Such soils are generally acidic, have relatively high concentrations of iron oxides, and are found in Puerto Rico, Hawaii, and Ireland. The third major group of soils would include the majority of soils in North America: soils having low concentrations of Se due to development on parent materials having low concentrations of Se or leaching of Se from the pedon in highly weathered profiles (Nation Research Council, 1976). Regions with lower concentrations of Se in the United States include the southeastern states, the Pacific northwest, and New England, areas with high levels of precipitation, relatively intense soil development and weathering, and exposure of old relatively stable land surfaces.

PLANT UPTAKE AND CONTENT OF SELENIUM

Plant species of the arid and semi-arid rangelands of the Great Plains have been divided into three groups according to their uptake and tissue concentrations of Se. The first and most interesting from the standpoint of Se are the accumulator species (those plants which have the capacity to incorporate several thousand ppm Se into their plant tissues) examples including some species of the <u>Astragalus, Stanleya, Castilleja, Aster, and Haplopappus</u> genera. The second group of plants have the ability to concentrate moderate levels (intermediate accumulators) of the available soil Se into plant tissue, concentrations ranging from 60 to a few hundred ppm Se tissue concentration of Se, and are represented by some species from the genera <u>Agropyron, Atriplex, Gutierrezia</u>, etc. The nonaccumulators are species that have limited accumulation capacity, generally less than 60 ppm, even when growing on potentially seleniferous soils.

Our data suggest that the highest plant tissue Se concentrations occur in the spring and decrease into the fall. The one exception encountered was that of <u>Grindelia squarrosa</u> which apparently increased in Se concentration from the spring into the fall (Fishier, unpublished data). The ranching community in this

region will take advantage of this plant/soil Se character by not allowing stock into pastures containing significant concentrations of accumulator species until much later in the growing season. The practice serves two functions: 1) to allow some decrease in Se content in accumulator plant tissue and 2) to allow full development of the more palatable species providing an alternative to the less palatable accumulator species.

Although there are some conflicting statements in the literature, plant uptake is limited to the selenate and selenite ion species which are dominant in alkaline soil and oxidizing conditions. High soil pH, presence of free lime (calcium carbonate) and salinity are major factors in increasing Se uptake by plants. Selenite is much more readily adsorbed in the soil and therefore much of the plant available Se is in the form of selenate (Mayland, et. al., 1995). Several studies have attempted to correlate either total or extractable soil Se with Se content of plant tissue with only limited success. The wide differences between plant species and their ability to accumulate Se noted earlier is a major limitation. Genetic differences between individual plants within a species and their ability to accumulate Se to be extracted after drying and sample preparation is a major problem in making such correlations. The chemophysical properties of the soil sample have a significant impact on the potential for plant uptake of Se. One of the most difficult factors to overcome is the variability that exists in the soil and its three dimensional nature. Munn (1995) describing a bimodal type of distribution and our work showing a relatively major increase in extractable Se in the soil parent material makes correlation with a single data point/sample to a naturally occurring soil pedon extremely difficult.

HERBIVORES & OMNIVORES AND SELENIUM NUTRITION

Se was the last essential nutrient for animals to be identified. The understanding of its roles in nutrition and health is not as advanced as that of other nutrients. The early work with Se found that in experimental animals it could replace to some extent some of the functions of Vitamin E. Se is an essential component of the enzyme glutathione peroxidase which functions in protection of cells from oxidative deterioration of lipids, proteins, and DNA. In recent years, several other enzymes are known to contain Se, the GPx enzymes, the deiodinase enzymes involved in thyroid hormone metabolism, and another antioxidant enzyme called thioredoxin reductase Se-proteins. Se replaces sulfur in the amino acid cysteine to form selenocysteine which is a component of these enzymes (Combs, 1999).

Se accumulates in animal tissue; in man the value is 1000 times the concentration in surface waters and in marine fish it is 50,000 times the concentration of Se in sea water. The Coastal Plain and the

Pacific Northwest have the lowest Se levels and people in these regions naturally ingest about 60 to 90 meg per day. This compares to a dietary range of 60 to 200 meg and an average of 125 meg for the United States. Se is an antioxidant that has been found to prevent the degradation of fats and cell membranes and block the action of cancer-causing chemicals. Se is naturally found in foods high in protein such as fish, poultry, cereals and other grains as well as in vegetables like mushrooms and asparagus (Wallach, 1999).

White muscle disease results from the lack of adequate amounts of Se in animal diets. The essentiality of Se was established by Klaus Schwarz (National Institute of Health, Bethesda, MD) followed by the identification of white muscle disease from the work of Eggert and Patterson (Oldfield, 1995b). Those minimal dietary levels have been reported to be in the 0.2 to 0.5 ppm Se. Hall (1999) notes that Virginia soils, like most of those in the southeast, are deficient in Se. Hall encourages cattle producers to inject newborn calves with sodium selenite to prevent white muscle disease. Increasing Se supplementation is also recommended for brood cows to enhance immune function and reproductive performance. Enjalbert, et. al, (1999) indicated that supplementing Se in the diets of beef cows in late gestation improves the health of calves.

Blind staggers, alkali disease, and selenosis are all terms that have been used to describe acute and chronic Se toxicity. In forage utilized by livestock over sustained periods of time 5 ppm of Se has been established as a maximum value (National Research Council, 1976) A great many factors influence this value and must be considered in any forage toxicity evaluation. Se was identified as the causative factor in cereal grains which were producing deformities in young animals (Franke, 1934). Raisbeck, et. al.(1995) indicated that the toxicity related to Se is not widespread and is probably due to other conditions entirely. They attribute the "blind staggers" condition described by earlier workers to be sulfur-induced polio encephalomalacia, malignant catarrhal fever, and/or starvation. These workers report that the only form of selenosis that they have been able to document is hair and hoof loss often described as "alkali disease". Anecdotal data suggest that livestock has the capacity to develop higher tolerances to Se either by individuals having constant exposure to forage and water with higher levels of Se or through genetic adaptation in livestock having been exposed for generations to the higher levels of Se in forage and water.

One of the most difficult Se issues is the narrow range in Se content between deficiency and toxicity, about a 100 fold difference. Se deficiency can occur at levels below 0.04 ppm and toxicity at above 4 ppm (Oldfield, 1995). Acute Se toxicity and mortality in livestock remains a relatively obscure event except possibly where stock has been on highly deficient range and are turned into pastures dominated by forage and water having very high Sc; concentrations. Chronic Se toxicity is another matter.

It is by far the most important issue as it may result in the lowering of animal vitality and productivity. In today's agricultural marketplace, any factor reducing animal productivity is of major concern.

EPA has found Se to potentially cause the following human health effects from acute exposures at levels above the MCL - hair and fingernail changes, damage to the peripheral nervous system, and fatigue and irritability. They note that Se has the potential to cause the following health effects from long term exposures at levels above the MCL - hair and fingernail loss, damage to kidney and liver tissue, and the nervous and circulatory systems. Keshan disease in China is an important human disease in the Se deficient areas of China. Se dietary supplements have been widely dispensed to prevent this disease in human populations. Finland require Se fertilization of agricultural lands to supplement both livestock and human populations (Oldfield, 1995a).

DISTURBED LAND RECLAMATION AND SELENIUM ISSUES

Literally millions of dollars have been spent on Se sampling and analytical work as a part of the coal mine permitting process in the Great Plains of the Western United States. A large percentage of these funds have been wasted, or perhaps better said, not well spent. For example, the use of available or extractable type of analysis when characterizing a highly reduced sample of spoil or overburden will yield data of limited value, particularly if this material is to be placed in the near surface zone of the reclaimed landscape. The use of a hot water extractant will produce lower values than ammonium bicarbonate DTPA along with much higher variability in replicate extractions (Soltanpour and Workman, 1980). Water extracts remain in use nearly twenty years after Soltanpour's work was published. Understanding the redox potential of the sample, the general nature of the depositional environment in which the sample material will be placed, and having a thorough understanding of why the analysis is being completed would allow better use of funds. Using either or both extractable or total types of analysis in characterizing a sample can provide important information about Se in the sample for reclamation planning. Quality assurance and quality control programs are generally either weak or not required so industry continues to pay for Se analytical work that can not be properly interpreted. The use of poor quality data can result in overburden quality problems surfacing that need not be raised, or worse, implementation of spurious special handling programs or related added regulatory requirements. In the coal fields, detailed site specific geologic investigations are the first step in developing a stratified Se sampling and analytical program having the capacity to limit the number of samples and allowing for more complete and accurate analysis of the individual sample.

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