Assessment of Risk of Adverse Effects of Cattle Exposure to Selenium on Southwestern Coal Mines<sup>1</sup>

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### Abstract

Consumption of grasses, herbs and shrubs is the primary pathway that may expose herbivores to potential risks associated with selenium (Se) at Southwest surface-coal mines. To assess the potential for adverse effects of exposure of grazing cattle to Se, average concentrations were estimated in biotic and abiotic media and compared to no observed effect levels to determine the potential for adverse effects. Risks were assessed separately for grasses, forbs and shrubs on native soil and regraded spoil areas. Average plant Se concentrations (Se<sub>PLT</sub>) for each plant type were estimated for each of 4 mines. Potential for toxic effects was assessed by comparing *mine-wide* average Se concentrations with a literaturederived no observed adverse effect concentration (NOAEC) of 5000 µg/kg. *Mine-wide* average comparisons was considered appropriate because it is representative of the integrated chronic exposures that cattle would likely encounter. Upper 95% confidence limits were below the NOAEC level for all combinations of plant and soil types. McKinley Mine had the highest average Septr for four-winged saltbush (1530 µg/kg) in regraded spoils. Risk was also assessed at a smaller scale (*mine-areas*) using linear regression to estimate correlations between average  $Se_{PLT}$  and hot-water soluble Se ( $Se_{HW}$ ) in soil materials for samples of specified depth intervals (top 1, 2 and 4 feet). The strongest correlations were found for cool season grasses and shrubs with the  $Se_{HW}$  in the top 4 feet of soil. The upper 95% prediction limit for the mine-area with the largest average Se<sub>HW</sub> was below the NOAEC for all combinations of plant and soil types. Risk of adverse effects due to Se exposure at the mines studied in the SW appears to be minimal or nonexistent at both the mine-wide and the *mine-area* scale.

# Introduction

#### Plant Absorption of Se

- Plant Se (Se<sub>PLT</sub>) was correlated with soil hot-water soluble Se (Se<sub>HW</sub>) in several greenhouse studies (Olson and Moxon, 1939; Soltanpour and Workman, 1980; Jump and Sabey, 1985).
- Similar correlations have been difficult to reproduce under field conditions.
- Soil Se is a poor index of potential toxicity (Trelease and Beath, 1949)
- Se<sub>HW</sub> and Se<sub>PLT</sub> relationships that appear significant in one area are not applicable to other locations (Prodgers, 1991; Sharmasarkar and Vance, 1995) or other species (Fischer et al., 1987) due to varied environmental factors and analytical methods.
- Selenium accumulation varies within the plant (Rosenfeld and Beath, 1964; Williams and Mayland, 1992), both seasonality (Prodgers, 1991) and annually (Johnsson, 1991), and following simulated herbivory (Banuelos and Meek, 1990)

# Introduction

#### Animal Toxicity

- Selenium is both a required nutrient and potentially toxic to grazing animals.
  - Low levels (50 to 3000 µg Se/kg) are required in feed for maximum productivity (CAST, 1994)
  - Wide range (3000-40,000 µg/kg Se<sub>PLT</sub>) proposed as potentially toxic (NRC, 1976; Mayland et al., 1989; James et al., 1989; CAST, 1994; Thacker, 1961)
  - Chronic selenosis is believed to occur after long-term exposure (weeks) when Se<sub>PLT</sub> > 5000 µg/kg (James et al., 1989)
- 5000 µg/kg Se<sub>PLT</sub> considered the no observed adverse effect concentration (NOAEC)
- Recently, questions have been raised whether the early livestock deaths (1856-1930) were a result of selenium poisoning or other misdiagnosed maladies (Geol Survey of WY, 1988; James et al., 1989)
- In the West, blood and tissue Se deficiencies are seen in both wild and domestic herbivores grazing native and reclaimed pastures (O'Toole et al., 1996; Riasbeck et al., 1996; Gov Se Task Force, 1989: Peabody, 1999, BHP, 2000).

# Rationale and Objectives

#### Primary Objective

To assess the potential risk of adverse effects of exposure of grazing cattle to Se at regraded SW surface-coal mines.

#### Risk was assessed in two ways:

Large scale, *mine-wide* risk assessment:

One-mine-at-a-time comparison of upper 95% confidence limits (UCL) for native and reclaim *mine-wide* average Se to the NOAEC.

Small scale, *mine-area* risk assessment:

Regression analysis of *mine-area* paired root-zone Se<sub>HW</sub> and Se<sub>PLT</sub> concentrations (Neter et al., 1996). Se<sub>HW</sub> and Se<sub>PLT</sub> were assumed to be positively correlated, that is, controlling for other factors, the more Se in the soil, the more expected in the plant. In those relationships where the regression is both positive and significant, the NOAEC was then compared to the the  $100^{*}(1-\alpha)$ % upper prediction limit (UPL) of the fitted regression. When the UPL is less than the NOAEC, then the risk of true Se<sub>PLT</sub> exceeding the NOAEC is less than  $\alpha$ . Risk assessment was made at the  $\alpha$ =0.05 level of significance.

#### Secondary Objective

Quantify both  $Se_{HW}$  and  $Se_{PLT}$  at the study's mines.

# Data Assembly

- Plant and soil material Se (total, extractable and hot-water soluble) data were gathered from mine permits, annual reports and other submittals by industry to both federal and state agencies. Most data sources were not electronic and were scanned into a spreadsheet format using translation software. Resulting files were randomly checked for accuracy, formatted for uniformity and collated into separate plant and material databases.
- Se<sub>HW</sub> data were available for 6 SW mines: Black Mesa, La Plata, Lee Ranch, McKinley, Navajo, and San Juan. Se<sub>PLT</sub> data was available for Black Mesa, McKinley, Navajo and San Juan, sampled from multiple mine-areas and accompanied by soil Se data. Midpoint depth of a soil sample increment was used to assign each sample a root zone depth. Both *mine-wide* and *mine-area* Se<sub>HW</sub> averages for the top 1, 2, and 4 ft of soil were calculated.
- Species-specific Se<sub>PLT</sub> data were averaged by life-form into 4 categories: grasses, forbs, herbs, and shrubs for both *mine-wide* and *mine-area* levels. Grasses were also split into both cool and warm season categories. Forbs included all herbaceous non-grasses. Herbs included all herbaceous vegetation (grass and forbs). Individual species data was also available for western wheatgrass (*Elymus smithii*) and 4-wing saltbush (*Atriplex canescens*).

## **Resulting Dataset**

#### Soil Materials: 12890 total soil, spoil and topdressing samples

> 3345 profiles sampled from 6 mines, totaling 12890 samples from spoil, topdressing and native soil from 3228 reclaim and 117 native sites collected between Oct '85 and July '97

#### Plants: 1216 total plant samples

52 plant species from 4 mines at 492 sampling locations collected between Oct '85 and July '97

# Non-detects and outliers

#### Censored data

- Assuming the true value of a non-detects to be uniformly distributed between zero and reporting limit of a mine's dataset, censored data were replaced with randomly generated values (Helsel and Hirsch, 1992)
  - 27.2% of plant data modeled
  - 33.3% of material data modeled

#### Accuracy

 When mine datasets for soil materials included total selenium (Se<sub>TOT</sub>) and/or extractable selenium (Se<sub>EXT</sub>), data was excluded when:

$$Se_{TOT} < Se_{HW}$$
 and/or  $Se_{EXT}$ 

• 182 samples removed from analysis (1.41%)

# Data Analyses

#### Mine-Wide average Se concentration

- Each Se<sub>PLT</sub> data point is the average Se over all areas of the same soil type (native or reclaim) at a mine
- Each material Se<sub>HW</sub> data point is the average Se over all areas of the same soil type (native or reclaim) at a mine at 1, 2, or 4 ft depth interval
- Risk assessment made one mine at a time

#### Mine-Area average Se concentration

- Raw data was reduced to the subset of sampled mine areas that had both Se<sub>PLT</sub> and Se<sub>HW</sub> data
- Native and reclaim Se data were averaged by mine-area for plants and soil materials
- > 38 areas (11 native, 27 reclaim) with 489 plant plots and 2075 soil samples from 603 profiles
- Sampling dates did not always coincide for plant and soil pairs
- Correlation between mean Se<sub>PLT</sub> and mean Se<sub>HW</sub> values were estimated by linear regression
- Risk assessment made simultaneously for all mines

## Mine-wide Results

#### ✤ Fig 1: Average Se<sub>HW</sub> by mine and depth

Spoil materials have higher Se<sub>HW</sub> than native soils

# Fig 2: Average Se<sub>PLT</sub> by mine and vegetation type

- Shrubs have the highest Se<sub>PLT</sub>
- Shrubs in reclamation have slightly higher Se, but Se for shrubs in both native soil and reclamation is far below the NOAEC level
- This suggests that *mine-wide* differences between native and reclaim have no biological significance with respect to livestock grazing.

# Figure 1: Material Se

Mean  $Se_{HW}$  in native and reclaim soil materials at SW mines by depth. Error bars represent the 95% UCL of the mean







# Figure 2: Plant Se

Mean  $Se_{PLT}$  for vegetation rooted in native and reclaim materials at SW mines. Error bars represent 95% UCL of the mean



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## Mine-Area Results

#### Table 1: Regression between Se<sub>PLT</sub> and Se<sub>HW</sub> paired by *mine-area* for 1, 2, and 4 ft depth intervals

- > Highly significant at p<0.05</p>
  - Cool season grasses (top 4 feet, |t|=2.55, p=0.02)
  - Shrubs (top 4 feet, |t|=2.09, p=0.05)
- Moderately significant at p<0.15</p>
  - 4-wing saltbush (top 4 feet, |t|=1.58, p=0.13)
  - Grasses (top 4 feet, |t|=1.48, p=0.15)

#### Figures 3 and 4: Regression plots for cool season grasses and shrubs.

- For both, the most significant depth (4 feet) is plotted.
- In both, the 95% UPL is below NOAEC, indicating a low probability (< 0.05) that even the largest observed average area Se<sub>HW</sub> would have a corresponding average Se<sub>PLT</sub> greater than the NOAEC.

# Table 1: Regressions for *Mine-area* data

Regression of *mine-area*  $Se_{PLT}$  (µg/kg dry wt) vs  $Se_{HW}$  (µg/kg) in the top 1, 2, and 4 ft of soil material

-	Depth	Intercept					Slope UPL for				
Variate	(ft)	n	Intercept	se	Т	Р	Slope	se	Т	Ρ	max soil Se
Grasses	1	33	227.2	39.3	5.78	<0.01	0.6	0.5	1.1	0.3	
	2	35	214.9	38.1	5.65	<0.01	0.7	0.5	1.3	0.2	
	4	35	209.4	38.7	5.4	<0.01	0.7	0.5	1.5	0.2	
Herbs	1	33	254.0	39.6	6.4	<0.01	0.4	0.6	0.6	0.5	
	2	35	239.7	38.8	6.2	<0.01	0.5	0.5	0.9	0.4	
	4	35	236.0	39.7	6.0	<0.01	0.5	0.5	1.0	0.4	
Shrubs	1	31	825.3	155.2	5.3	<0.01	2.7	2.7	1.0	0.3	
	2	33	0.7	0.2	4.8	<0.01	4.3	2.4	1.8	0.1	
	4	33	669.0	150.3	4.5	<0.01	4.9	2.4	2.1	0.1	2366
Forbs	1	26	366.1	97.8	3.7	<0.01	0.0	1.6	0.0	1.0	
	2	27	340.1	97.6	3.5	<0.01	0.3	1.4	0.2	0.9	
	4	27	327.0	99.1	3.3	<0.01	0.5	1.4	0.4	0.7	
Cool Season Grasses	1	29	101.5	21.9	4.6	<0.01	0.6	0.3	2.0	0.1	
	2	31	96.0	20.4	4.7	<0.01	0.6	0.3	2.3	0.0	
	4	31	92.3	20.3	4.5	<0.01	0.7	0.3	2.6	0.0	342
Warm Season Grasses	; 1	22	350.2	76.0	4.6	<0.01	-1.0	1.1	-0.9	0.4	
	2	23	347.3	76.2	4.6	<0.01	-0.9	1.1	-0.9	0.4	
	4	23	349.1	78.5	4.5	<0.01	-1.0	1.2	-0.9	0.4	
Western Wheatgrass	1	25	338.2	82.0	4.12	<0.01	0.0	1.1	0.0	>0.9	
	2	27	333.6	77.7	4.29	<0.01	0.1	1.0	0.1	0.9	
	4	27	316.4	78.3	4.04	<0.01	0.4	1.0	0.4	0.7	
Four-Wing Saltbush	1	28	881.6	195.9	4.5	<0.01	2.7	3.7	0.7	0.5	
	2	29	776.1	192.9	4.02	<0.01	4.4	3.3	1.4	0.2	
	4	29	735.3	196.4	3.7	< 0.01	5.1	3.3	1.6	0.1	

Notes:

1) Depth represents feet from surface, e.g., depth=4 includes all samples from top 4 feet of material.

2) P = p-value for a two-sided Student's t test that the given regression parameter is 0.

4) Far right column represents plant Se upper one-sided 95% prediction limit (UPL) for the sample area with the largest observed soil Se concentration (ug/kg; water soluable). Calculated only for slopes with observed significance less than or equal to 0.05.

# Figure 3: Cool season grasses regression

Regression of *mine-area* averages for cool season grasses  $Se_{PLT}$  (µg/kg dry wt) vs  $Se_{HW}$  (µg/kg) in the top 4 ft of soil material



# Figure 4: Shrub regression

Regression of *mine-area* averages for shrub Se ( $\mu$ g/kg dry wt) vs Se<sub>HW</sub> ( $\mu$ g/kg) in the top 4 ft of soil material



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## Conclusions

- Mine-wide: Though reclamation has higher Se<sub>PLT</sub>, biological significance of increase is questionable since (1) all Se<sub>PLT</sub> mine averages for herbaceous vegetation are at least an order of magnitude below NOAEC of 5000 µg/kg and the largest UCL is 16.6% of that value, and (2) the highest Se<sub>PLT</sub> mine average for shrubs is 29.6% of the NOAEC and the largest UCL is approximately 1900 µg/kg, or 38.2% of the NOAEC.
- Mine-areas: Positive soil-plant correlations were highly significant for both cool season grasses and shrubs for *mine-areas*. This finding allows risks to be assessed for these plant types over the entire observed range of Se<sub>HW</sub> values in SW coal mines using the UPL of the fitted regression line. Likewise, risk could be speculated for Se<sub>HW</sub> higher than those observed. For the largest observed material Se<sub>HW</sub>, the UPL for cool season grasses was 342 µg/kg, over an order of magnitude less than than the NOAEC. The respective UPL for shrubs was 2366 µg/kg, 47.3% of the NOAEC.
- Together, these results suggest that at these SW coal mines, regraded spoils that have higher average Se<sub>HW</sub> present little to no potential to produce average Se<sub>PLT</sub> that would adversely affect domestic herbivores. On both a *mine-wide* or *mine-area* scale, Se<sub>PLT</sub> in reclamation are similar to those on native soils and well below toxic concentrations. It appears that the resumption of grazing following mining poses a minimal to nonexistent risk of Se toxicity to livestock in the Southwest.

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  - La Plata Mine