Evaluating the Toxicity of Arsenic and Lead in the Tacoma Smelter Plume and the Hanford Site Old Orchards Areas

By Janice Sloan and Dave Sternberg, Washington State Department of Ecology, Olympia, WA

Study Questions:

Are current arsenic and lead soil screening levels under the Model Toxics Control Act (MTCA) protective of wildlife in the Tacoma Smelter Plume and Hanford Old Orchards areas?

Do arsenic species and soil type need to be considered for setting ecologically-relevant soil screening levels for arsenic and lead?

In Washington State, air emissions from metal smelters and the use of lead arsenate pesticides have resulted in widespread arsenic and lead soil contamination well above natural background concentrations. Elevated levels of these metals can pose a risk to wildlife including decreased reproductive success, decreased growth, and behavioral changes.[1,2]

Study Areas

The two study areas are the Tacoma Smelter Plume (TSP) footprint, a 1,000 square mile area surrounding Tacoma, WA (arsenic & lead source = smelter stack emissions) and the Hanford Old Orchards (HOO) area within the U.S. Department of Energy Hanford Site (arsenic & lead source = use of lead arsenate pesticides). Figure 1 shows the study area locations, and Figure 2 is a timeline of each area's history.

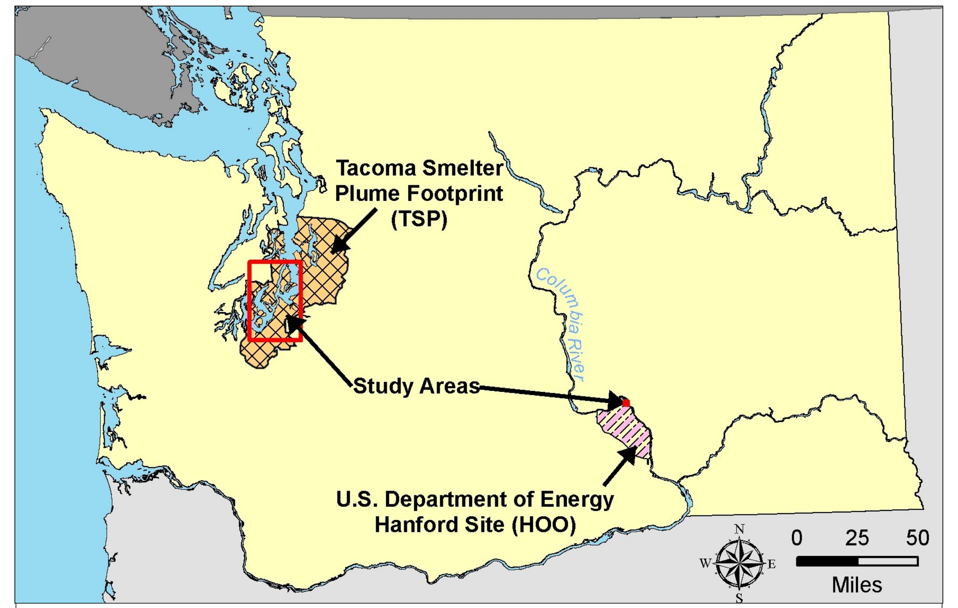

Figure 1. Map of study and contaminated areas.

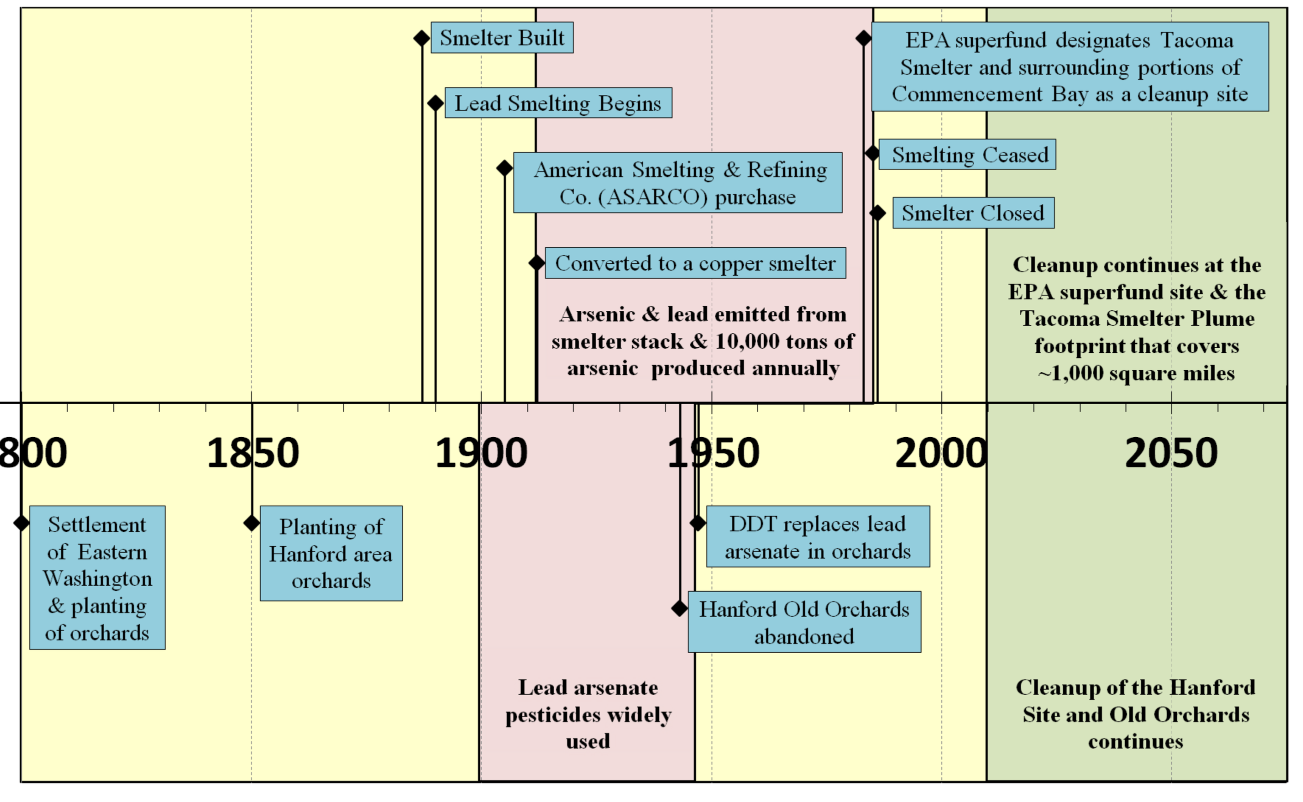

Figure 2. Timeline for historical contamination in the two study areas.

Why now?

In both areas cleanup has been focused on human health concerns, but as cleanup progresses ecological impacts are being considered. However, the ecological impacts of arsenic and lead contamination in these two areas is poorly understood. The data from this study will influence ecologically-relevant cleanup decisions for arsenic and lead contaminated soils in these two areas. In particular, risks to wildlife posed by contaminated soils were evaluated to determine if current soil screening levels accurately predict risks to wildlife in the Tacoma Smelter Plume footprint and the Hanford Old Orchards area.

Why was this study needed?

The size of the two study areas makes it difficult to conduct an in-depth Terrestrial Ecological Evaluation (TEE) under the Washington Department of Ecology's, Model Toxics Control Act (MTCA). Gaining knowledge about the toxicity of arsenic and lead in these areas will inform targeted cleanup efforts about ecological risks particular to each area. TEE ecological risk assessments use soil screening levels (SSL) derived from simple bioaccumulation models to evaluate ecological risk. If SSLs a re exceeded, the SSL may be used as a conservative cleanup level for the site, or additional site-specific evaluations may be performed. Therefore, it is important that SSLs adequately protect wildlife while considering the ecological and monetary expense of setting these values too low.

Site selection

A total of 25 sampling sites were selected in the Tacoma Smelter Plume area, and 11 sites were selected in the Hanford Old Orchards area. A variety of locations were sampled to cover a range of factors and concentrations. Sampling locations were selected based on the following criteria:

- ♦ located within the study areas.
- ♦ represent a range of major soil types (TSP area only, Table 1, Figure 3).
- ♦ represent a range of arsenic and lead concentrations.
- ♦ accessible for sampling.
- ♦ relevant to or part of a cleanup site.
- ♦ support or have the potential to support wildlife.

Figures 4a and 4b show typical HOO and TSP sampling locations, respectively. Figure 6 shows the names, locations, and soil types of the selected sampling locations.

Figure 4a. HOO typical location.

Figure 4b. TSP typical location.

Table 1. Tacoma Smelter Plume footprint soil series used as the basis for distinguishing between soil types.

Soil Series	Origin	Drainage	Texture	Friable?	Sticky?	Plastic	Other Characteristics	% of TSP footprint [1]
Alderwood (Ald)	Glacial Till	Moderate	Gravelly Ashy Sandy Loam	Very	Slightly	Slightly	Prone to high water table due to cemented layer	25% +1% Everett[2] +3% Kitsap[2]
Everett (Evt)	Glacial Outwash	Excessive	Very Gravelly Sandy Loam	Very	No	No	Rocks clean and arranged in layers	8%
Harstine (Har)	Glacial Till	Moderate	Gravelly Ashy Sandy Loam	Very	-	Slightly		8%
Spanaway (Spn)	Glacial Outwash	Excessive	Gravelly Sandy Loam	Very	No	No	High organic matter content	4% +5%[3]
Kitsap (Kit)	Glacial Lakebed	Moderate	Silt Loam	Moderate	Slightly	Slightly		2%

Un-mapped area of the TSP = 25%.
[1]TSP = Tacoma Smelter Plume. Areas do not include waterbodies.
[2]Mapping was not detailed enough to distinguish between these series.
[3]Other very similar soil series.
Soil Survey Staff (2008) and personal communication with Chuck Natsuhara, Natural Resource Conservation Service.

Figure 3. KCO-Everett soil

Sampling

At each sampling location, soil, soil biota, and plants (roots, stems, and leaves) were collected. Standard EPA approved methods were used for all analyses. The parameters measured were:

Soil Only	Soil, Soil Biota, and Plants
pH	Arsenic
Arsenic (III)	Lead
Arsenic (V)	
Solids	
Total Organic Carbon	

Soil Collection

Soil was collected from 0-6 inches in depth from a central location and from each cardinal direction a randomized distance from the center. The soil was composited, homogenized, then split for analysis.

In the TSP area, arsenic speciation was conducted on soils from two highly contaminated sites in each soil series.

Soil Biota Collection

Soil biota were collected within the soil sampling area or no further than 10 feet from any soil sample. Soil biota were depurated for 48 hours, rinsed with deionized water, and frozen until analysis. Earthworms were targeted in the TSP area and darkling beetles in the HOO area (Figure 5).

For TSP sites BURTON-Evt and MIMP-Ald-UNK, an assortment of soil biota and at KOPA-Har no soil biota, were collected. At HOO sites 01, 05, 06, 07, and 09, darkling beetles were found. Two samples of darkling beetles and two samples of other invertebrates were collected throughout the HOO area to obtain enough mass for analysis.

Plant Collection

Plants were collected within the soil sampling area or no further than 10 feet from any soil sample. Plants were rinsed with tap water to remove soil, then rinsed with deionized water, and frozen until analysis. English ivy, evergreen huckleberry, grass, Oregon grape, salal, and one unknown plants were collected in the TSP area. Cheatgrass was the only plant collected at the HOO locations.

Figure 5. Weighing collected earthworms.

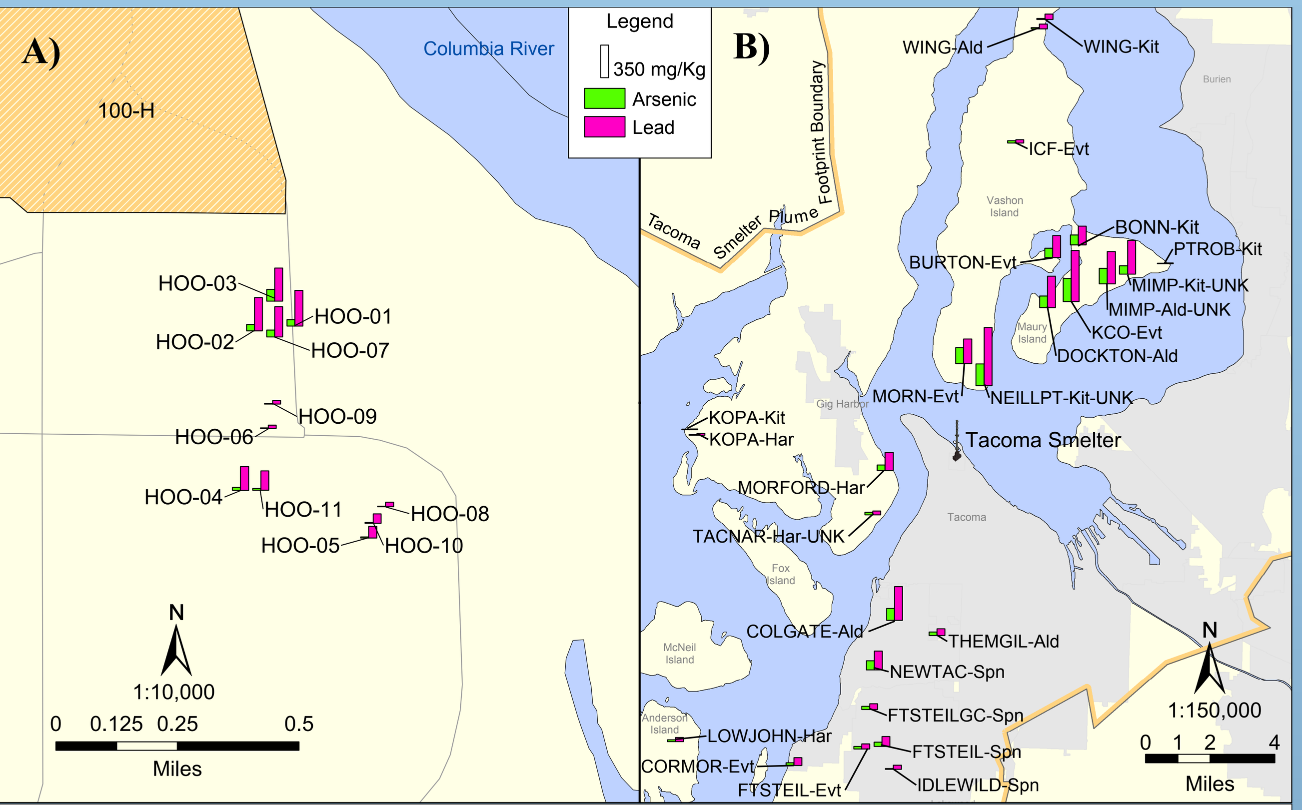

Figure 6. Maps of study areas and arsenic and lead concentrations.
Map of the two study areas relative to contaminated areas: (A) Hanford Old Orchards locations, and (B) Tacoma Smelter Plume footprint locations labeled with abbreviation of name - Soil Series (see Table 1) - "UNK" soil series was not verified.

Tacoma Smelter Plume versus Hanford Old Orchards

Combining the two areas for analysis was considered as part of this project to facilitate a more statewide approach to setting SSLs. However, it was decided to keep them separate due to several factors:

- ♦ Linear regressions of arsenic-versus-lead soil concentrations of the TSP and HOO areas were compared. The slopes of the regressions were not statistically different, while the intercepts were statistically different. This indicates that the two data sets are distinct but parallel and therefore should be considered separately (Figure 7).
- ♦ The wildlife exposure model receptors are unique to each area and resulted in different SSL values.
- ♦ These two sites are being considered individually for cleanup actions.

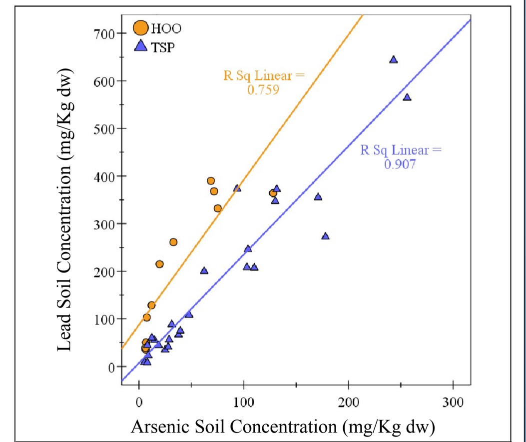

Figure 7. Arsenic versus lead soil concentration.

Wildlife exposure model

Per the TEE process, a simple wildlife exposure model was used to determine SSLs. The model uses soil and food chain interactions to predict the accumulation of contaminants in three wildlife receptor species groups, avian predators, mammalian herbivores, and mammalian predators. Each receptor species group for this study was represented by a surrogate species as shown in Figure 8. The parameter values used are from MTCA Tables 749-3 through 5 for the TSP and HOO, additional HOO specific surrogate species values from Doctor et al. (2000) were also used. The lowest SSL produced from the exposure model can be used for site evaluations.

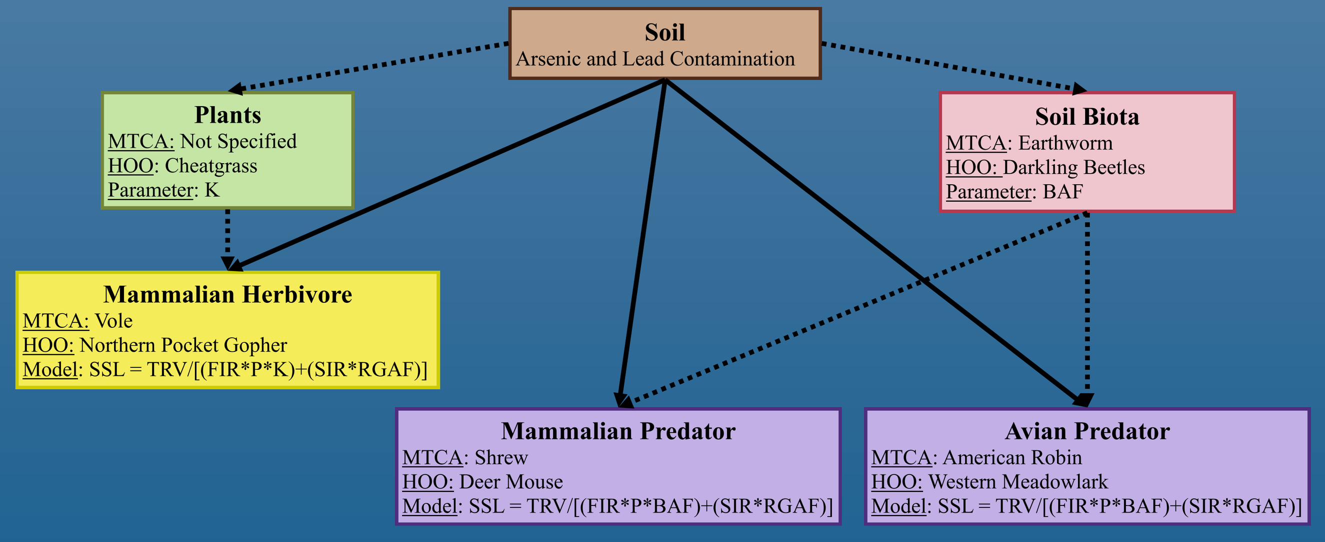

Figure 8. Diagram of the wildlife model used in a Terrestrial Ecological Evaluation.
Outline of receptor species group parameters for the TSP and HOO areas used in the TEE wildlife model. Dotted lines represent the food pathway while solid lines represent the direct ingestion of soil pathway for arsenic and lead to enter organism.

Measured Parameters
BAF = Bioaccumulation factor
K = Plant Uptake Coefficient

Parameters from MTCA or Doctor et al. (2000)
P = Diet contamination
FIR = Food ingestion rate
SIR = Soil ingestion rate
RGAF = Gut adsorption factor
TRV = Toxicity reference value

Is arsenic species important to consider in terrestrial ecological risk?

Arsenic (III) is the more toxic than the arsenic (V) species in laboratory toxicity tests.[4,5] Therefore separate SSLs were set in MTCA for arsenic (III) and arsenic (V), rather than using total arsenic.

Arsenic (III) concentrations in this study ranged from a minimum of 0.086 mg/Kg dw at HOO-06 to 1.930 mg/kg at KCO-Evt (Figure 9). The MORFORD-Har arsenic (III) concentration represented the greatest proportion of total inorganic arsenic at 12.3%, while at the remaining sites arsenic (III) represented less than 1.1% of the total arsenic. None of the arsenic (III) concentrations exceed the MTCA TEE SSL of 7 mg/kg dw for the protection of wildlife. The arsenic (V) concentrations were only slightly less than the total arsenic concentration, and only three sites exceed the MTCA TEE SSL of 132 mg/Kg dw.

Due to the lack of arsenic (III) in the samples, use of total arsenic values for the protection of wildlife is justified. However, this only applies to dry soils, not saturated or inundated soils where arsenic (III) may be more prevalent.

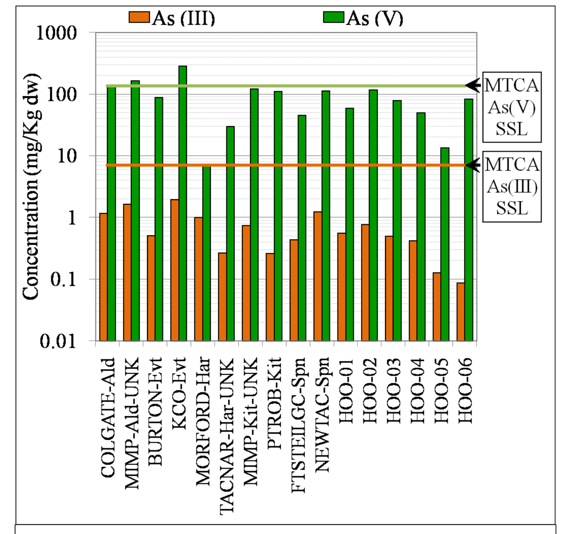

Figure 9. Arsenic species results.

Does soil type influence plant and worm bioaccumulation of arsenic and lead?

The null hypothesis was that soil type does not influence the body burden of arsenic and lead in plants and worms. This hypothesis was evaluated for TSP locations with confirmed soil types. Additional factors that may influence uptake of metals were combined with soil type to see if there was a significant predictive model that included soil type. K and BAF values were used as the dependent variables.

The only significant combination of factors was for predicting arsenic K. The factors consist of soil type, arsenic level, and plant type (Model p = 0.008, soil type p = 0.014, $R^2 = 0.783$; Table 3). Tukey HSD multiple comparisons of arsenic K values for the Alderwood, Everett, and Spanaway series were significantly lower than those of the Kitsap series (Table 4). Since the Kitsap series is the only silt loam soil while the remaining series are sandy loams, it makes sense that this series may exhibit different accumulation of arsenic in plants. Table 3 summarizes the best model runs, Table 4 shows the multiple comparison results, and Figure 10 shows box plots of K and BAF by soil type and area.

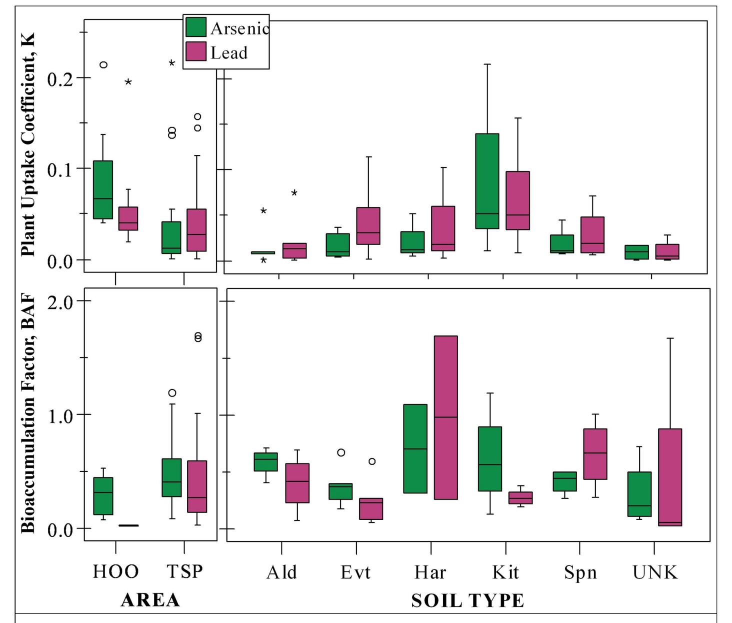

Figure 10. K and BAF boxplots by area and soil type.

Table 3. Arsenic and lead K and BAF predictive model results.

Test	Dependent	Model	Sig.	R^2
GLM	Arsenic K	Intercept + SOIL_TYPE + AS_LEVEL + PLANT_TYPE + SOIL_TYPE * PLANT_TYPE + AS_LEVEL * PLANT_TYPE	0.008**	0.783
	Arsenic BAF	Intercept + SOIL_TYPE + TOC_SOIL	0.053	0.532
Kruskal-Wallis	Lead K	SOIL_TYPE	0.397	
	Lead BAF	SOIL_TYPE	0.180	
GLM	Lead K	PLANT_TYPE	0.055	0.087

AS_LEVEL = Expected arsenic level that was used to select each location; represents the project design.
PLANT_TYPE = Herbaceous or woody.

Table 4. Arsenic K multiple comparisons of soil type.

	Ald	Evt	Har	Spn
Evt	1.00			
Har	0.999	0.999		
Spn	1.00	1.00	1.00	
Kit	0.013*	0.004**	0.062	0.024*

* p≤0.05
** p≤0.01

Calculating a Plant Uptake Coefficient (K) or Bioaccumulation Factor (BAF) value for the Tacoma Smelter Plume and Hanford Old Orchards areas

This study aims at establishing an area-specific K and BAF value for use in large-scale cleanup efforts that span a range of concentrations. Therefore a regression model approach for calculating K and BAF values was not appropriate even though it may be more accurate for site-specific uses. Instead the median K and BAF values for the data sets will be used. This approach follows previous establishment of statewide K and BAF values by Ecology and the EPA method used when an acceptable regression model is not available. Table 5 shows the median K and BAF values by soil type and area.

SSL values for the Tacoma Smelter Plume and Hanford Old Orchards areas

Using current MTCA parameters, SSL values based on the median K and BAF for the TSP and HOO areas are considerably higher than the current MTCA SSLs of 132 and 118 mg/Kg for arsenic (V) and lead, respectively (Table 5). This indicates that the median area-specific K and BAF values are lower than current K and BAF values in MTCA. Thus, the current MTCA SSL values are overprotective of wildlife if current MTCA model parameter values are used.

Using HOO-specific surrogate receptor species increased the HOO SSL values from 408 to 593 mg/Kg and 482 to 504 mg/Kg for arsenic and lead, respectively. Therefore the default MTCA surrogate receptor species provide a more conservative estimate of risk.

New updated literature values for the wildlife exposure model were not evaluated here but are expected to result in lower SSL values even when using field-derived K and BAF values.

Table 5. K, BAF, and SSLs for the Tacoma Smelter Plume area, the Hanford Old Orchards area, and current MTCA values.

	Median Plant Uptake Coefficient, K			Mammalian Herbivore SSL		Median Bioaccumulation Factor, BAF			Mammalian Predator SSL		Avian Predator SSL		Lowest SSL Value	
	N	Arsenic	Lead	Arsenic	Lead	N	Arsenic	Lead	Arsenic	Lead	Arsenic	Lead	Arsenic	Lead
TSP	32	0.0125	0.0271	2,954	1,217	24	0.403	0.268	367	309	**339**	**225**	339	225
HOO[1]	11	0.0667	0.0403	1,211	971	9	0.302	0.018	484	2340	**408**	**482**	408	482
HOO[2]				1,211	1,700				1,452	3,528	**593**	**504**	593	504
MTCA Values [3]		0.06	00047	1,306	2,132		1.16	0.69	**132**	125	150	**118**	132	118

Lowest SSL bolded.
[1] HOO SSLs using MTCA default surrogate species values.
[2] HOO SSLs using Doctor et al. (2000) surrogate species values.
[3] Arsenic (V) values displayed.

Conclusions and recommendations

- ♦ Based on current MTCA wildlife exposure model parameters, MTCA SSLs are overprotective of wildlife. Area-specific median K and BAF values produced arsenic and lead SSLs that were higher than SSLs derived from current MTCA K and BAF values.
- ♦ Total arsenic should be used for establishing SSL values for dry soils; arsenic species should only be considered for wetted soils.
- ♦ Only the significant model including soil type was for arsenic K values. The difference may logically be attributed to differences in soil texture; however, more evidence needs to be collected to conclusively determine this effect.
- ♦ Despite the evidence that soil texture may influence arsenic uptake by plants, it is recommended to establish one K and one BAF value for each of the study areas. This recommendation reflects the practicality of using one value for an entire area and the fact that the Kitsap soil, which was significantly different from the other soil series, represents a very small portion of the TSP soils.

Further information

The final report for this project will: look at wildlife exposure model parameters from literature, compare XRF data to laboratory data for arsenic and lead, evaluate bioassays conducted on these soils, recommend SSLs for the TSP and HOO areas, and provide more detail on the findings presented here.

This poster can be viewed online at www.ecy.wa.gov/biblio/1003062.html along with the project plan and the final report (early 2011) in the *Related Publications* section. For questions about this project, contact Janice Sloan at Janice.Sloan@ecy.wa.gov. For questions about the TEE process, contact Dave Sternberg at DAST461@ecy.wa.gov.

Abbreviations

Ald	- Alderwood Series	Har	- Harstine Series	SIR	- Soil Ingestion Rate
As	- Arsenic	HOO	- Hanford Old Orchards	Spn	- Spanaway Series
BAF	- Bioaccumulation Factor	K	- Plant Uptake Coefficient	SSL	- Soil Screening Levels
dw	- Dry weight	Kit	- Kitsap Series	TRV	- Toxicity Reference Value
Ecology	- Washington State Department of Ecology	MTCA	- Model Toxics Control Act	TEE	- Terrestrial Ecological Evaluation
EPA	- U.S. Environmental Protection Agency	P	- Diet Contamination	TSP	- Tacoma Smelter Plume
Evt	- Everett Series	Pb	- Lead	UNK	- Unknown
FIR	- Food Ingestion Rate	RGAF	- Gut Adsorption Factor		

Acknowledgements

- ♦ Funding for this project was provided by (1) EPA Region 10 through the 128 (a) State and Tribal Response Program Grant and (2) Washington State Department of Ecology's Nuclear Waste Program.
- ♦ Land owners and park managers granted access to their properties and allowed sampling.
- ♦ U.S. Department of Energy Hanford Site staff helped plan this project and provided access to the site.
- ♦ Nautilus Environmental Laboratory conducted the bioassay tests and provided interpretation of the results.
- ♦ Brooks Rand Laboratory conducted the arsenic speciation analysis.
- ♦ Chuck Natsuhara, USDA Natural Resource Conservation Service, helped with soil identification and guidance on soil series selection.
- ♦ Washington State Department of Ecology staff helped with this project from conception to sampling to analysis, including staff at Manchester Environmental Laboratory who provided analysis of chemical analytes and contracting.

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