TECHNICAL MEMORANDUM

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TO: Dave Bradley, Washington State Department of Ecology

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RE: ARSENIC AND LEAD MOBILITY IN AREA-WIDE CONTAMINATION-IMPACTED SOIL

INTRODUCTION

In June 2003, the Area-Wide Soil Contamination Task Force (Task Force 2003) issued a comprehensive report on the nature and extent of low to moderate shallow soil metals contamination dispersed over large geographic areas of the State. The report also outlined a remedial action strategy for addressing area-wide contamination. Subsequently, the Washington State Department of Ecology (Ecology) established a remedial action strategy for area-wide soil with moderate levels of arsenic and lead, based on preventing direct contact, especially by children, with contaminated soil. Concentrations that are considered moderate levels depend on the land use and are shown in Table 1. One of the assumptions underlying the area-wide strategy is that shallow soil containing moderate levels of arsenic and/or lead is unlikely to impact underlying aquifers. The purpose of this technical memorandum is to summarize existing technical information on arsenic and lead mobility as it relates to the potential of area-wide contamination to impact groundwater.

TABLE 1
MODERATE CONCENTRATIONS OF ARSENIC AND LEAD

Land Use	Arsenic (mg/kg)	Lead (mg/kg)
Schools, childcares, residential properties	20 - 100	250 - 500
Parks, commercial properties	20 - 200	250 - 700

SUMMARY OF CONTAMINATION IMPACTS

Area-wide contamination includes arsenic and lead soil impacts primarily from historical metal smelters and agricultural operations. The primary smelter contamination source is historical operation of the Tacoma Smelter near Tacoma, Washington from 1890 to 1986.[1] The primary agricultural

[1] Three other smelters (Everett, Northport, and Trail) were identified in the Task Force Report (2003); however, the soil impacts from these other smelters in Washington State are appreciably less significant than the Tacoma Smelter impact.

contamination source is application of lead arsenate pesticides to apple and pear orchards in central and eastern Washington during the first part of the 20th century (Task Force 2003).

Tacoma Smelter Plume Impacts

Tacoma Smelter area-wide impacts are associated with gas and particulate emissions from the 562 ft-high main stack. Total atmospheric emissions were estimated at 300 metric tons/year. Stack emissions were composed of up to 70 percent arsenic and lead (Crecelius et al, 1974). Deposition of the stack emissions occurred over a wide area in the principal wind directions, extending from approximately the Snohomish/King County line to north Thurston County.

Widespread arsenic and lead soil contamination in Puget Sound is attributed to the Tacoma Smelter (Task Force 2003). In the 2003 Task Force Report, a Level 1 area of 240 square miles around the smelter was defined where undisturbed shallow soil concentrations of arsenic were likely to exceed the Model Toxics Control Act (MTCA) Method A soil cleanup level of 20 mg/kg. More recently, Ecology issued a final "Footprint" report on the Tacoma Smelter Plume (TSP) (Pacific Groundwater Group and TeraStat Inc. 2005).[2] Over 4,000 TSP soil samples were analyzed for arsenic and lead, typically in relatively undisturbed soil. These soil samples defined the footprint of soil contamination originating from the smelter. Outside of the immediate Superfund site area in the town of Ruston, the maximum arsenic concentration was 1,100 mg/kg; the maximum lead concentration was 6,700 mg/kg. However, excluding north Pierce County, Maury Island, and south Vashon Island, arsenic concentrations are typically less than 200 mg/kg and lead concentrations are typically less than 400 mg/kg. Arsenic and lead concentrations decrease in the principal downwind directions with distance from the smelter. Concentrations also decrease with soil depth (Glass 2004, Landau Associates 2003). Arsenic and lead concentrations tend to be highly variable over short distances within impacted areas due, in part, to soil disturbance [Seattle-King County Public Health (SKCPH) and Glass 2000]. Various reports that document soil concentration data in the TSP are available from the Ecology web site (http://www.ecy.wa.gov/programs/tcp/sites/tacoma_smelter/ts_hp.htm).

Agricultural Impacts

Widespread arsenic and lead soil contamination in historical orchard areas of central and eastern Washington is attributed to the use of lead arsenate pesticides. Lead arsenate was used to control chewing insects; in particular, the codling moth, a pest that infests apple and pear fruit. Apple and pear production

[2] Note that the TSP "footprint" was defined in phases with interim "footprint" reports issued for portions of Pierce and King Counties. The final footprint study report represents a summary of these earlier reports along with more recent data.

peaked in the 1920s and 1930s in Washington State (Peryea 1998). The use of lead arsenate was virtually eliminated in 1948 with the introduction of DDT as an alternative pesticide.

The composition of acid lead arsenate was approximately 22 percent arsenic and 44 percent lead.[3] Application rates peaked in 1943 at an estimated 191 lbs/acre lead and 71 lbs/acre arsenic (Peryea 1989). The primary areas that were likely affected by lead arsenate use were the dominant fruit growing regions of Washington State in the first part of the 20th century; namely Yakima, Chelan, Okanogan, and Spokane Counties (Task Force 2003); however, historic orchard lands also exist in other counties, including counties in western Washington. Yakima County had the highest estimate of potentially impacted land at 58,050 acres (equivalent to about 90 square miles).

Unlike estimates of the TSP, the estimate of the impacted area associated with agricultural practices is based on estimates of cultivated acres. However, there are numerous studies that document the distribution of arsenic and lead in historical orchard soil. Washington State University research documented the effect of lead arsenate on orchard soil (Peryea 1989, Peryea and Creger 1994). There are also numerous reports submitted to Ecology under MTCA that document the distribution of arsenic and lead in historical orchard soil; some of these reports are summarized in the 2003 Task Force report. The U.S. Bureau of Reclamation (USBR) collected over 1,600 soil samples in the Manson area along Lake Chelan at four different depths to study the affect of lead arsenate on replanting failure of apple trees (Ecology 2003). In the Washington State University studies, lead was detected up to 2,213 mg/kg; arsenic was detected up to 363 mg/kg. In the Manson area study, arsenic was detected up to 441 mg/kg (lead was not analyzed) [4]. Arsenic and lead concentrations in agricultural soil also tend to be highly variable over short distances due to pesticide application practices (Veneman et al. 1983).

CONCEPTUAL MODEL OF ARSENIC AND LEAD MOBILITY

Arsenic and lead tend to have low mobility in shallow soil. Factors that affect mobility include metal concentration, metal speciation, soil redox and pH conditions, precipitation, soil composition and permeability, and application of phosphate fertilizers.

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[3] The lead/arsenic ratio found in agricultural soil ranges from about 3 to 5. In eastern Washington, acid lead arsenate was typically used due to the alkaline nature of the soil. In other areas with more acid soil, basic lead arsenate was used.

[4] A single sample out of 1,600 exceeded 441 mg/kg. The concentration of this outlier was 1,100 mg/kg.

Arsenic

Arsenic can occur in four oxidation states. In the range of Eh and pH found in soil, the +5 and +3 states predominate, corresponding to arsenate (AsO_4^{-3}) and arsenite (AsO_3^{-3}) (Kabata-Pendias 2001). The attached Eh-pH diagram shows the dominant forms of arsenic in a water-arsenic system (Figure 1). Shallow oxidized soil conditions (i.e., conditions that are not flooded) have a redox potential from about +400 to +700 mV (Dempsey 1991, Peryea 1989). In this environment, arsenate is the dominant arsenic form. In flooded, poorly drained soil, arsenite may be the dominant form.

Arsenic is discharged from the Tacoma Smelter stack predominantly as arsenic trioxide (i.e., arsenite). However, arsenite is rapidly oxidized to arsenate in oxidizing conditions (Dempsey 1991). For example, chemical speciation of rainwater arsenic in five samples collected downwind of the Tacoma Smelter showed 48 percent to 82 percent as arsenate rather than arsenite (Dempsey 1991). Consequently, arsenite is not expected to persist long in an aerated soil environment. In general, arsenate is the dominant arsenic species under most environmental conditions (Kabata-Pendias 2001, Sadiq 1997). Therefore, it is a reasonable assumption that arsenic from both smelter deposition (which originates as arsenite) and agricultural deposition [which originates as arsenate ($PbHAsO_4$)], occurs in shallow soil predominantly as arsenate.

Arsenic released onto the ground will either be bound to the soil or dissolved into groundwater. Arsenic adsorption to soil is a complex process that is not completely characterized. In most shallow soil conditions, arsenic occurs primarily as anionic arsenate in the form of an oxyanion ($H_2AsO_4^-$) (Sadiq 1997, Kabata-Pendias 2001). Therefore, arsenic retention is controlled by the anion exchange capacity, which is related to the positive charge available on soil surfaces (Dempsey 1991). Positive soil charge in turn is related to oxides; aluminum and iron oxides appear to be the most important parameters controlling arsenate retention in acid to neutral soil (Kabata-Pendias 2001, Tokunaga and Hakuta 2001, Sadiq 1997). According to Darland and Inskeep (1997), arsenate is preferentially adsorbed to iron and aluminum hydroxides compared to kaolinite, montmorillonite, calcite, and quartz; and iron and aluminum hydroxide adsorption capacity increases with decreasing pH to a maximum at about pH 3. Adsorption onto calcite and layered silicate minerals (i.e., clays) was highest in the pH range 4 to 6. Arsenate adsorption onto crystalline quartz was negligible. Coprecipitation is a related process where arsenic may be incorporated into the mineral structure as it forms. For example, arsenic can be coprecipitated during the formation of iron oxides.

Tokunaga (2005) evaluated arsenic retention capacity for six different soil types (Table 2 below).

TABLE 2
CHEMICAL COMPOSITION OF SOIL SAMPLES
(AFTER TOKUNAGA 2005)

Soil	Al2O3	SiO2	Fe2O3	org-C	Water
PY Pseudogleyed yellow-brown forest soil	4.06	14.95	67.91	0.30	4.63
RY Red-yellow soil	15.18	54.36	20.31	0.49	4.52
YB Yellow-brown forest soil	21.04	362.86	11.78	0.70	29.33
KS Kuroboku soil	19.45	37.98	15.02	7.33	13.49
KP Kanuma pumice	34.34	46.67	2.73	0.10	11.31
TG Thionic gley soil	20.62	47.74	8.14	0.81	8.34

The percent adsorption for arsenate was near 100 percent for most soil types in the pH range of 3 to 7.[5] At a pH above 7, sorption capacity dropped off significantly. Adsorption of arsenite on the same soil was lower, but less pH dependant. Arsenite and arsenate adsorption results are shown on Figures 2a and 2b, respectively. Tokunaga (2005) also estimated the sorption capacity of each of the six soil types. Sorption capacity ranged from 0.038 mmol/g (for the RY soil) to 0.282 mmol/g (for the PY soil) (see Table 2 for a description of soil type). These values equate to adsorbed arsenic concentrations between 2,850 mg/kg and 21,130 mg/kg. Note that the sorption capacity was highest for the soil with the highest percentage of iron and aluminum oxide and lowest for the soil with the lowest percentage of iron and aluminum oxides.

Based on the experiments of Tokunaga (2005), arsenic has low mobility in typical shallow soil in the acid to neutral range. Soil in the Puget Sound region tends to be strongly acid (pH 5.1 to 5.5) to slightly acid (pH 6.1 to 6.5) based on soil surveys for King and Pierce County. Soil in agricultural areas of central Washington, however, tend to be neutral (pH 6.6 to 7.3) to moderately alkaline (pH 7.9 to 8.4) based on soil surveys of Yakima and Chelan Counties. However, the optimum pH range for growing tree fruit is slightly acid to neutral and application of ammonium sulfate or ferrous sulfate was recommended for orchard cultivation in alkaline soil (Soil Conservation Service 1975). Consequently, the alkaline nature of central Washington soil may have been modified to some degree to optimize orchard production. The effect of this recommended practice on soil pH and arsenic and lead mobility has not been quantified.

Phosphate fertilizer[6] has been used in replanted orchards to enhance the growth of young trees. Laboratory column experiments performed by Washington State University found that the addition of phosphate fertilizers significantly increased the movement of arsenic, while lead movement was not

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[5] 25 mL of As(V) at a concentration of 0.2 mmol was mixed with 25 grams of soil. The resultant suspension was filtered and the arsenic concentration analyzed using ICP-AES. A similar procedure was used for arsenite.
[6] Typical phosphate fertilizers include monoammonium phosphate (MAP) and triple superphosphate (TSP).

affected (Davenport and Peryea 1991, Peryea 1989). Column experiments by Darland and Inskeep (1997) produced similar results. Increased arsenic mobility appears to be related to exchange of solution-phase orthophosphate for solid-phase arsenic. The concurrent lack of mobility of lead may be related to the formation of lead-phosphate minerals of low solubility (Peryea 1989). The application of phosphate fertilizer is probably not a high risk factor for arsenic remobilization in residential soil as loading rates and application areas are likely to be limited in extent.

In summary, arsenic likely occurs as arsenate in typical shallow soil conditions. Arsenic mobility in typical shallow soil conditions is likely to be limited, but potentially variable depending on soil pH and soil type. Arsenic mobility can increase by either increasing pH or reducing the oxidation state from arsenate to arsenite. In alkaline soil or silica-rich sandy soil, arsenic would be expected to migrate more readily. Similarly, soil liming or mixing soil with cement could increase pH and mobilize arsenic. Arsenic is assumed to persist as arsenate; however, more mobile arsenite may persist in poorly drained soil where reducing conditions predominate (e.g., wetland or low lying areas). Many poorly drained soil areas represent groundwater discharge locations and are limited in areal extent.

Lead

Lead occurs in soil as a stable Pb^{2+} cation (Kabata-Pendias 2001). In this form, lead will typically complex with organic matter, sorb on oxides and clays or precipitate as carbonates, hydroxides, and phosphates. The dominant forms of lead in a lead-water system are shown on Figure 3.

Lead in lead arsenate is probably not stable in a shallow soil environment and is likely converted to relatively insoluble lead hydroxide and lead phosphate minerals (Peryea 1998). Various lead minerals have solubility products that indicate lead solubility would be low under typical Eh-pH conditions (Fetter 1993). Cation exchange capacity (CEC) also affects lead mobility. The CEC is determined by the amount of negative surface charge on soil surfaces and is correlated with soil surface area (i.e., silt and clay content). Generally, soil CEC capacity increases with increasing pH up to neutral to slightly alkaline conditions (Dempsey 1991). Scokart et al. (1983) measured the mobility of lead near a smelter in two soil types; a sandy soil with a well developed humus layer (podzolic soil) and a stony loam. In this study, lead was found to be immobilized in the upper soil layer and that variation in soil pH from very strongly acidic (pH 4.5; podzolic soil) to neutral (pH 7.0; loamy soil) had no discernable effect on mobility. In a study for the U.S. Environmental Protection Agency (EPA), Allison and Allison (2005) determined that lead had the highest soil-water distribution coefficient (i.e., the lowest water mobility) of nine metals studied in soil/water, suspended matter/water, and sediment/water systems. Friedland (1991) concluded that lead (as Pb^{2+}) had the highest potential to form stable metal-humic substances of eight metals studied.

In summary, lead likely is present in the Pb^{2+} oxidation state in typical shallow soil conditions. Lead is expected to strongly partition to soil due to a number of processes in a variety of soil types. Lead partitioning or mobility is not particularly pH dependant in the range of conditions expected for shallow soil.

FIELD DATA AS AN INDICATOR OF ARSENIC AND LEAD MOBILITY

Extensive data exists on the occurrence of arsenic and lead in area-wide contamination-impacted soil. Groundwater data also exists, but is less extensive and less well documented. To some degree, the subsurface mobility of arsenic and lead can be inferred from their distribution in soil and groundwater, decades after the initial contaminant release.

Soil Profile Data

For both agricultural- and smelter-related impacts, arsenic and lead are deposited at the ground surface. These metals migrate into the subsurface through two primary processes, mechanical mixing (e.g., tilling)[7,8] or partitioning into solution at high concentrations and percolating as a dissolved phase in the soil column (i.e., leaching). The occurrence of elevated concentrations of arsenic and lead in soil below the level of mechanical mixing is an indication of the leaching of these metals. However, the metal concentration profile with depth suggests that leached metals readsorb (i.e., are redistributed) deeper within the soil column.

Tacoma Smelter Sites

In areas around the Tacoma Smelter, sampling in relatively undisturbed soil indicates that arsenic and lead is concentrated in the upper 6 inches of the soil column. Elevated concentrations of arsenic and lead generally do not occur below about 24 inches in the absence of soil disturbance.

Crecelius et al. (1985) evaluated the movement of arsenic and lead in soil impacted by the Tacoma Smelter. Three sample profiles were evaluated at a relatively undisturbed site about 1 mile southwest of the former smelter in the City of Tacoma. The soil is described as sandy (the sand and gravel content exceeded 85 percent); the pH ranged from 3.4 (extremely acid) to 5.1 (strongly acid). Samples were collected from seven depth intervals (0 to 2 inch, 2 to 4 inch, 4 to 6 inch, 10 to 12 inch, 23

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[7] Biologically controlled mixing (i.e., insects, earthworms, and rodents) may also be a factor. Peryea and Creger (1994) noted filled-in rodent tunnels in test pits from two of six plots where they evaluated the vertical leaching of lead arsenate in soil. They did not consider this type of mixing as a major process resulting in redistribution of arsenic and lead in the soil column.

[8] Clearly filling and grading can cause burial of shallow arsenic and lead contamination; however, this process is not considered metals migration.

to 26 inch, 35 to 37 inch, 46 to 48 inch). Concentrations of arsenic and lead were highest in the organic-rich upper interval (maximum concentrations between 393 mg/kg and 586 mg/kg) and declined to background concentrations by the 10 to 12 or 23 to 26 inch-interval depending on location. Arsenic and lead concentrations from porewater from the 10 to 12 inch-interval was analyzed from holes 1 and 3 in triplicate. Porewater concentrations were low and usually undetectable.[9] The soil sampling results for the Crecelius et al. (1985) study are presented on Figures 4 and 5 for arsenic and lead, respectively.

Dempsey (1991) evaluated the movement of arsenic at nine sites within the TSP area. Five of the sites were on south Vashon and Maury Islands; one site was on north Vashon Island, two sites were on the King County mainland, and one site was in Kitsap County. The soil was Everett (excessively drained soil that forms under conifers and is underlain by very gravelly sand at 18 to 36 inches) and Alderwood series (moderately well drained soil that forms under conifers and has a weakly consolidated substratum at a depth of 24 to 40 inches); the soil pH typically ranged from extremely acid (below 4.5) to medium acid (5.6 to 6.1). At each location, typically about 6 to 10 samples were collected along a depth profile between a maximum depth of 1.6 ft and 3.7 ft depth depending on location. Arsenic concentrations were as high as about 160 mg/kg; lead concentrations were as high as 1,100 mg/kg. Dempsey (1991) concluded that little or no arsenic moved beyond the O (organic) and A soil horizons (i.e., typically within the upper 6 inches of the soil column). The limited amount of soil movement relative to studies in other areas was attributed to the very low pH and high organic matter of the soil and the presence of sufficient iron, aluminum, and manganese oxides to adsorb the arsenic. Although not a focus of this study, lead showed a similar depth distribution pattern to arsenic in the sample data.

Two gravel pits exist on Maury Island. Arsenic and lead concentration depth profiles were evaluated by Terra Associates (Foster Wheeler Environmental 1999) at the Northwest Aggregates/Glacier gravel mine along the southeast shore of the island. Samples were collected at the surface, 9 inches, and 18 inches at twenty separate locations.[10] Concentrations of arsenic dropped from a maximum of 433 mg/kg at the surface to a maximum of 64 mg/kg at 18-inches depth. Similarly, lead concentrations dropped from 830 mg/kg to 36 mg/kg. At the 18-inch depth, eleven of the twenty samples exceeded the arsenic background concentration of 7 mg/kg (Ecology 1994); three of the samples exceeded the lead background concentration of 24 mg/kg. These data are indicative of some leaching and readsorption of arsenic and lead to a depth of at least 18 inches. A summary of the Terra Associates data is presented on Figure 6. Landau Associates (2001) found similar results in the evaluation of metal contamination depth profiles at the Maury Island Regional Park, a 289-acre former gravel mine that is now a King County park located a few miles north of the Northwest Aggregates/Glacier gravel mine.

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[9] The detection limit for both arsenic and lead was 0.4 µg/L.
[10] Sixteen of the twenty samples were collected from forested (i.e., undisturbed) sample locations at the gravel mine.

For the TSP footprint studies, most samples were collected within the 0 to 6 inch-depth range for undisturbed soil sampling locations. This sampling design reflected the underlying conceptual understanding that arsenic and soil concentrations were highest in this interval. The Vashon-Maury Island study (SKCPH and Glass 2000) compared soil concentrations in the 0 to 2 inch- and 2 to 6 inch-depth intervals; generally, concentrations were slightly higher in the 0 to 2 inch interval, particularly for undisturbed (forest) soil. A similar conclusion was reached during the Pierce County footprint study (Glass 2004). Arsenic was slightly depleted relative to lead in the 0 to 2 inch interval and slightly enriched relative to lead in the 2 to 6 inch interval. These relationships are consistent with arsenic being more mobile than lead in this Pierce County soil. During the Pierce County study, 94 samples were also collected from the 6 to 12 inch interval. These data confirmed that the highest arsenic and lead concentrations occurred at shallower depth intervals.

In summary, two studies looked specifically at the movement of arsenic and lead in the area affected by the TSP and concluded that most of the arsenic and lead in soil associated with the TSP is sequestered in the shallow O or A soil horizons. Additional studies associated with the TSP footprint studies or MTCA cleanups reached a similar conclusion; however, these data suggest that elevated concentrations of arsenic may reach deeper than 18 inches due to leaching and readsorption deeper in the soil column.

Agricultural Sites

In agricultural areas, mechanical mixing or tilling is clearly an important mechanism affecting the depth profile of arsenic and lead contamination. However, tillage[11] typically affects only the top 6 to 8 inches of soil (Peryea and Creger 1994). The presence of arsenic and lead in agricultural soil below this depth is an indication these metals leach and move downward in the soil column.

Veneman et. al. (1983) investigated the distribution of arsenic and lead in a former orchard[12] in western Massachusetts. Arsenic and lead concentrations in surface soil were as high as 330 mg/kg and 1,400 mg/kg, respectively; well above background concentrations. A depth profile of contaminant concentrations was evaluated by sampling every 2.5 cm to a depth of 95 cm (3.1 ft). The highest concentrations of both metals were detected in the upper part of the organic-rich Ap soil horizon[13] within the upper 23 cm (8 inches) of the soil column. Based on arsenic and lead ratios with nickel (a metal that is not a contaminant at the site), it appears that some leaching and redistribution of arsenic and lead has

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[11] Tillage was used in orchards up until about the 1960s for weed control (Peryea and Creger 1994).
[12] The orchard was apparently in production from the late 1800s to 1962.
[13] The Ap horizon refers to the portion of the A soil horizon (the surface horizon characterized by mineral leaching and organic accumulation) disturbed by man's activities.

taken place within the Ap horizon, but not below it. Lead/arsenic ratios decreased with depth in the Ap horizon suggesting that arsenic was leached preferentially to lead. The soil was a silt loam with relatively high iron and aluminum content; the pH was in the medium acid range (5.6 to 6.0). A summary of these data is presented on Figure 7.

Peryea and Creger (1994) evaluated the vertical distribution of arsenic and lead from six contaminated orchard plots in north-central Washington (likely Chelan County) that had been used for commercial fruit production since 1915. The soil pH in each of these plots was similar; the topsoil pH was 5.5 to 6.0 (medium acid), deeper soil pH ranged from 6.5 to 7.0 (neutral). Two of the plots were Cashmont series soil (well drained, moderately coarse textured soil that forms in alluvial and colluvial material of granite, gneiss, schist, and basalt origin); four of the plots were Burch series soil (well drained, medium-textured, and moderately coarse-textured soil that forms in valley fill chiefly of sandstone origin). Soil samples were collected every 5 cm to 10 cm to a depth of 125 cm (4.1 ft). The study showed that most of the arsenic and lead was concentrated in the upper 40 cm (1.3 ft) of soil (i.e., within and below the Ap horizon); maximum arsenic concentrations ranged from 0.77 mmol/kg (57 mg/kg) to 4.85 mmol/kg (359 mg/kg); maximum lead concentrations ranged from 2.15 mmol/kg (444 mg/kg) to 10.69 mmol/kg (2,386 mg/kg). Arsenic was depleted relative to lead in the topsoil and enriched relative to lead in the subsoil indicating preferential movement of arsenic relative to lead. Absolute enrichment of lead occurred to 1.6 ft, while the absolute enrichment of arsenic occurred from 1.5 ft to greater than 4 ft in some plots. The relatively greater mobility of arsenic and lead in this study relative to the Veneman et. al. study was attributed to higher lead arsenate loading rates, coarser soil texture, and lower organic soil content. Iron and aluminum oxide content and soil pH may also have played a role. Results from the Peryea and Creger (1994) study are shown on Figure 8.

The USBR collected 1,600 arsenic soil samples in 253 separate tracts in the Manson area of Chelan County to evaluate orchard replanting failure (i.e., stunting of new trees attributed to high arsenic soil concentrations associated with historical applications of lead arsenate). These data were subsequently evaluated as part of the area-wide soil contamination project to evaluate the effect of historical land use on arsenic soil distribution (Ecology 2003). Predominant soil types were Antilon series soil (well-drained, medium-textured soil formed in loess and material weathered from basalt; mildly to moderately alkaline) and Chelan series soil (well-drained, moderately coarse-textured soil that forms in pumice, volcanic ash, and loess over cobbly deposits of ablation till). The upper Ap soil horizon of both soil types is classified as mildly alkaline (pH 7.4 to 7.8); deeper soil horizons are classified as moderately alkaline (pH 7.9 to 8.4). Samples were collected from four intervals (0 to 12 in, 12 to 18 in, 18 to 24 in, and 24 to 36 in) and segregated into three land use categories (orchard, mixed, and non-orchard). Arsenic concentrations ranged from non-detect to 441 mg/kg (except for one sample result that was 1,100 mg/kg).

Arsenic concentrations were elevated for all three land use types (Figure 9), although the orchard category had the highest concentrations. Concentrations decreased progressively from the shallowest interval to the deepest sample interval. The median arsenic concentration of all three land use categories was below 20 mg/kg by the 18 to 24 inch interval (Figure 10). However, about 20 percent of the samples from all three categories exceeded 20 mg/kg concentration in the deepest interval (24 to 36 inches) (Figure 11).

In summary, arsenic in shallow agricultural soil will redistribute deeper in the soil column due to leaching and readsorption. The degree of leaching is apparently variable. In more acid soil with high iron and aluminum content, leaching may be limited to the A soil horizon. In more alkaline soil typical of central Washington, where metals loading is high, leaching may cause enrichment of arsenic to a depth of over 4 ft. Lead also is leached and redistributed in the soil column, but to a lesser degree than arsenic.

Groundwater Quality Data

Arsenic and lead groundwater quality associated with area-wide contamination have not been evaluated in a focused study. However, groundwater quality data sets do exist that provide a basis for evaluating the area-wide impact to groundwater.

Washington Department of Health (WDOH) maintains a groundwater quality database for Group A and Group B water systems.[14] These data for wells where arsenic has been detected above 10 µg/L can be accessed at the WDOH website (http://www.doh.wa.gov/ehp/dw/our_main_pages/arseniclist.htm). While there are numerous water systems across the State that have detected arsenic above 10 µg/L [the federal maximum contaminant level (MCL) and the Washington State standard for Group A systems], exceedances do not follow a pattern predicted by area-wide contamination trends. For example, Island County has the most arsenic detections (93) and Whatcom County has the most detections above 100 µg/L. A graphical summary of the WDOH data by county is presented on Figure 12 (note this figure just shows detections greater than 10 µg/L).

On Vashon and Maury Islands in King County where area-wide arsenic contamination of soil is relatively severe, three wells have detected arsenic above 10 µg/L as reported by WDOH.[15] However, the reported depth of these wells range from 150 ft to 689 ft suggesting the arsenic is from a natural source. Landau Associates recently assisted Westside Water Association, located on the north end of Vashon Island, with a Group A well installation. The well was installed 300 ft deep and screened in a sand and gravel seam beneath about 280 ft of massive Pre-Vashon age clayey silt. This well had total arsenic concentrations in the 30 µg/L to 40 µg/L range; the cause of elevated concentrations was attributed to a

[14] Group A systems serve 25 or more people or 15 or more residential connections. Group B systems serve less than 25 people and 2 to 14 residential connections.

natural source, probably desorption or dissolution of naturally occurring arsenic under anaerobic groundwater conditions.

King County also maintains a groundwater quality database for wells and springs throughout the County. This database (http://dnr.metrokc.gov/wlr/wq/groundwater-data.htm) includes water quality data on Vashon Island for five Group A systems that are supplied by springs.[16] All of these spring sources have been sampled for arsenic multiple times, but arsenic has not been detected. King County also maintains one surface water sampling location and 19 wells on Vashon and Maury Island that they sample annually as part of a long-term monitoring program that began in 2001. These sampling sites are distributed to provide geographic coverage throughout Vashon and Maury Islands. This program includes sampling for arsenic, chloride, and nitrate as environmental indicator constituents (King County 2006). Of the 20 locations sampled in June 2005, the highest arsenic concentration was 7 µg/L with the exception of wells w-04 (17.5 µg/L) and w-07 (24.1 µg/L). These two wells are 305 ft and 297 ft deep; consequently, it is very unlikely that the source of arsenic in these wells is from area-wide contamination. King County technical personnel also did an evaluation of arsenic in groundwater in King County (Ferguson and Johnson 2006). They noted that two wells (i.e., w-04 and w-07) on Vashon Island exceeded the MCL for arsenic of 10 µg/L; however, the most exceedances and highest concentrations were in East King County, (generally outside the TSP impacted area). They interpreted all exceedances to be consistent with natural leaching of arsenic from locally derived rocks and soil minerals.

The most extensive groundwater quality data set for arsenic appears to be the WDOH and King County data from Vashon and Maury Island. The relatively numerous studies have not concluded there is any anthropogenic arsenic impact to groundwater despite the relatively high concentrations that occur in shallow soil. Lead has generally not been studied to the same degree; however, lead is less mobile than arsenic. Additionally, lead data would be more difficult to interpret due to the presence of potential lead sources in water system piping. In addition to Vashon and Maury Island, the United States Geological Survey (USGS) evaluated water quality in south King County (Woodward et. al. 1995). South King County is within the area impacted by the TSP. The USGS sampled 23 wells for arsenic and other metals. Twelve of the wells were in alluvial or recessional outwash aquifers (likely shallow aquifers). The highest arsenic concentration in any of the 23 wells was 6 µg/L.

Ecology conducted a study of groundwater quality in agricultural areas of Yakima, Franklin, and Whatcom Counties (Ecology 1990). Eight shallow wells were sampled in Yakima County where alfalfa, hops, apples, and cherries are grown (there was no information on historical land use presented in the

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[15] Heights Water and King County Water District 19 wells.

[16] These fives systems are: Westside Water Association, Beulah Park Water System, Dockton Water Association, Maury Mutual Water Company, and Sunwater Beach Water Association.

study). Arsenic concentrations ranged from 2.0 μg/L to 11.7 μg/L. No conclusion was reached in the study on whether the elevated arsenic concentrations were associated with arsenic soil contamination. In Whatcom County and Franklin Counties, the maximum arsenic concentrations were 1.7 μg/L and 13.3 μg/L. Additional study would be required to evaluate if the elevated occurrence of arsenic associated with these studies is related to area-wide contamination impacts.

MODELING

Groundwater impacts associated with area-wide arsenic and lead soil contamination would result from partitioning or leaching of arsenic and lead from the soil phase to infiltrating groundwater. Partitioning can be estimated by modeling.

MTCA/EPA Partitioning Model

One commonly used approach to estimate potential groundwater impacts is the equilibrium partitioning approach that is presented in the MTCA regulation, WAC 173-340-747(4). The same approach is specified in the EPA *Soil Screening Guidance Technical Background Document* (EPA 1996). For metals, this equation is equivalent to:

$$\text{Equation 1: } C_s = C_w (UCF) DF \left[K_d + \frac{\theta_w}{\rho_b} \right]$$

Where:

C_s	=	Soil concentration (mg/kg)
C_w	=	Groundwater concentration or cleanup level (μg/L)
UCF	=	Unit conversion factor (1 mg/1,000 μg)
DF	=	Dilution factor (dimensionless: 20 for unsaturated zone soil)
K_d	=	Distribution coefficient (L/kg; default arsenic = 29; default lead = 10,000)
θ_w	=	Water-filled soil porosity (ml water/ml soil: 0.3 for unsaturated zone soil)
ρ_b	=	Dry soil bulk density (1.5 kg/L).

Equation 1 can be solved for a groundwater concentration that would result from a specified soil concentration. For example, if the average arsenic soil concentration was assumed to be 25 mg/kg, the calculated arsenic groundwater concentration would be 43 μg/L.

Discrepancies Between Model Results and Field Observations

Results of the equilibrium partitioning equation are inconsistent with groundwater data from area-wide contamination areas discussed above. While average area-wide arsenic concentrations in soil are commonly above 25 mg/kg, resulting groundwater concentrations are not commonly above 10 µg/L in underlying groundwater. This discrepancy is potentially due to a number of factors:

- The partitioning model assumes that the entire soil column above groundwater has a uniform concentration equal to the specified soil concentration. For area-wide contamination, the highest concentrations occur typically in the upper 12 inches of the soil column and concentrations typically decline to near background levels within a few feet; therefore, this assumption is likely to result in an overestimate of the groundwater concentration. Implicit in the model assumption of uniform soil concentrations is that arsenic and lead do not resorb deeper in the soil column. However, from soil sampling data we know that resorption is an important process.
- The model assumes equilibrium partitioning between soil and liquid phase. Equilibrium is a conservative assumption in that it represents the end member of a process that produces the highest groundwater concentration. Model equilibrium would be less likely to be representative of actual conditions in heterogeneous soil (i.e., coarse grain soil where most water movement occurs may have lower concentrations).
- The model also assumes that the source is infinite (does not decline over time). This assumption may not be appropriate for moderate source concentrations.
- The predicted groundwater concentration is directly related to the assumed DF. The DF is the ratio of the soil leachate concentration to the receptor point concentration. The default DF value of 20 is based on an assumed 30-acre source size and accounts for mixing in the aquifer directly beneath the source. It does not account for aquifer attenuation mechanisms in the aquifer downgradient of the source; consideration of aquifer attenuation would result in a larger DF and, consequently, a smaller predicted groundwater concentration. For source areas smaller than 30 acres, a larger DF would be appropriate; for source areas larger than 30 acres, a smaller DF would be appropriate. For example, using the MTCA formula for DF, the same assumptions for other factors included in the formula[17], and varying the size of a square source area, the calculated DF would be 33 for a 10-acre site, 17 for a 40-acre site, and 11 for a 100 acre site. Based on the review of historical aerial photographs performed as part of the Manson Area study (Ecology 2003), it appears that the typical plat size was about 10 to 30 acres so use of an assumed source size of 30 acres may be reasonable. The smelter plume areas are significantly larger than 30 acres, although soil concentrations vary significantly over small distances (i.e., less than 50 ft) (SKCPH and Glass 2000). Smelter plume concentration variability attributed to depositional factors include wind channeling, water-shore air turbulence, slope aspect, and slope steepness (Landau Associates 2003). Variability attributed to post-depositional factors are related to soil disturbance. In agricultural areas, concentration variability is attributed to agricultural spray factors (Veneman et al. 1983)

[17] Other factors included in the formula for DF include groundwater flow volume, hydraulic conductivity, aquifer mixing zone, gradient, volume of water infiltrating, infiltration per year, and unit width.

- The partitioning model assumes a default K_d value that may not be appropriate for site-specific soil conditions. For example, the arsenic K_d of 29 L/kg specified in MTCA is much lower than typical literature values.

Rationale for Use of Alternate Input Parameters

The K_d value (L/mg) is equal to total metal concentration in soil divided by the dissolved metal concentration in groundwater, where total metal is in mg/kg and dissolved metal is in mg/L. The K_d value is used to represent metal mobility and leaching loss from the metal sorbed soil. Experimentally determined values of metal K_d vary over a wide range. Sauve et. al. (2000) compiled literature K_d values for both arsenic and lead. For arsenic, the mean (average) and median (50th percentile) of 66 compiled K_d values were 13,119 L/kg and 1,825 L/kg, respectively. For lead, the mean and median of 204 compiled K_d values were 171,214 L/kg and 102,410 L/kg respectively. Sauve attributed much of the variability in metal K_d values to soil solution pH and identified total metal content and soil organic matter content as two other significant factors that contribute to K_d variability. Other factors, such as soil texture, oxidation state (e.g. arsenic +3 versus arsenic +5), and mineral content also affect K_d.

Recently, the EPA performed an evaluation of metals partition coefficients (Allison and Allison 2005). Their results were similar to the Sauve et. al. (2000) findings. According to the recent EPA study, the mean K_d for arsenic was 1,585 L/kg and the median of 21 compiled values was 2,512 L/kg. For lead, the mean and median of 31 compiled K_d values were 5,012 L/kg and 12,589 L/kg, respectively.

Table 3 presents the minimum, median, mean, and maximum K_d values for arsenic and lead from the two studies. Also presented for comparison are the K_d values specified in MTCA.

TABLE 3
K_d VALUES FOR ARSENIC AND LEAD

	Sauve et. al (2000)	Allison & Allison (2005)	MTCA
Arsenic (L/kg)			29
Minimum	1.60	1.99	
Median	1,830	2,510	
Mean	13,100	1,580	
Maximum	530,000	20,000	
Lead (L/kg)			10,000
Minimum	60.6	5.01	
Median	102,000	15,800	
Mean	171,000	5,010	
Maximum	2,300,000	100,000	

Distribution coefficients can be based on linear sorption isotherms (i.e., the K_d does not change with concentration) or non-linear sorption isotherms (i.e., the K_d is concentration dependant). In practice, a single number is typically used to represent a given contaminant. For example, the MTCA regulations

specify an arsenic K_d of 29 L/kg and a lead K_d of 10,000 L/kg. The arsenic K_d in MTCA is the same as the EPA published numbers for arsenite (i.e., As^{+3}) at a soil pH of 6.8 (EPA 1996). Arsenite is unlikely to be the dominant form of arsenic associated with area-wide contamination. A distribution coefficient based on arsenate (i.e., As^{+5}) would be higher. In contrast to arsenic, the lead K_d used in MTCA (10,000 L/kg) appears to be consistent with the central tendency of literature values presented by the recent EPA study.

Results Using Alternate Input Parameters

If the Kd values are based on median values presented in the EPA study (Allison and Allison 2005), the equilibrium partitioning model would predict the following groundwater concentrations:

- Arsenic soil concentration of 251 mg/kg = groundwater concentration of 5 μg/L (MTCA Method A cleanup level)
- Lead soil concentration of 3,777 mg/kg = groundwater concentration of 15 μg/L (MTCA Method A cleanup level.

CONCLUSIONS

Evaluating the mobility of arsenic and lead relies on an understanding of geochemical relationships as well as an empirical understanding of soil and groundwater occurrence in different settings. Arsenic is expected to occur as an arsenate oxyanion in shallow soil systems and be strongly adsorbed to iron and aluminum oxides. Data from the TSP in Puget Sound and agricultural areas of central Washington consistently determined that the majority of arsenic and lead deposited at the surface over the past century is still sequestered in the upper 6 to 12 inches of soil. Conclusions regarding the mobility of arsenic and lead in the Puget Sound region and in central Washington are summarized below.

Arsenic in Puget Sound Region

In the acid soil of the Puget Sound region, arsenic mobility is expected to be quite limited. Empirical data from the TSP in Puget Sound indicate arsenic soil concentrations of up to 586 mg/kg did not result in enrichment of arsenic below about 2 ft. .

Soil profile data suggest that arsenic has low mobility in Puget Sound soil for moderate concentrations indicative of area-wide contamination. Extensive groundwater databases for wells on Vashon and Maury Island in King County are consistent with a lack of groundwater impact from area-wide arsenic contamination.

Arsenic in Central Washington

In alkaline soil of central Washington, arsenic mobility is expected to be somewhat greater than in the Puget Sound region. Empirical data from former orchard areas indicate arsenic soil enrichment to a depth of more than 4 ft in some samples; however; the majority of arsenic deposited at the surface 60 or more years ago is still sequestered in the upper 6 to 12 inches of soil. Groundwater data for historical orchard areas was limited for central and eastern Washington. The one study completed in 1990 was inconclusive on whether arsenic concentrations in groundwater were elevated due to natural causes or historical lead arsenate pesticide use.

Lead in Puget Sound Region

Lead mobility appears to be less than arsenic mobility. Empirical data indicate lead enrichment to depths of about 18 inches; however, concentrations were lower (relative to background) than arsenic.. Extensive groundwater databases for wells on Vashon and Maury Island in King County are consistent with a lack of groundwater impact from area-wide lead contamination. The low mobility of lead is consistent with literature data on distribution coefficients for lead.

Lead in Central Washington

Lead mobility in central Washington is quite limited, similar to that in the Puget Sound region.

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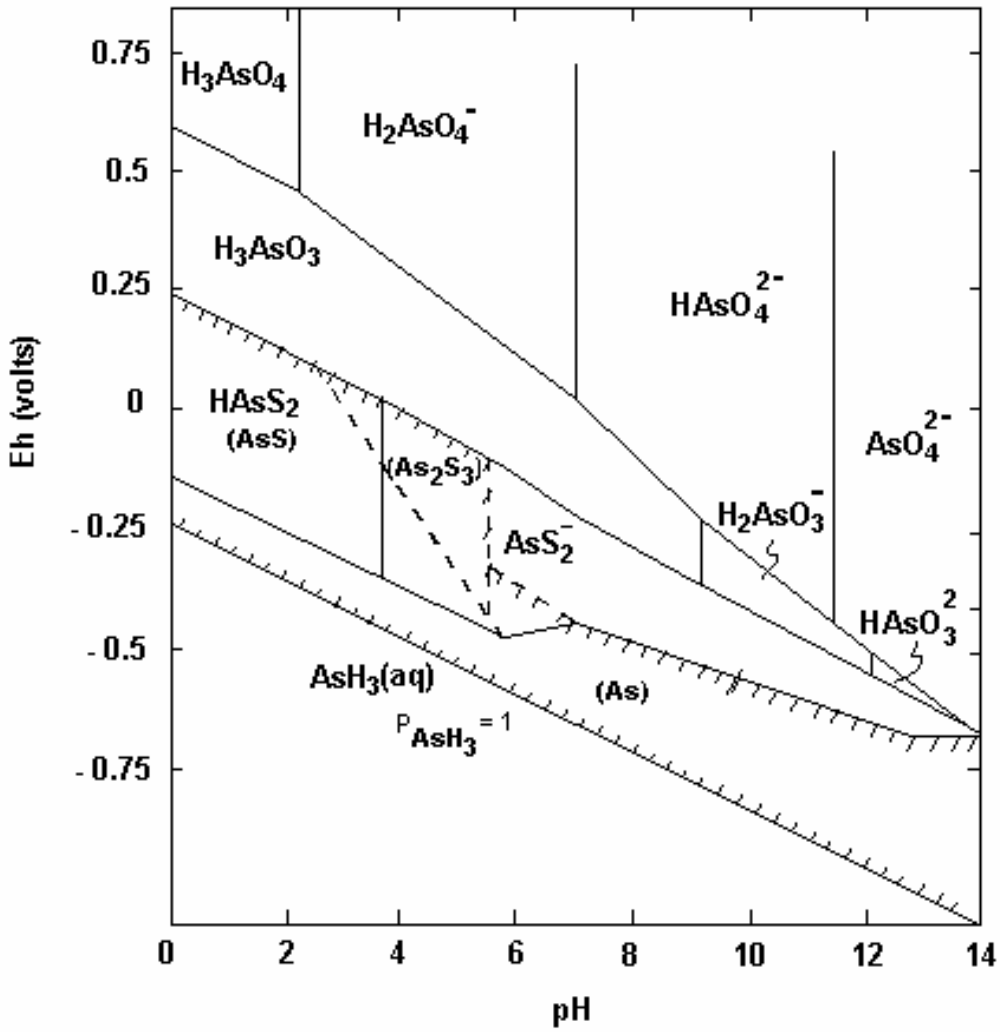

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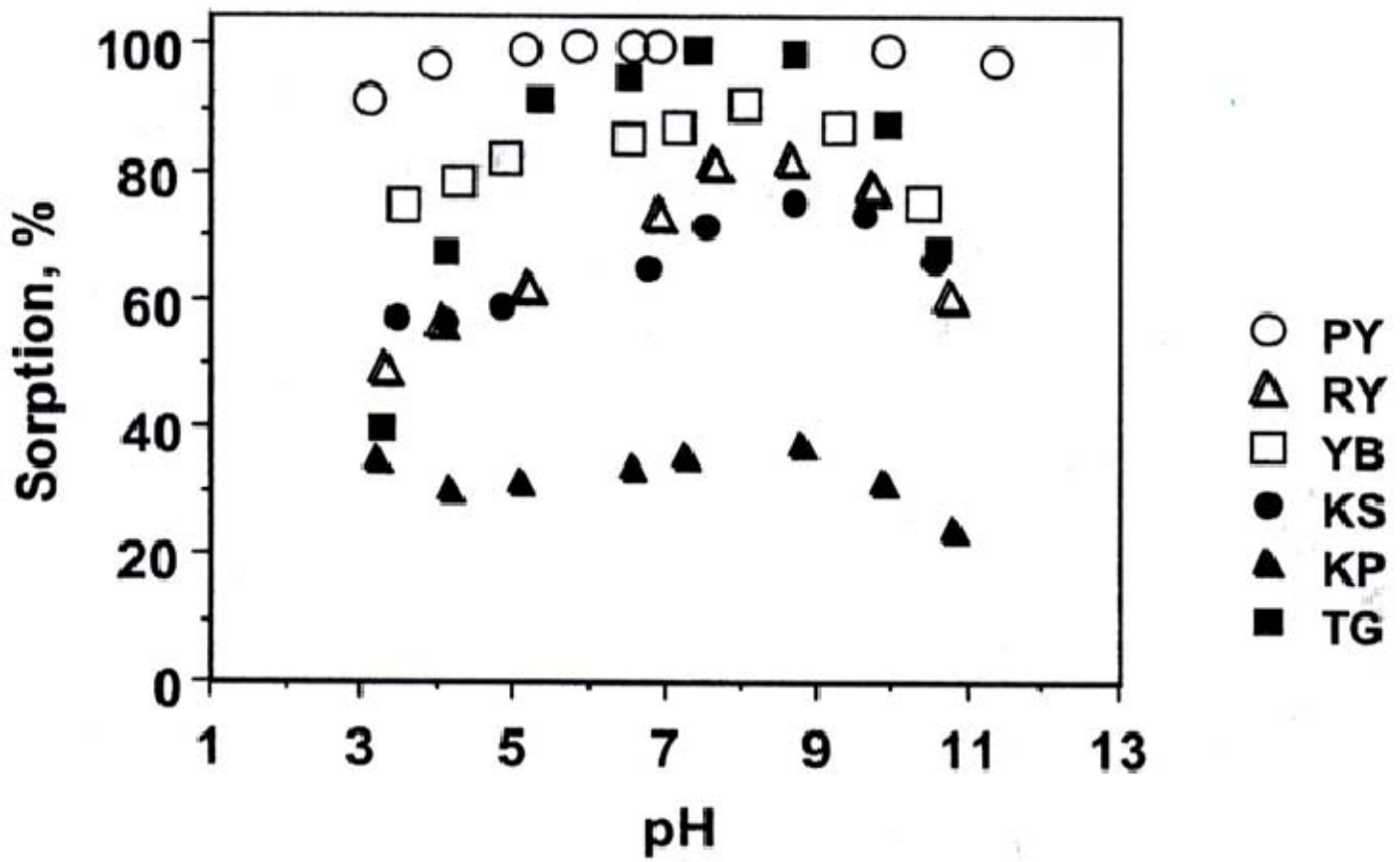
Area-Wide Implementation Project Washington Department of Ecology	**Arsenic-Water System Eh-pH Diagram**	Figure 1



Note

1. See Table 1 for description of Soil Types

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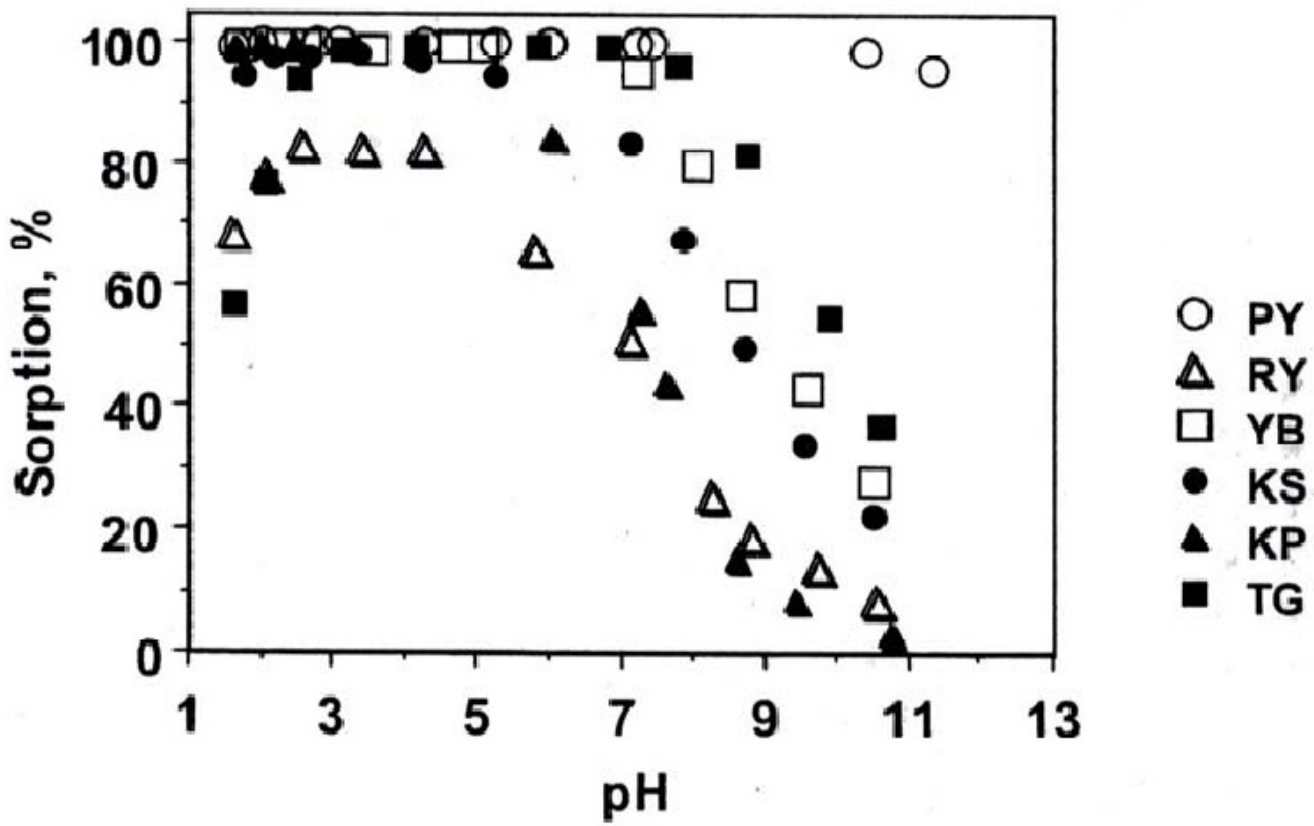
Area-Wide Implementation Project Washington Department of Ecology	**Adsorption of Arsenite on Six Soil Types After Tokunaga 2005**	Figure 2a



Note

1. See Table 1 for description of Soil Types

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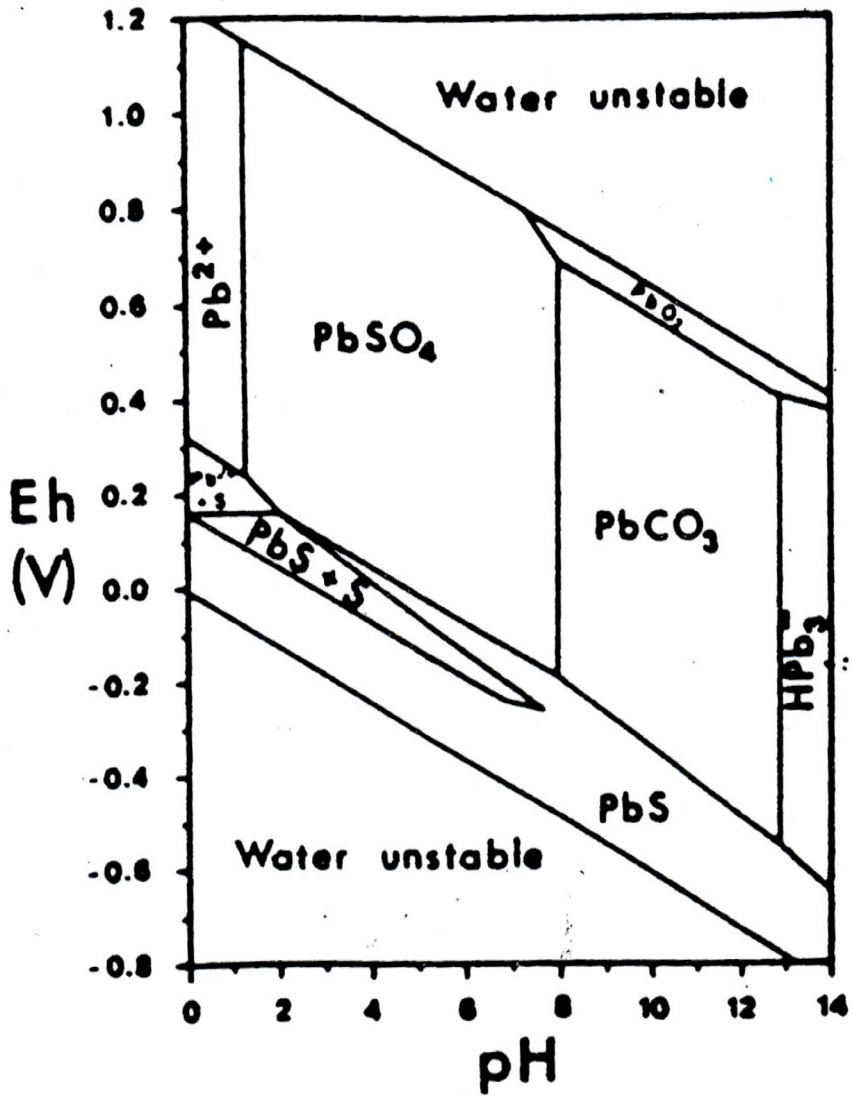
Area-Wide Implementation Project Washington Department of Ecology	**Adsorption of Arsenate on Six Soil Types After Tokunaga 2005**	Figure 2b



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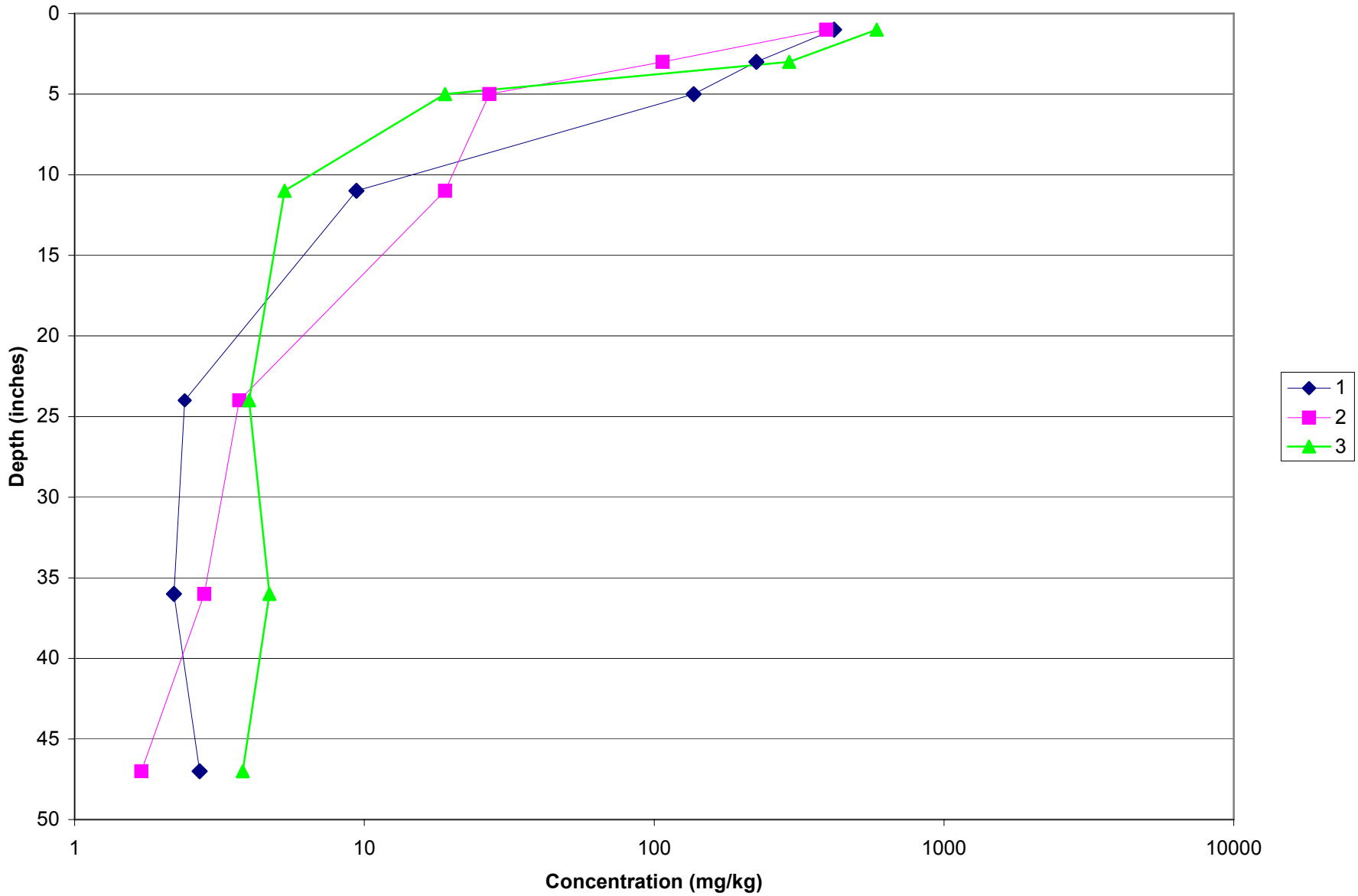
Area-Wide Implementation Project Washington Department of Ecology	**Lead-Water System Eh-pH Diagram**	Figure 3



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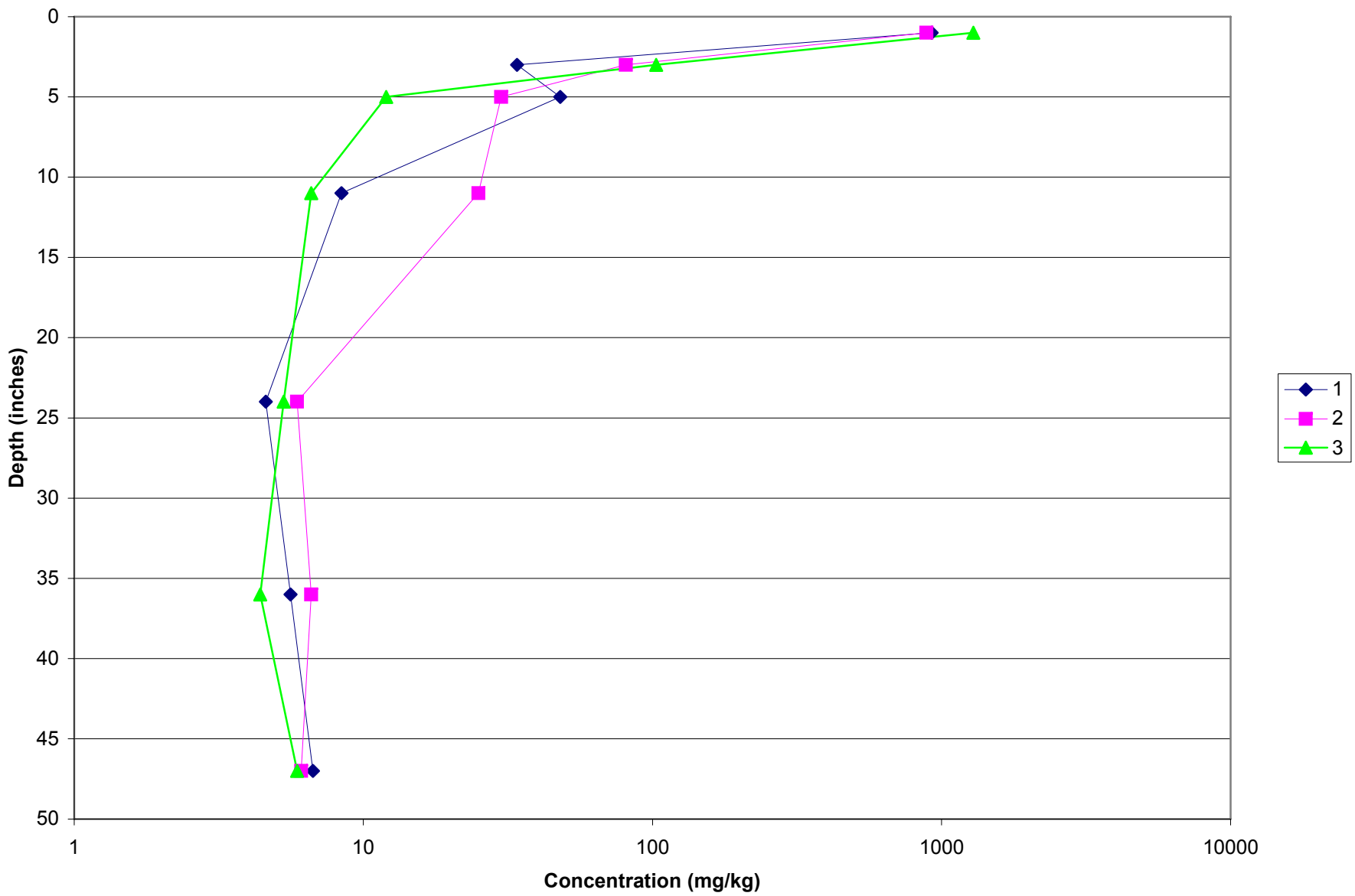
Washington Department of Ecology

Arsenic Concentrations with Depth in Tacoma after Crecelius et al. 1985

Figure 4



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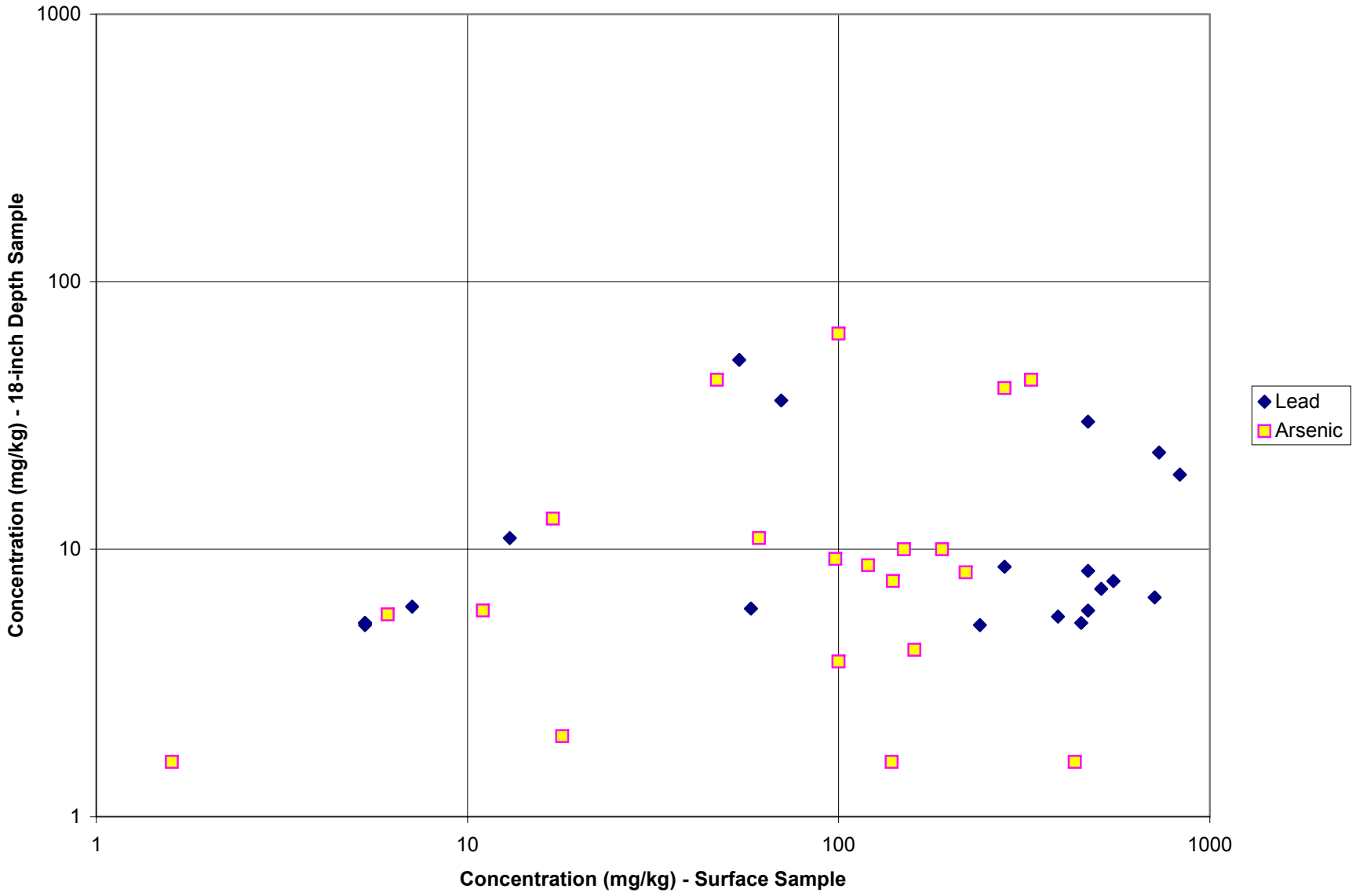
Lead Concentrations with Depth in Tacoma after Crecelius et al. 1985

Figure 5



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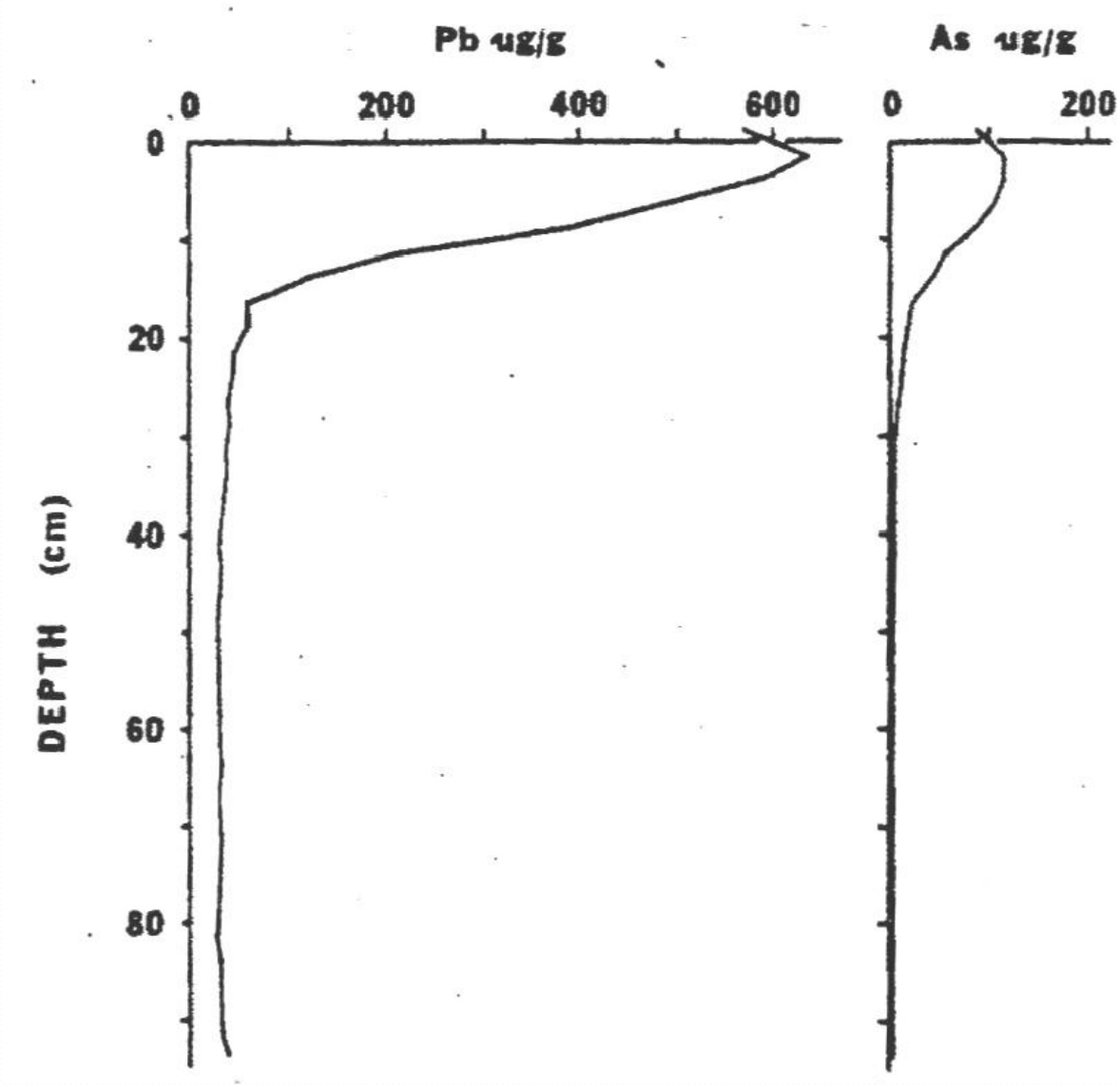
Shallow vs. Deep As Concentrations at the NW Aggregate Mine on Maury Island after Foster Weeler 1999

Figure 6



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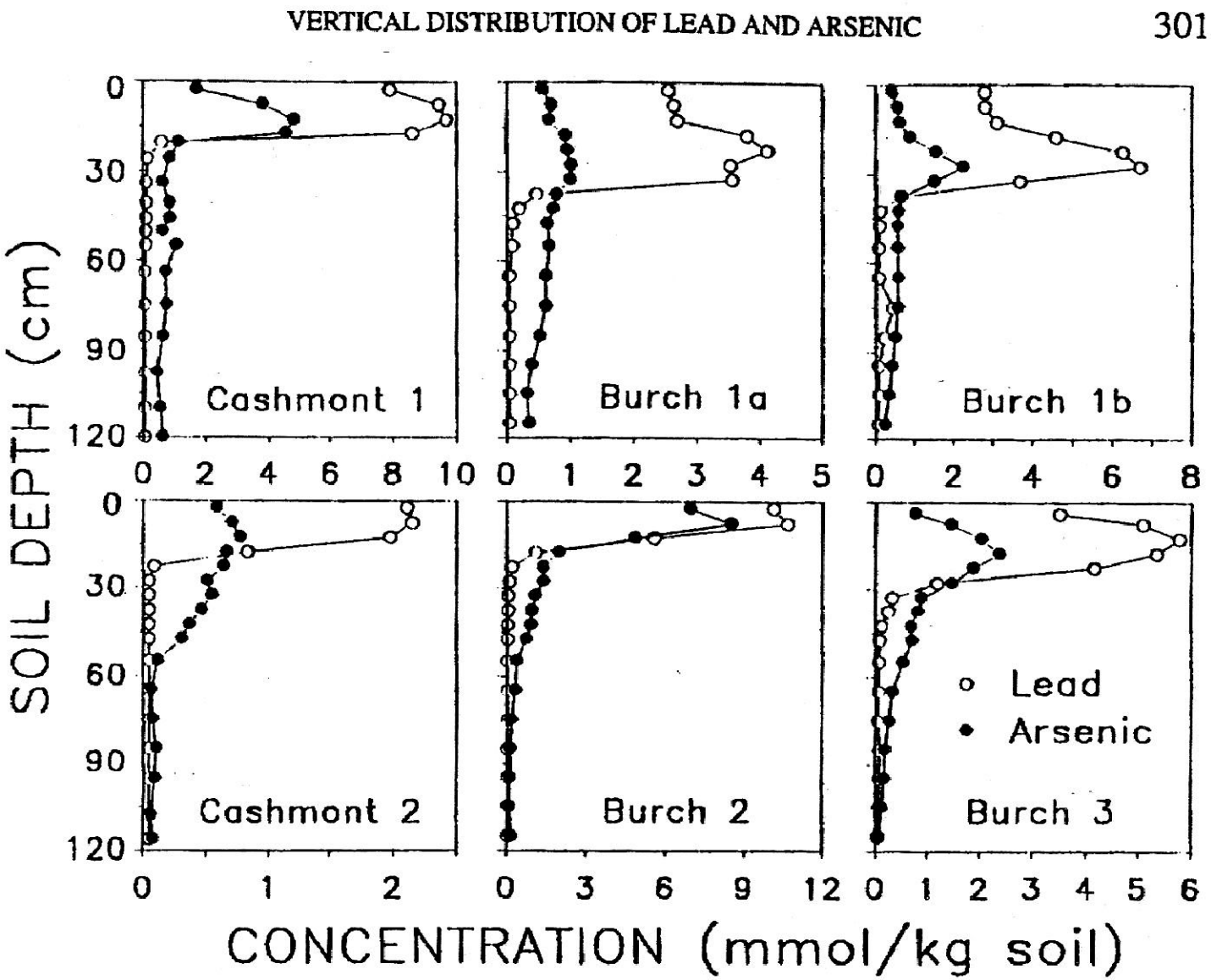
Area-Wide Implementation Project Washington Department of Ecology	**Arsenic and Lead Distribution with Depth after Vereman et al. 1983**	Figure 7



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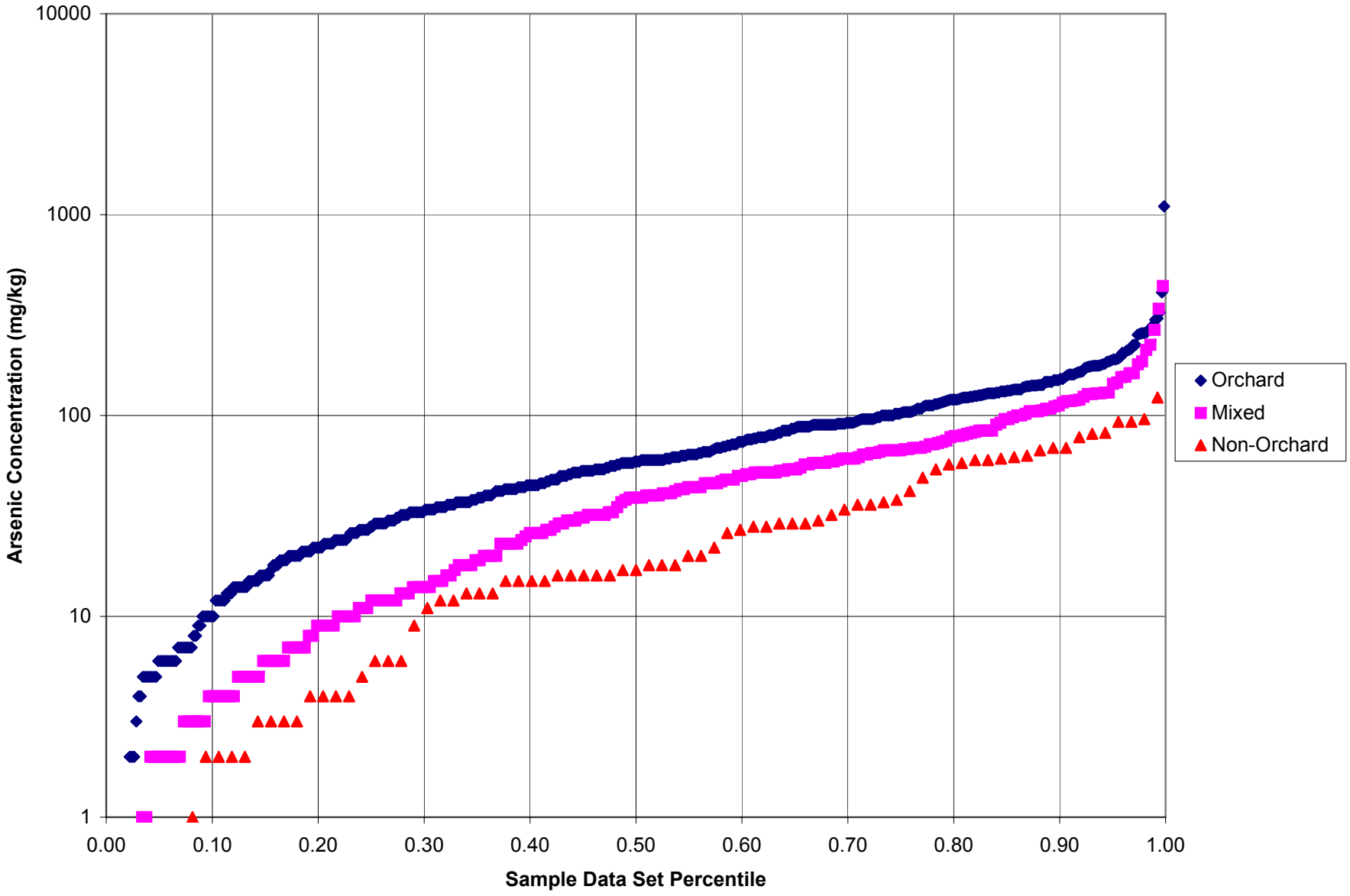
Area-Wide Implementation Project Washington Department of Ecology	**Vertical Distribution As and Pb in Six Contaminated Soils after Peryer and Creger 1994**	Figure 8



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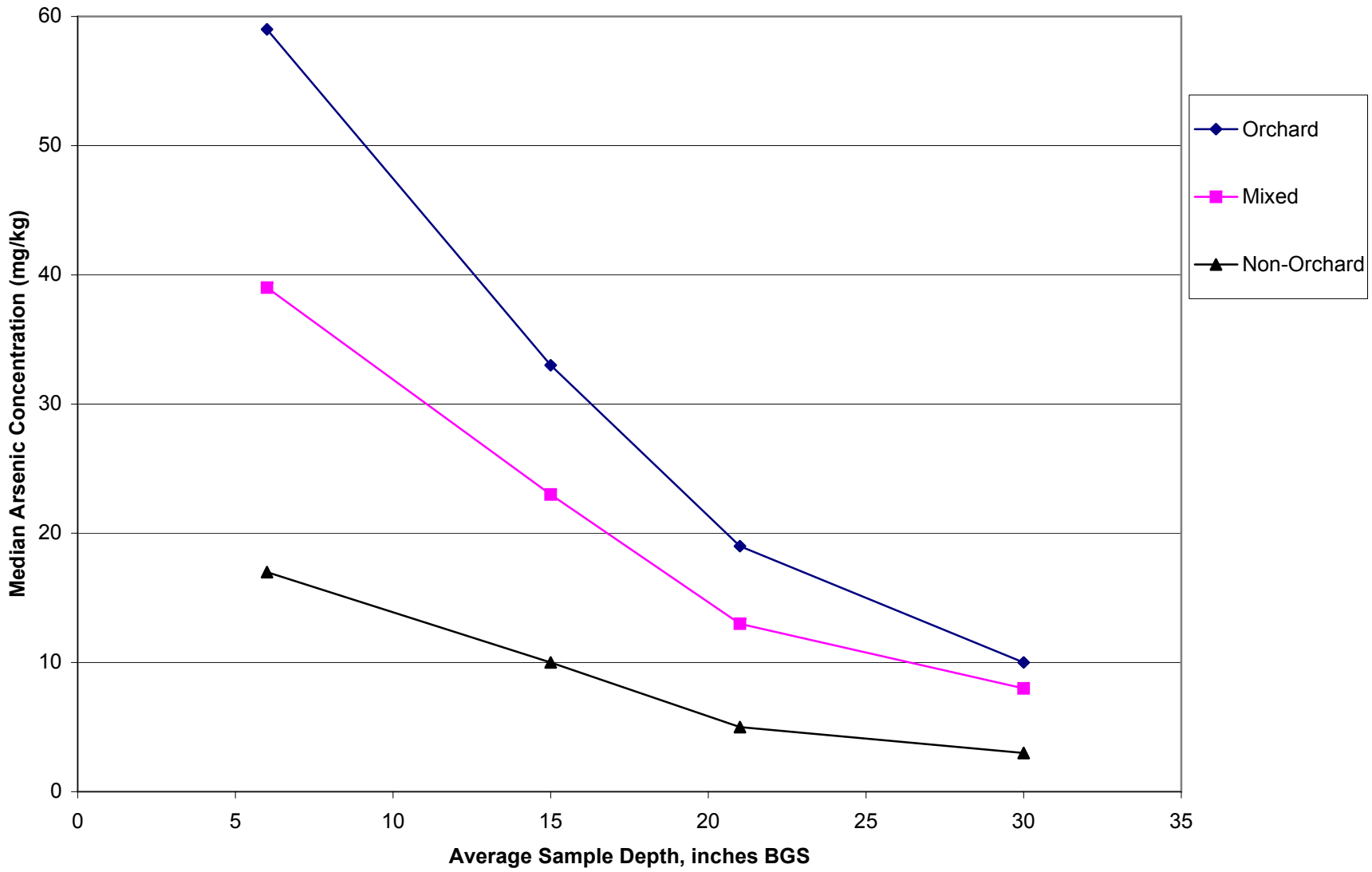
Arsenic Soil Concentraton for 0 to 12 Inch Interval by Land Use Manson Area of Chelan County

Figure 9



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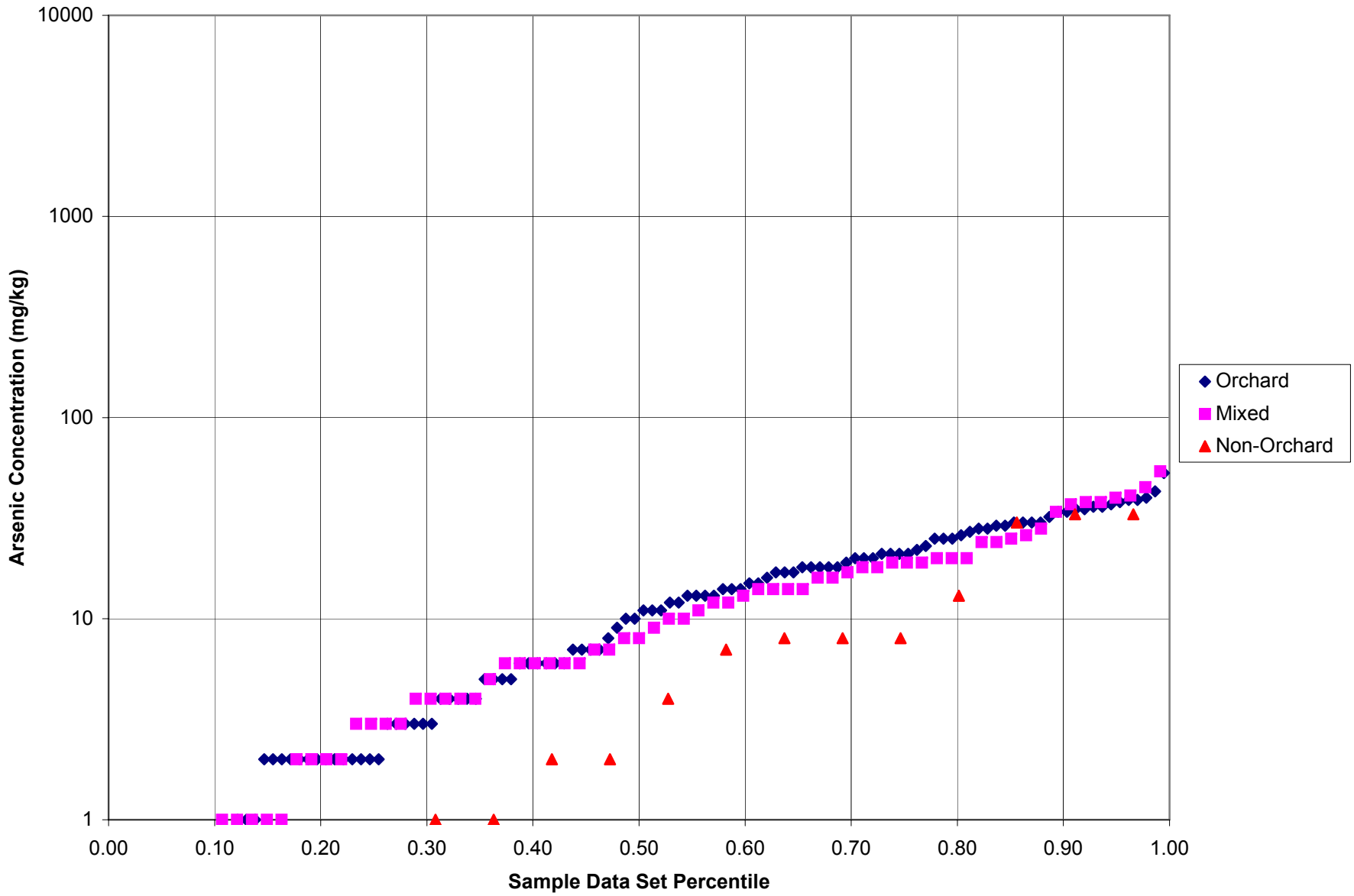
Median Arsenic Soil Concentration Variation with Depth Manson Area of Chelan County

Figure 10



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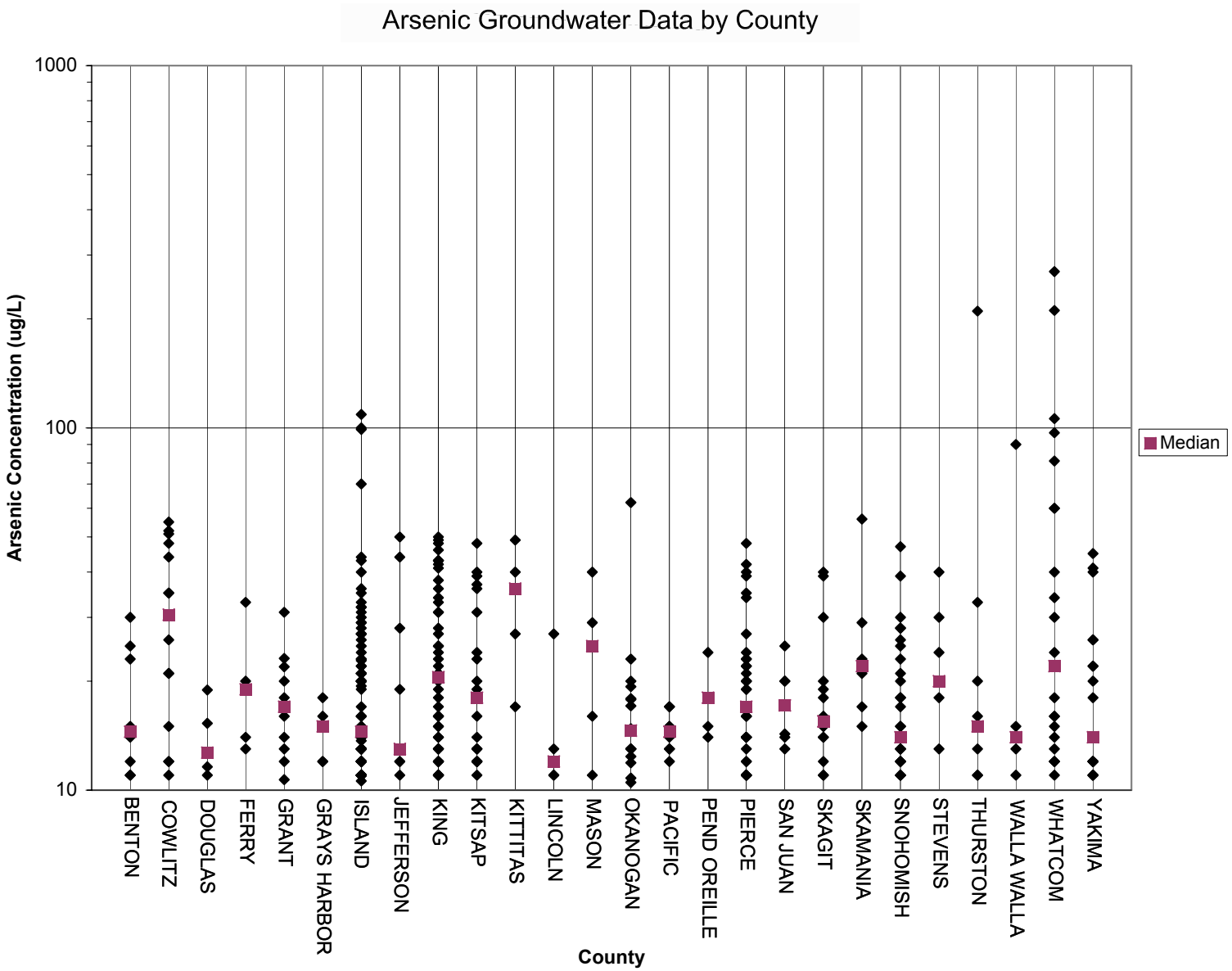
Arsenic Soil Concentration for 24 to 36 Inch Interval by Land Use Manson Area of Chelan County

Figure 11



Note

1. From WDOH database
2. Only shows detections greater than 10 µg/L

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Washington Department of Health
Arsenic Groundwater Data by County

Figure
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