



# Contributions of Pesticide use to Urban Background Concentrations of Arsenic in Denver, Colorado, U.S.A.

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Soil investigations near a former smelter have revealed that historic use of arsenical pesticides has contributed significantly to anthropogenic background concentrations of arsenic on certain residential properties in Denver, Colorado, U.S.A. Remedial investigation data, based on samples collected in relatively undisturbed locations, had previously indicated that the “upper limit” of background arsenic concentrations was 28 mg/kg in the site vicinity. This value compares reasonably well with more regional data, which indicate increasing arsenic concentrations moving from rural to urban land use. Soil sampling during cleanup, however, revealed the presence of arsenic concentrations of a few hundred to more than 1000 mg/kg on a large number of residential lawns due to historic applications of a crabgrass killer, which was missed by the earlier investigation samples because of the sampling bias toward undisturbed land. Data from over 20,000 soil samples now show that several different populations comprise urban background levels of arsenic and that these populations may be stratified by land use and have spatial patterns that should be considered during any background study. A variety of forensic techniques, including spatial analysis, arsenic speciation, and calculation of metals ratios were necessary to separate the smelter impacts from pesticide impacts. © 2001 AEHS

Keywords: anthropogenic; stratified; herbicides; speciation.

## Introduction

The investigation of soil contamination around smelters and other metal refining sites often requires the determination of background concentrations for the metals of concern. Proper evaluation of background values may be critical if used to establish action levels for cleanup, define class action boundaries, or allocate costs for cleanup. Although often (and erroneously) considered to be as simple as a single concentration, the background population that underlies any specific source of contamination, particularly in urban areas, is a complex combination of natural soil, widespread anthropogenic, spatially-correlated anthropogenic, and stratified random anthropogenic populations. A number of anthropogenic sources can contribute to these background populations, including emissions from metal processing and chemical industries, fossil fuel emissions, pesticides and phosphate fertilizers, and wood treatment chemicals (e.g. [Kabata-Pendias and Pendias, 1992](#)). It is essential to recognize the potential existence of each of these sources and background populations and to be able to distinguish their impacts from those caused by the source of concern. Otherwise, soil cleanup programs relying on background values to determine the extent of cleanup may extend well

beyond the actual extent of impact or, in the worse case scenario, never reach a boundary.

Recent investigations of soils surrounding a former smelter site in Denver, CO, U.S.A. were complicated by the presence of a stratified random pesticide population that was initially mistaken for smelter impacts. Initial investigations indicated that the arsenic anthropogenic background concentrations were higher than expected in natural soils, but typically less than 30 mg/kg, consistent with other studies in the Denver area (e.g. [Tourtelot 1973](#)). Sampling during the cleanup program, however, identified the presence of high arsenic concentrations on a number of properties located well beyond the expected extent of smelter impacts. Further investigations identified historic application of an arsenical pesticide as the source of the anomalous arsenic concentrations, using a variety of fingerprinting techniques as described herein. The pesticide population is stratified because it only occurs on residential properties developed prior to the early 1950s; it is also random and widespread. In some cases, these pesticide “background” concentrations were greater than 1000 mg/kg and significantly exceeded the maximum off-site concentrations due to smelter emissions.

Although arsenical pesticides were used extensively in the early part of the 20th century for weed and insect control (e.g. [Gile 1936](#); [Welton and Carroll 1938](#); [Gianessi and Phillips, 1998](#)) and the potential for arsenical pesticides to impact soils has been known for

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some time (e.g. McMurtrey and Robinson, 1938), most, if not all, case histories in the literature have been limited to fruit orchards, cotton fields, or commercial facilities that produced, stored, or disposed of pesticides. The authors are not aware of any other cases in the literature where application of arsenical pesticides on residential properties has resulted in such high and widespread impacts as the case reported herein.

This paper describes the investigations and methods initially used to establish the widespread anthropogenic arsenic background population, reasons why the initial sampling missed the additional stratified random pesticide population, the nature of the pesticide background population, and methods used to distinguish pesticide impacts from those due to the smelter emissions.

## Site Description

The ASARCO Incorporated (Asarco) Globe Plant was originally built in a rural area north of Denver in 1886. Today, the site is located within an urban area of the city along the South Platte River Valley, a major transportation and industrial corridor with a mixture of residential, commercial, and industrial land use (Figure 1). Soils in the site vicinity are neutral to alkaline sandy to clayey loams with organic concentrations typically exceeding 1% in the surface horizons and clay accumulations in the B-horizons (SCS, 1974). The climate is semi-arid with a mean annual precipitation of 13.8 inches and a mean annual temperature of 52°F (TRC, 1988).

The Globe Plant smelted ore to produce lead bullion, silver, and gold until 1919, when lead-smelting operations ceased. From approximately 1912 until 1927, the plant produced arsenic trioxide by refining bag house dust from other lead smelters. From 1927

until 1993, cadmium refining was the principal operation at the Globe Plant. Since 1993, only relatively small scale, high-purity metal refining has been conducted at the plant.

The State of Colorado sued Asarco in 1983 under the Natural Resource Damages (NRD) provision of the Comprehensive Environmental Reclamation, Cleanup, and Liability Act of 1980 (CERCLA). In an effort to settle the lawsuit, a Remedial Investigation and Feasibility Study (RIFS) was carried out between 1985 and 1990 to identify, among other things, the nature and extent of soil contamination in the surrounding community due to historic emissions of arsenic, cadmium, lead, and zinc from the facility (TRC 1988, 1990). Soil cleanup or action levels for each metal were set by the State of Colorado (CDH 1993) based on risk assessment calculations (Putnam, 1989). The primary action level for arsenic is 70 mg/kg, while a voluntary or secondary arsenic action level was set equal to the “upper limit of background” for arsenic, or 28 mg/kg. The primary action levels for cadmium and lead are 73 mg/kg and 500 mg/kg, respectively.

The community soil cleanup program was subsequently defined by a Statement of Work (SOW), part of a Consent Decree entered by the U.S. District Court on 15 July 1993 that settled the NRD lawsuit. The SOW requires sampling of every property surrounding the Globe Plant in each direction until a “clean block” is reached. A provision in the SOW allows Asarco to petition the State to not clean up properties that were contaminated by other sources not related to the Globe Plant. As a result, proper delineation of background and the identification of other sources is critical. Yard-by-yard soil sampling began in late 1993, as described below.

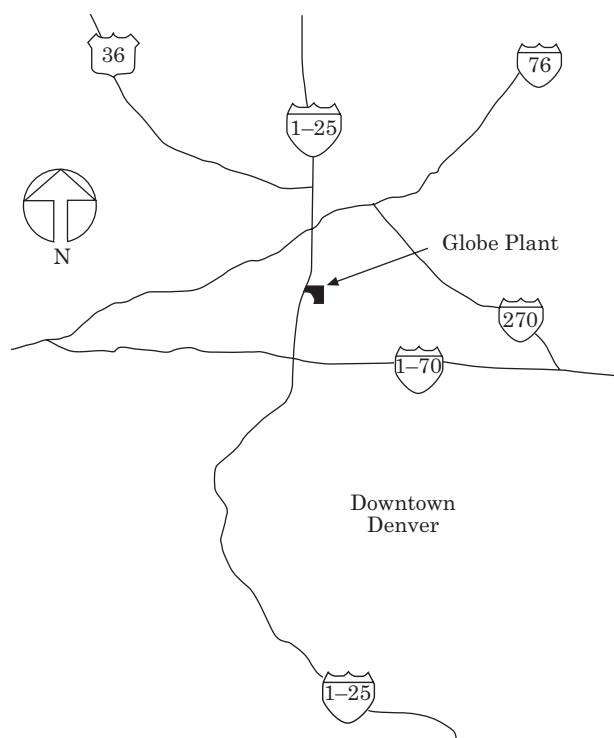


Figure 1. Site location.

## Investigation Procedures

### RIFS Investigation

RIFS soil samples were collected at approximately 150 locations within 2 miles of the Globe Plant over 0–5 cm, 5–20 cm, and occasionally 15–30 cm depth intervals, based on a polar grid system that resulted in a greater sample density near the plant (Figure 2). The sampling program was deliberately biased or stratified to select sites with the least amount of apparent disturbance (e.g. vacant lots) to maximize the likelihood that metals due to deposition of historic air emissions would still be present and not removed or obscured by recent development or landscaping activity (TRC, 1988). Therefore, established lawns and other landscaped areas were not sampled during the RIFS. Discrete samples were collected at each site from random locations within a 8 × 8 m area judged to be relatively undisturbed. The <math>-2</math> mm fractions of all soil samples were analysed for arsenic, cadmium, lead, and zinc concentrations. Arsenic analyses were conducted by EPA Methods 206.1 and 206.2, with a detection limit of 0.5 mg/kg. More detailed information is provided in TRC (1988, 1990).



Figure 2. RIFS sample locations.

### *SOW Investigation*

To date, over 20,000 samples have been collected at residential and commercial properties over an area of approximately 5 km<sup>2</sup> during the SOW cleanup program (Figure 3). Properties were divided into at least four sectors not exceeding 2500 ft<sup>2</sup> (232 m<sup>2</sup>) in size. Core samples were collected at four random locations within each sector over the 0–5 cm and 0–15 cm depth intervals and composited to create one sample for laboratory analysis for each depth interval in each sector. The samples were air dried and sieved through a 10 mesh screen (–2 mm fraction) prior to aliquoting for digestion by SW-846 Method 3050B. Samples were then analysed for arsenic, cadmium, lead and zinc by either inductively coupled plasma–atomic emission spectrometry (ICP) by EPA Method 6010A (6010B after 1998) or laboratory X-Ray Fluorescence (XRF), with arsenic detection limits of 6 and 10 mg/kg, respectively, following standard EPA method

laboratory quality control procedures. During the time period the XRF was used, one in 20 samples were split and also analysed by ICP, with ICP arsenic concentrations generally being about 5–10% lower than XRF concentrations. This difference in the two techniques should not affect any of the observations or conclusions presented herein, which are based on very large concentration differences between properties. The results of all tests are available in the Design Investigation Reports (DIRs) for the project submitted by Asarco every year to the Colorado Department of Public Health and Environment (EnviroGroup, 1994 *et seq.*)

### **Results and Discussion**

The results of the RIFS investigations indicated that arsenic concentrations decreased from levels of approximately 100–200 mg/kg near the Globe Plant

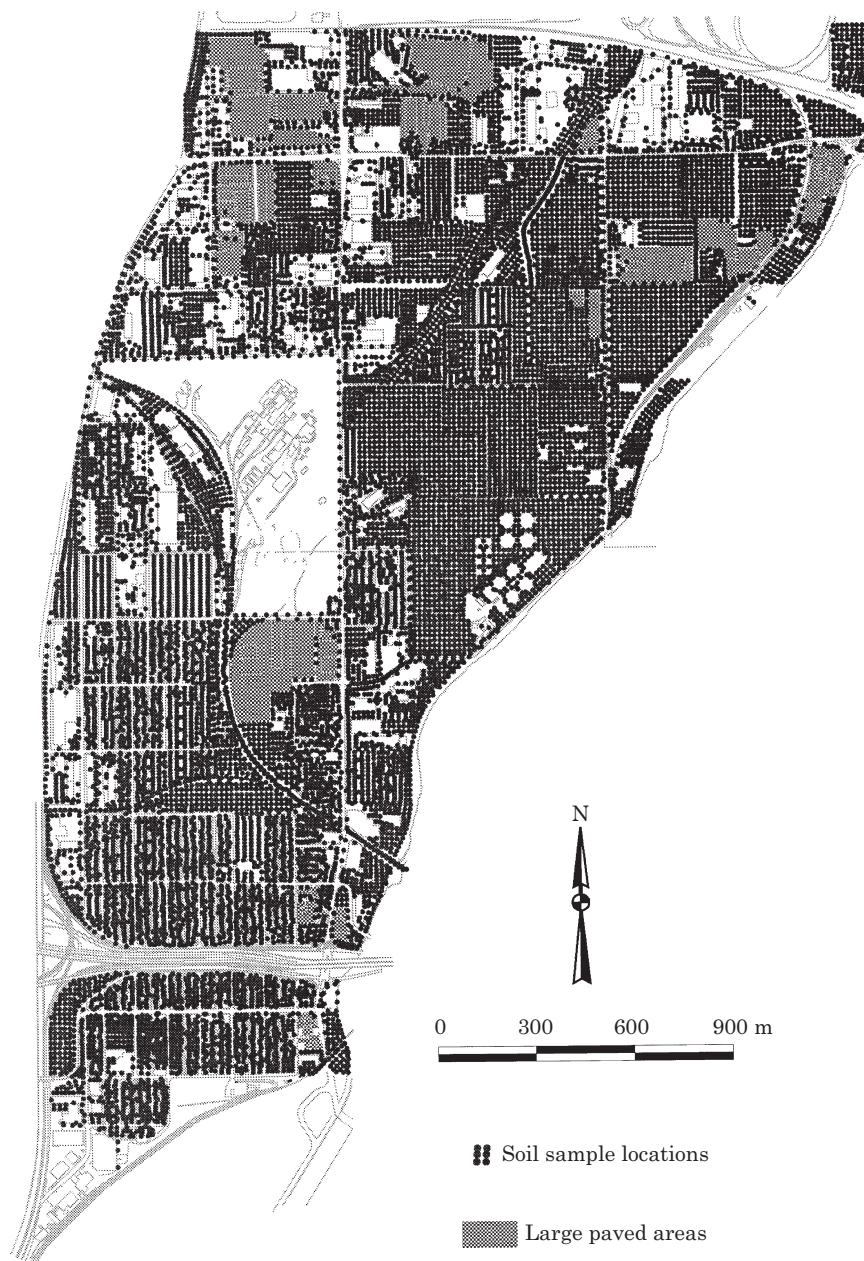


Figure 3. SOW sample locations.

to levels typically 20 mg/kg or less within 2000 m of the center of the site (Figure 4). Similar findings were expected at the start of the SOW investigations. Arsenic concentrations, however, continued to remain elevated during the SOW investigations and even increased with distance, as shown on Figure 5. In fact, arsenic concentrations were much higher at properties more than 1000 m from the Globe Plant than found near the Globe Plant during either the RIFS or SOW investigations, exceeding 1000 mg/kg on several properties (Figure 6).

The following sections of the paper describe the evaluation of the widespread anthropogenic arsenic background population in the Globe Plant area based on the RIFS data, and the discovery and characterization of the pesticide background population during the SOW investigations.

#### *Widespread Anthropogenic Background*

Two approaches are typically available for evaluating background populations. The first and most common approach is to collect samples from areas that are beyond the impact of the source, usually at upwind and remote locations (e.g. Breckenridge and Crockett, 1995). However, because the wind usually blows in all directions at least part of the time and the impacts of airborne emissions decrease asymptotically with distance, it can be difficult to determine how far you have to go to reach background conditions. Remote locations are also problematic because soil type and other anthropogenic sources of arsenic may be different from the source area, resulting in a non-representative background population. In the case of the Globe Plant, sufficient data were collected to surmise that reasonable background conditions might be encountered beyond

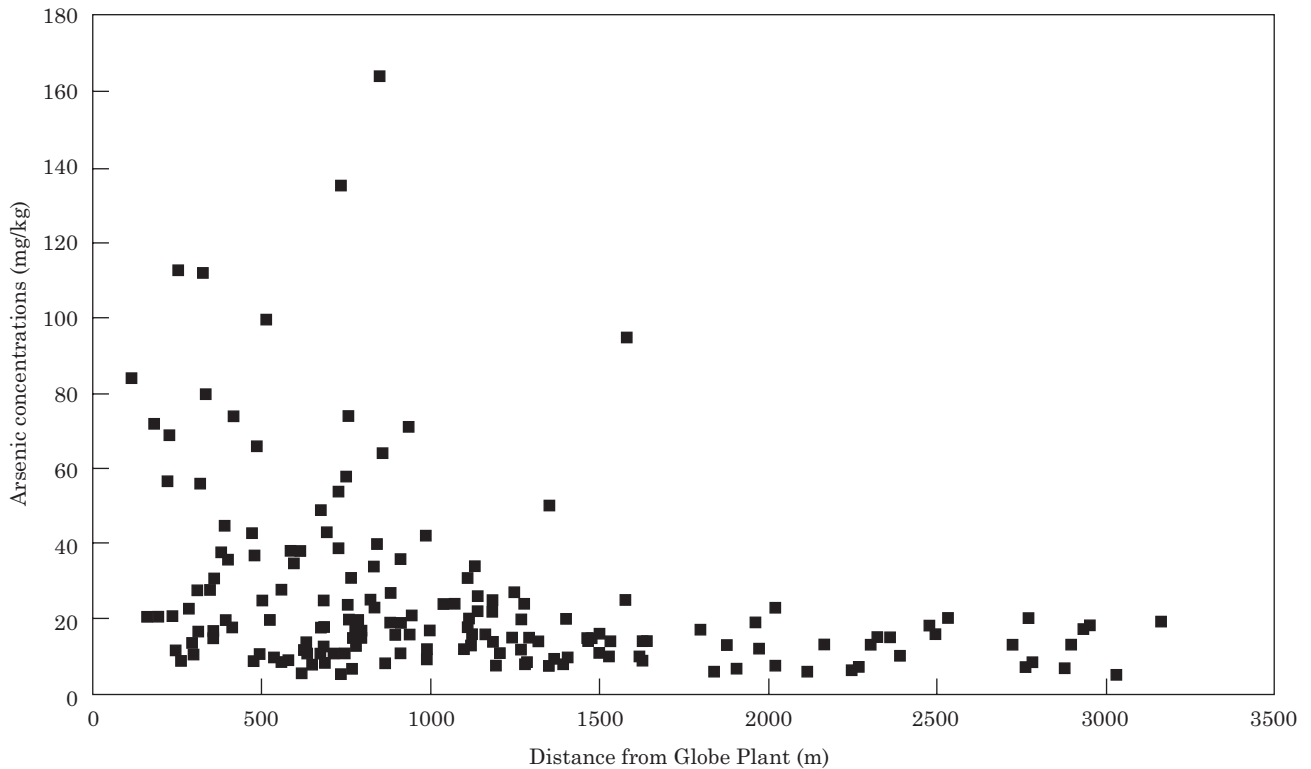


Figure 4. RIFS arsenic concentrations v. distance.

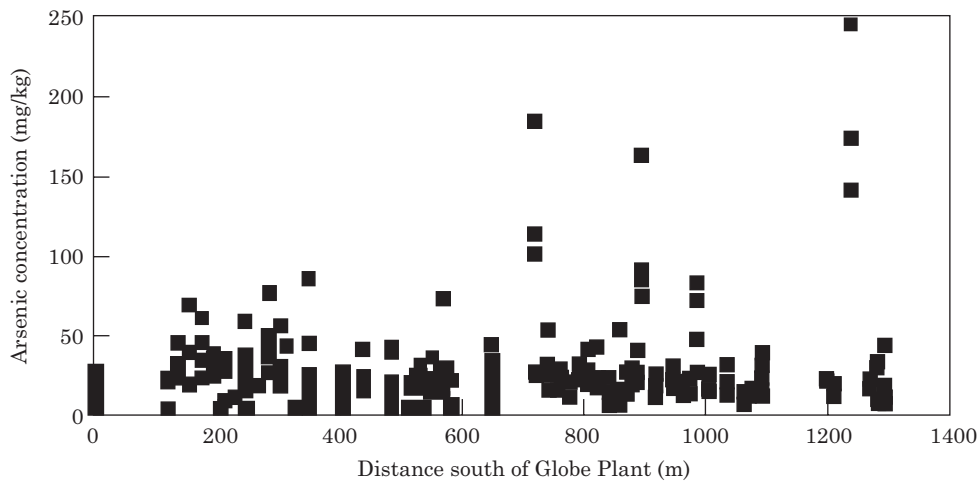


Figure 5. SOW arsenic concentrations v. distance.

one mile from the plant (Figure 4), although some Globe Plant contributions might still be present.

The second, less common approach for determining background is to statistically analyse all of the concentration data and separate the background from the contaminated population. Rose, Hawkes and Webb (1979) suggest two methods for doing this, including plotting the logarithms of the concentrations in a histogram and graphically identifying low points or saddles in the distributions that might represent a reasonable threshold between the two populations. The second method involves preparing a probability plot and identifying inflection points in the data curves, after the procedures described by Sinclair (1976). Data separation techniques have the advantage of using background values from the site area, which are more

likely to reflect natural soil and local anthropogenic background sources than remote samples. However, the selection of threshold concentrations that separate the populations is somewhat subjective, particularly if the background and contaminated soil populations overlap significantly.

Because of the uncertainty inherent in any background determination, both of the approaches described above were used to evaluate the widespread, anthropogenic background population in the Globe Plant vicinity. First, summary statistics were calculated using all of the samples collected beyond 1 mile from the center of the Globe Plant during the RIFS. Based on the relatively linear nature of the data when plotted on a log scale probability plot (and excluding the three highest values which are skewed from the line), this

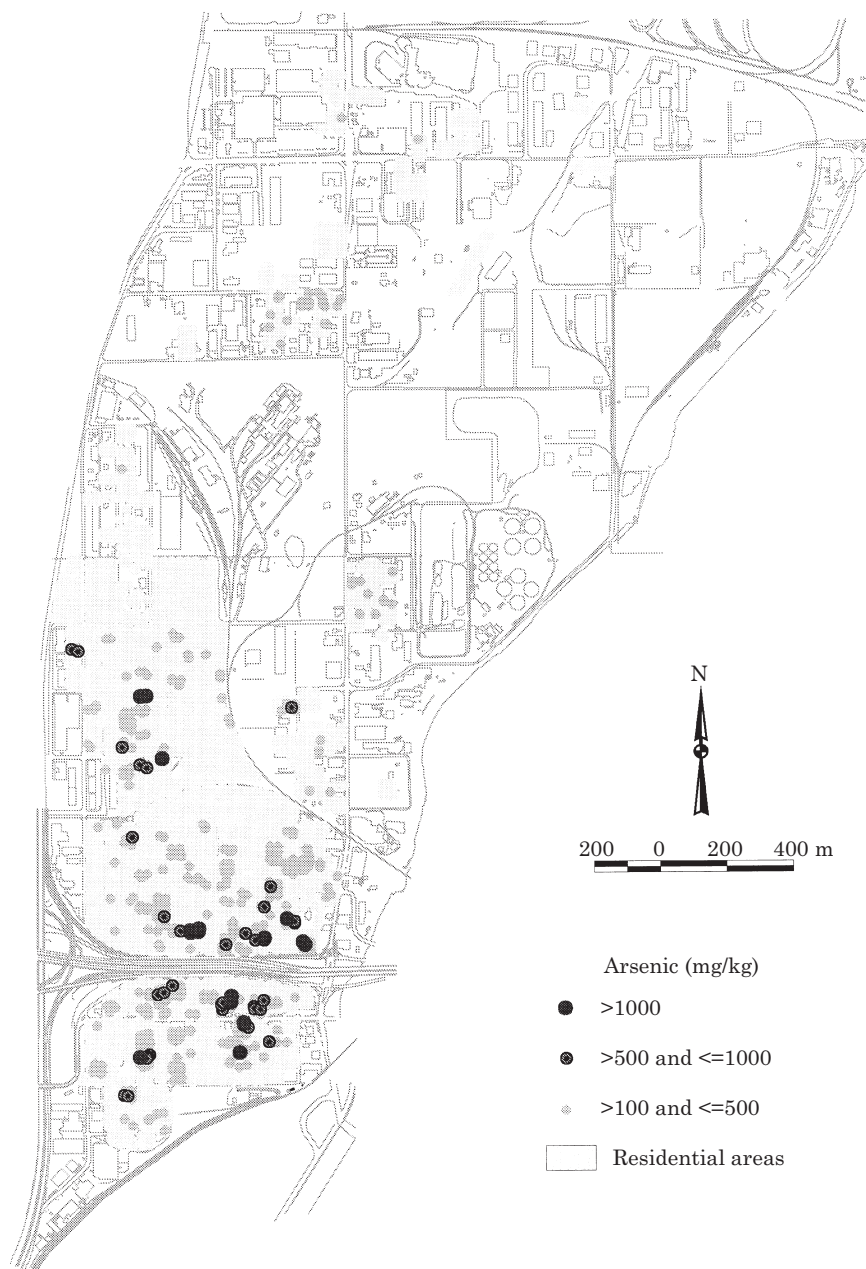


Figure 6. Arsenic concentrations on residential properties.

“background” population was considered to be log-normal in distribution, with a geometric mean and geometric standard deviation (SD) of 12.6 and 1.6 mg/kg, respectively (Table 1). Rose Hawkes and Webb (1979) and Breckenridge and Crockett (1995) suggest using the mean plus two or three SD, respectively, as a threshold concentration to determine whether individual sample results are from the background or contaminated soil populations; i.e. threshold values of 20 mg/kg and 32 mg/kg, respectively. Including the three highest values, which are between 1 and 5 miles from the Globe Plant and, in at least one case, in a predominant upwind direction, the geometric mean and geometric SD values are 13.3 mg/kg and 1.7, respectively, with upper two and three SD concentrations of 23 and 38 mg/kg, respectively (Table 1).

The upper limit of background for arsenic in the Globe Plant vicinity was also estimated by plotting all off-site RIFS data on a log histogram plot (Figure 7)

and graphically separating the background and contaminant populations, according to the method described by Rose, Hawkes and Webb (1979). The saddle between the two populations at 28 mg/kg is not distinct and arguably a second saddle occurs at about 67 mg/kg. However, the 28 mg/kg value appears consistent with the distance trend in Figure 4, and the evaluation of the RIFS data collected beyond one mile, as discussed above. The impact of the polar grid (varying data density with distance from the Globe Plant) on the log histogram method is not clear, although it greatly increases the proportion of the data representing the contaminated population. Based on the evaluations described above, an upper limit for arsenic background of 28 mg/kg was selected for the cleanup program (CDH 1993).

The authors also evaluated the widespread anthropogenic background using the RIFS data and the method of Sinclair (1976), as shown on Figure 8. An

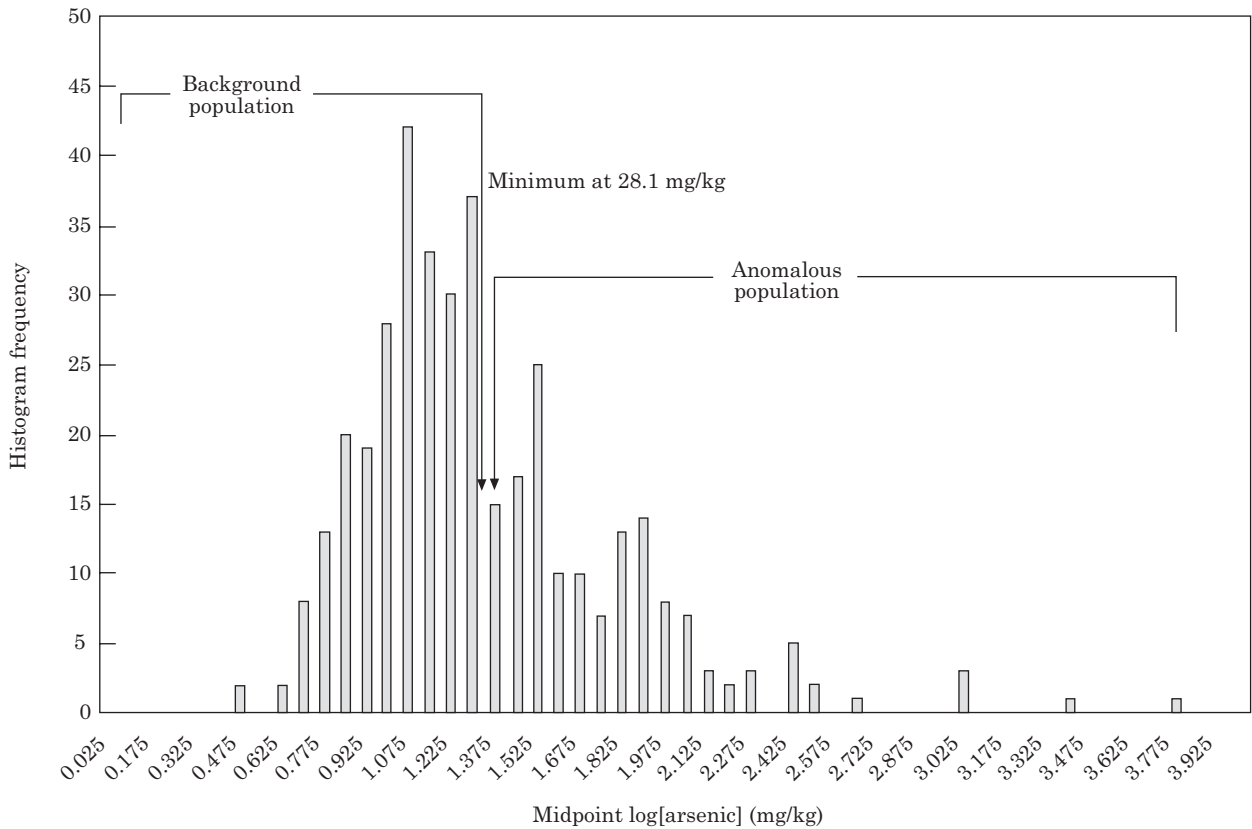


Figure 7. Log histogram of RIFS arsenic data.

Table 1. Estimates of widespread, anthropogenic arsenic background concentrations in surface soils

Data*	Location	Land Use†	Method‡	Geo Mean (mg/kg)	Geo SD	MEAN + 2 SD (mg/kg)§	Mean + 3 SD (mg/kg)	Ref¶
USGS	Western US	Varies	SS	6.1	1.82	20	37	A
USGS	Longmont, CO vicinity	Rural	SS	5.4	2.2	12	26	A
USGS	Denver	Rural	SS	3.7	2.0	15	30	B
USGS	Denver	Suburban	SS	5.7	2.0	23	46	B
USGS	Denver	Urban	SS	6.8	2.5	42	106	B
RIFS	> 1 mile from Globe	Mixed	SS(-3)¶¶	12.6	1.6	20	32	C
RIFS	> 1 mile from Globe	Mixed	SS	13.3	1.7	23	38	C
RIFS	All off-site data	Urban	HIST	na	na	28	na	D
RIFS	All off-site data	Urban	PROB	13.6	1.55	33	51	C
SOW	> 1 mile from Globe	Mixed	PROB	10.6	2.11	47	100	C

\*USGA—US Geological Survey (Tourtelot, 1973); RIFS—Remedial Investigation Feasibility Study (TRC, 1988, 1990, from relatively undisturbed sample locations; SOW—Statement of Work investigations, Globe Plant cleanup, (see EnviroGroup Limited DIR reports beginning 1994)

†Predominant land use indicated

‡SS—summary statistics on entire data set, HIST—log histogram separation, PROB—log probability separation

§The geometric mean is multiplied by the geometric standard deviation

¶A—Connor and Shacklette (1975), B—Tourtelot (1973) data as analyzed herein, C—TRC, (1988, 1990) data as analyzed herein, D—TRC (1988), E—EnviroGroup (1994) *et seq.* Design Investigation Report data, as analysed herein

¶¶Three highest data points excluded because of non-linear fit with population

inflection point was interpreted at approximately 30 mg/kg, resulting in a background population with a geometric mean concentration of about 14 mg/kg and a mean plus two standard deviations of about 33 mg/kg. Once again, these data compare well with the summary statistics calculated by the other methods presented above (Table 1).

As a further check on the validity of these background characterizations, the authors reviewed available data. In 1973, the USGS collected 780 random surficial soil samples at 195 “localities” (each 4 km<sup>2</sup> in

size) in a grid pattern at 16 km centers, from approximately Ft. Collins to Colorado Springs, including the Denver metropolitan area (Tourtelot 1973, Tourtelot and Neiman, 1974).

Nine of the 195 localities sampled by Tourtelot (four samples per locality) were located in urban areas of Denver, while an additional 15 localities were located in suburban areas of Denver. The distinction between urban, suburban, and rural areas was subjective, made by the authors based on the amount and age of development. Urban areas are generally older

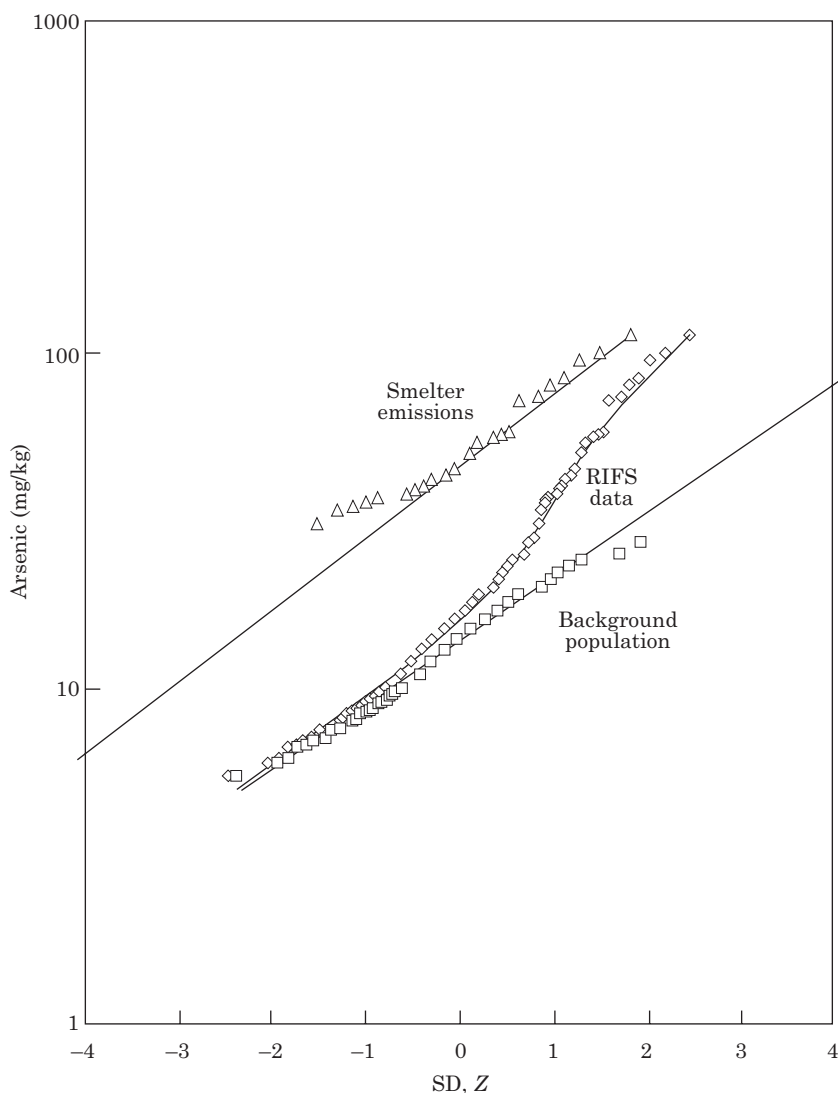


Figure 8. Log probability plot of RIFS arsenic data.

industrial, commercial, and inner city residential neighborhoods, while suburban areas are generally residential, commercial and light industrial areas that were developed after World War II. Rural areas are generally ranch and agricultural land with isolated residential and commercial/industrial properties. Most of the Front Range Urban Corridor that was sampled by Tourtelot is rural; for the purposes of this study, rural sampling localities within 32 km (i.e. two sampling localities) of the furthest suburban or urban locality were evaluated.

Geometric mean and standard deviations of arsenic concentrations in urban, suburban, and rural localities in the metropolitan Denver area are shown in Table 1, based on the Tourtelot data. The geometric mean increases from 3.7 mg/kg in the rural areas to 6.8 mg/kg in the urban area, indicating that human activities have a small effect on the mean arsenic background concentration. The increase in SD is more significant, from 2.0 in the rural and suburban areas to 2.5 in the urban area. This suggests that significant variances from the mean occur, particularly in the urban area, although not frequently enough to have a large impact on the mean concentration. The resultant mean plus

2 SD (the geometric mean multiplied by the geometric SD twice) concentration is about 42 mg/kg for the urban background population, compared to 15 mg/kg for the rural background population.

The increasing trend in arsenic concentrations moving from rural to urban settings is shown by data from east-west transects running through Denver, plotted v. distance from the downtown area on Figure 9. It should be noted that none of the sample localities (i.e. 16 km<sup>2</sup> areas within which samples were collected) were within 2 km of the Globe Plant, and that the locality with the highest arsenic concentrations was over 6 km from the Globe Plant.

The various interpretations of the widespread, anthropogenic background arsenic population in the urban Denver area based on evaluation of the RIFS data compare well with the Tourtelot (1973) data, as shown in Table 1. While none of the methods used to estimate background population characteristics is without bias, the general agreement between the methods is encouraging, and suggests that a threshold or upper limit value for the widespread anthropogenic background population in the range of 30 mg/kg is reasonable.

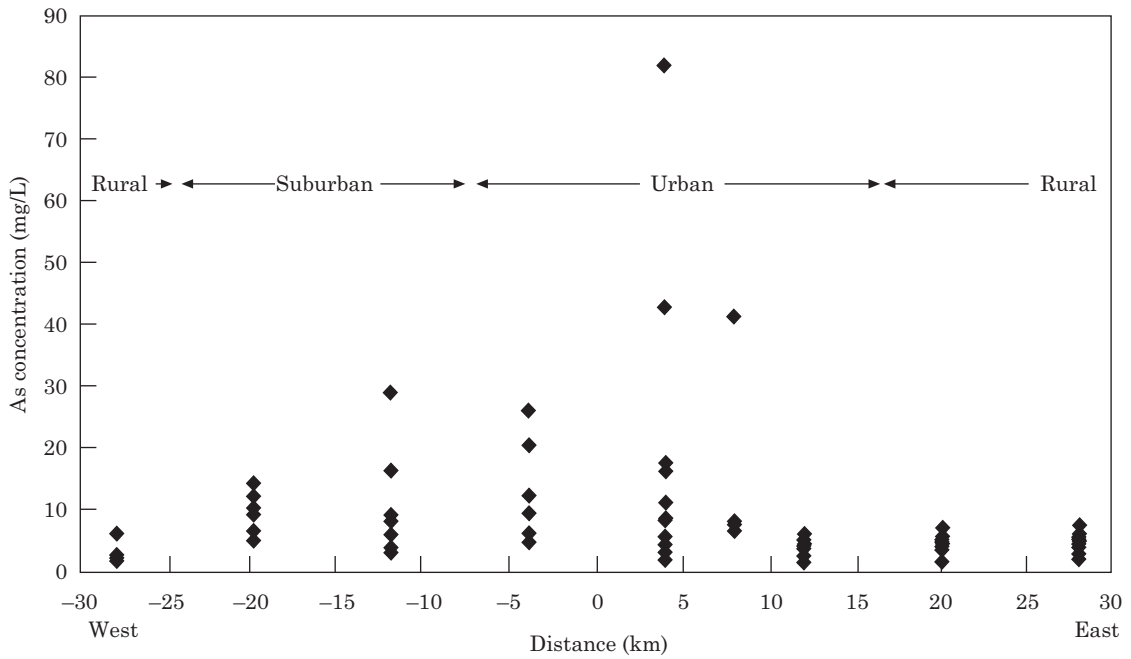


Figure 9. Arsenic concentrations across Denver (based on Tourtelot, 1973).

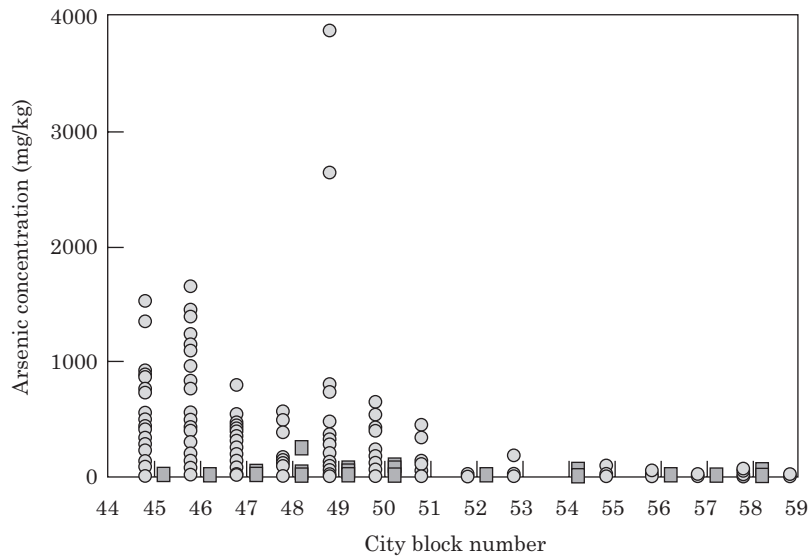


Figure 10. Arsenic concentrations v. land use. Developed residential (○); undeveloped (■).

*Pesticide Background Population*

The impacts of smelter emissions on soil generally decrease rapidly with distance from the site because of dilution caused by the geometrically expanding air volume, plume dispersion due to air turbulence, and plume depletion due to fallout, as observed at numerous smelter sites reported in the literature (e.g. Ball, Rom and Glenne, 1983; Karczewska, Szerszen and Kabala, 1998; Kuo, Heilman and Baker, 1983; Balsley and Caldwell 1974; Brams, 1985; Temple, Linzon and Chai, 1977; Bolter et al., 1974; and others). Because the randomly located, high arsenic concentrations encountered during the SOW were not consistent with widespread anthropogenic background levels or air emissions from the Globe Plant smelter (Figures 5 and 6), some other source or stratified

background population appeared to be responsible for these anomalous arsenic concentrations.

Extensive studies were undertaken to determine the source of the anomalously high arsenic concentrations encountered during the SOW investigations (Enviro-Group 1997; Folkes, Helgen and Litle, 2000). The standard techniques for separating background populations from smelter impacts were not applicable (i.e. determining a threshold background concentration), since the anomalous background concentrations were generally higher than those due to smelter emissions. In addition, the anomalous background levels only occurred on certain properties and could not be used as a general background for other properties. Further complicating the evaluation, the distribution of the anomalous arsenic concentrations was random and



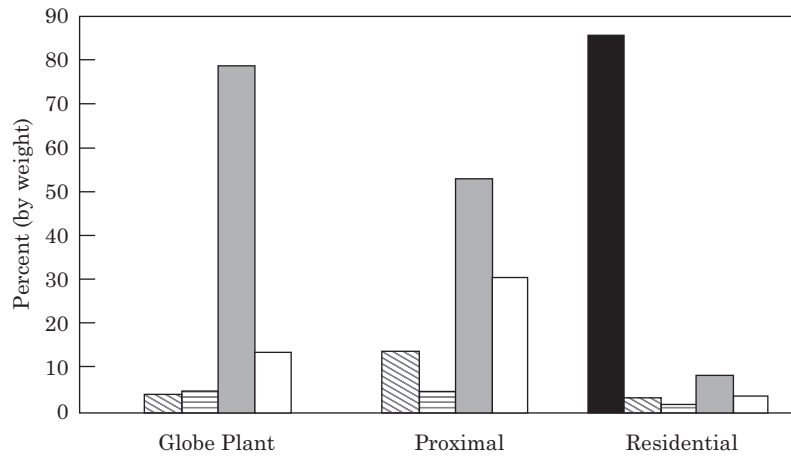


Figure 13. Arsenic phases. ASO (■); As? (▨); Fe/As (▤); Pb/As (▥); other (□).

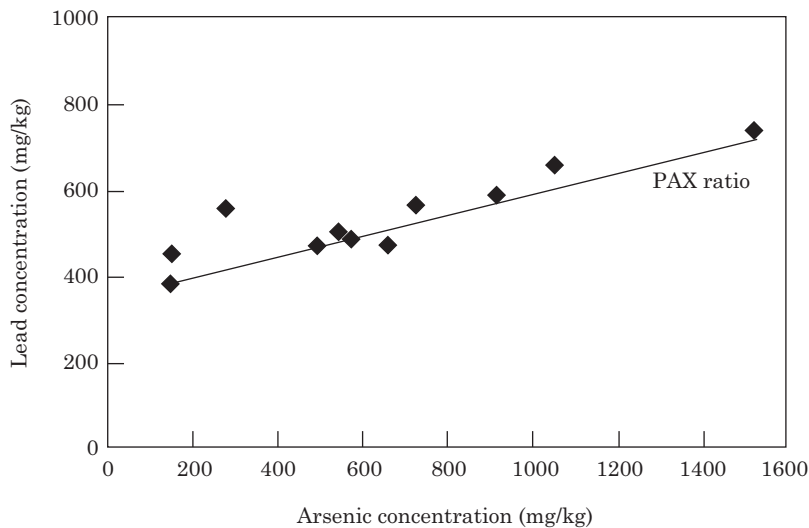


Figure 14. Correlation between soil data and PAX Pb:As ratio.

arsenic match the PAX formula almost exactly (Figure 14). Finally, a one to one correspondence between high arsenic and the presence of perlite, an inactive ingredient of PAX (US Patent 3057709, 1962) was observed. Therefore, while applications of lead arsenates on fruit trees and other arsenical pesticides may contribute to the high arsenic concentrations found on some residential yards, use of the product PAX is the most likely source of the high arsenic on most of the properties.

The statistical properties of the pesticide background population were examined using the method of Sinclair (1976) and SOW data from an area over 1 km south of the Globe Plant, where RIFS data (collected on relatively undisturbed properties) indicated smelter impacts were minimal. An inflection in the data curve was noted at about 50 mg/kg (Figure 15), resulting in a background population with a mean concentration of about 11 mg/kg and a mean plus 2 SD concentration of about 47 mg/kg, similar to the results of the widespread anthropogenic background evaluations presented in Table 1. The pesticide population, based on this separation, has a geometric mean concentration of 141 mg/kg and a geometric SD of 2.56.

### Conclusions

Experience at the Globe Plant has demonstrated the complexity of arsenic background populations in urban areas. Widespread anthropogenic sources clearly elevate arsenic concentrations in soil above natural soil levels on a significant number of properties (Figure 6), resulting in small increases in mean concentrations but more significant increases in variance (Table 1). Because of the biases inherent in any background assessment method, a number of techniques should be employed and compared for consistency, including evaluation of sampling data from remote sites and separation of contaminated and background populations using statistical techniques. Based on the Globe Plant evaluation, an upper limit of background for the widespread anthropogenic arsenic population of about 30 mg/kg is reasonable, although higher concentrations clearly exist.

Stratified background populations may also exist, such as pesticide impacted residential properties in the Denver area. This population only impacts a certain portion of older residential properties, but these have arsenic concentration ranges that exceed those due to

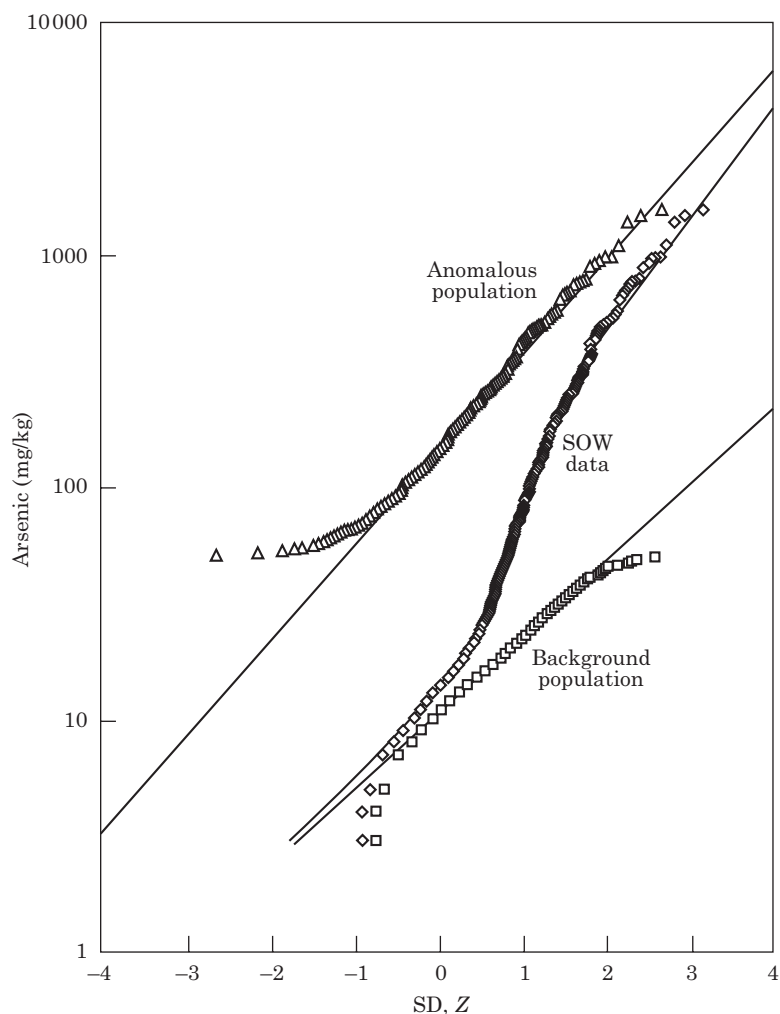


Figure 15. Log probability plot of SOW data.

smelter emissions. This particular pesticide, which was likely applied to a large number of residential properties in the Denver area and other cities, appears to have resulted in a mean arsenic soil concentration of about 141 mg/kg and with several values over 1000 mg/kg.

Forensic fingerprinting, rather than threshold concentrations, are necessary to separate this population from smelter impacts, including correlation with land use, geochemical ratios, and indicator compounds. In addition, the smelter impacts can be distinguished by spatial trends to establish likely boundaries and concentration ranges for given distances from the source, potentially limiting the number of potentially expensive fingerprinting analyses. Spatial trends can be interpreted from site data (e.g. from data sets excluding the stratified background population based on land use) and by modeling of historic air emissions and deposition (e.g. [EnviroGroup, 1998](#)).

In addition to the widespread anthropogenic and stratified background populations, other spatially correlated background populations may exist, such as impacts due to other air emission point sources. Both fingerprinting and spatial correlation techniques may be used to separate the contributions of these sources from the source of concern. In all cases, it is important to recognize the potential for varied and complex

background populations and to utilize the available analytical tools to separate their impacts.

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