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Abstract

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1. Introduction

Mercury is a neurotoxin that accumulates in the food chain and is therefore a health concern. Concentrations of mercury in the air are of little direct health concern. However, mercury in the air falls onto the Earth's surface through dry and wet deposition processes. This mercury can enter water bodies where a small percentage (< 10%) is transformed to methyl mercury. This chemical form of mercury readily enters the food chain and bioaccumulates. Upper trophic level fish can have mercury concentrations several orders of magnitude greater than that found in the water or sediment. As mercury accumulates in these organisms, ecological risks occur and potentially human health risks may occur through fish consumption.

- On December 15, 2003, the U.S. Environmental Protection Agency (EPA) signed its first ever proposal to substantially cut mercury emissions from coal-fired power plants. The Utility Mercury Reductions proposal would cut mercury emissions by nearly 70 percent when fully implemented. The Utility Mercury Reductions rule would permanently cap emissions from coal-fired power plants and provide companies with flexibility to achieve early reductions of mercury. On March 15, 2005, EPA selected a market-based "cap and trade" program that, if implemented, would reduce nationwide utility emissions of mercury in two phases. Emissions would be capped at 33 tons per year by 2010 and 15 tons per year in 2018. When fully implemented mercury emissions would be reduced by 33 tons per year from current levels (nearly 70 percent).

There are technical doubts pertaining to local deposition of mercury leading to “hot spots.” This has received widespread attention in the literature (ES&T, 2004a, b, c). The following are selected quotes from public health officials and the popular press in the past year.

- “Unlike most pollutants, mercury is highly toxic and does not disperse easily, creating “hot spots” of contamination.” (Kathleen McGinty, Director of Pennsylvania Department of Environmental Protection, July 2, 2004. (http://www.dep.state.pa.us/newsletter/default.asp?NewsletterArticleID=8850&SubjectID=)
- “Specifically, we are concerned that local "hot spots" of elevated mercury may result or worsen, especially if the required reduction levels are not sufficiently strict.” (Renee Cipriano. Director of Illinois Environmental Protection Agency, February 26, 2004 (Testimony to the U.S. EPA regarding the U.S. Environmental Protection Agency’s Proposal to Control Emissions of Hazardous Air Pollutants from Utilities (January 30, 2004, 69 Federal Register 4652) Docket ID No. OAR 2002-0056.
- “We generally support market-based approaches such as cap and trade schemes, yet we have an equally strong objection to the exclusive use of cap and trade schemes where local emissions “hot spots” are a concern. While mercury pollution and emissions are widespread, indeed a global problem, we share the concerns of many states that EPA’s proposed rule understates the needs for local controls as well”, letter from Stephen Mahfood, Director Missouri Department of Natural Resources to Michael Leavitt, Director U.S. EPA.
- “Sulfur dioxide is light, and travels long distances; power plants in the Midwest can cause acid rain in Maine. So a cap on total national emissions makes sense. Mercury is heavy; much of it precipitates to the ground near the source. As a result, coal-fired power

plants in states like Pennsylvania and Michigan create “hot spots” – chemical Chernobyls – where the risks of mercury poisoning are severe. … That probably means thousands of children will be born with preventable neurological problems.” Paul Krugman, New York Times, p. A-23, April 6, 2004.

- After passing the rule, “Jeffrey R. Holmstead, head of the EPA's Office of Air and Radiation, said the rule would eliminate "hot spots," or high mercury levels, in lakes and streams near big power plants.” Tom Hamburger and Allan C. Miller, LA Times, March 16, 2005.

- In contrast, that same article in the LA times stated “The market approach has proved effective in reducing the gases that contribute to acid rain, but even some advocates of that program say it won't work with mercury. Mercury is heavier than the acid rain gases, so it tends to fall out of the air near the power plant that emits it, creating the hot spots that could be dangerous.

  "Hot spots are a concern with me," said John A. Paul, a Republican environmental regulator and fisherman in Ohio who served as co-chairman of an EPA advisory committee on mercury. "I advise anyone who eats fish caught in a lake or a stream near a power plant that they are at risk, and that this rule will do nothing to protect them — and might make things worse." Tom Hamburger and Allan C. Miller, LA Times, March 16, 2005.

1.2 Defining a "Hot Spot."

Although the term "hot spot" appears frequently in the health and environmental literature, precise definitions do not. Generally speaking, a "hot spot" is a spatial anomaly, i.e., a location whose properties exceed those generally expected in the area. In statistical terms, a hot spot is an location whose properties exceed more than about 2 or 3 standard deviations above the relevant mean. Methods to statistically define a hot spot for metals in soils generally compare the data to a log normal distribution and look for variations between the two (Tobias, 1997). Some authors have simply defined "hot spots" as the highest observed values; for example, Worm et al. (2003) found a range of diversities in ocean predator species of less than one order of magnitude, among which the highest groups were termed "hot spots". Lebret et al. (2000) used the term to refer to locations where the ambient air quality standard for NO_2 is routinely exceeded. These two examples of "hot spots" would not meet the statistical definition.

However, the expected ranges of environmental concentrations depend heavily on the "natural" or background values and on the length of the measurement period (i.e., the averaging time). In many cases, environmental concentrations are log-normally distributed (skewed towards high values), so that the distribution is best described by the log-mean and the geometric standard deviation (GSD). As an example, Lu et al. (2005) studied the distribution of the polycyclic aromatic hydrocarbon naphthalene in Southern California and found a large range of values, in part because there is little or no natural background. The log-mean and GSD for 13 one- to two-month averages were 227 ng/m^3 and 1.57, leading to an expected range of a factor of 6. However, for 16 four-day averages in other locations, these values were 269 ng/m^3 and 3.12, leading to an expected range of a factor of 94. In both of these distributions, the top 3 or 4 values appear to be outliers and thus bona fide “hot spots”, since deleting them reduces the range of the distribution by about a factor of 2.

While small-scale transient hot spots may be a valid concern for inhalation exposures, the situation with mercury is quite different. The exposure pathway is through diet, and the relevant human exposure times relate to the development of the fetus and are of the order of months. Although a large point-source of Hg indeed constitutes an emission “hot spot”, it does not constitute a fetal exposure hot spot. In addition to the substantial global background in Hg air concentrations and deposition, the following processes act to smooth out spatial anomalies:

- Atmospheric variability, including winds and precipitation.
- Re-emission of mercury from vegetation.
- Terrestrial leaching and washout in transferring watershed deposits into water bodies.
- Aquatic mixing within water bodies.
- Spatial and temporal variability in biomagnification processes.
- Variability among fish species.

Only atmospheric variability is included in the models that may be used to define deposition “hot spots.” The other processes involve spatial variability, especially with regard to mixing within the receiving waters, for which the size of the water body may be key. In order for a local Hg deposit to pose a risk to a developing fetus, its mother must routinely consume high-Hg fish from an affected water body for several months, probably at the rate of 2 or 3 meals per day. While this scenario is unlikely in any event, it also requires a substantial fish population, which requires a substantial body of water, say of the order of tens of square km.

Many empirical findings concerning Hg are subject to experimental error, which may be considerable in some cases. Accordingly, statistical methods may be required to gain an understanding of the data. They include averaging, correlation, and linear regression, using established relationships to try to reduce experimental variability. In all cases, "statistical significance" implies a 95% probability that the finding is not due to chance alone, denoted as "$p < 0.05$".

This study examined the possibility that coal-fired power plants act as local sources leading to mercury "hot spots", using two types of evidence.. First, the world-wide literature was searched for reports of deposition around mercury sources, including coal-fired power plants. Second, soil samples from around two mid-sized U.S. coal-fired power plants were collected and analyzed for evidence of "hot spots" and for correlation with model predictions of deposition.

2. Evidence from the Literature for "Hot Spots" Near Mercury Emissions Sources

The rationale for regulating air emissions of mercury from U.S. coal-fired power plants largely depends on mathematical dispersion modeling, including the atmospheric chemistry processes that affect the partitioning of Hg emissions into elemental (Hg(0)) and the reactive gaseous mercury (RGM or Hg^{+2}) forms that may deposit more rapidly near sources. Mercury is a global pollutant and therefore, modeling estimates are often based on a large scale. Fine scale, (< 20 km) modeling of point sources is not performed often. In addition, there is evidence (Edgerton 2004, EPRI 2004) that reactive gaseous mercury in coal-fired power plant plumes quickly reduces to elemental mercury. This chemical process, which would greatly affect the amount of local deposition however, the corresponding chemical reactions are not included in the available local mercury deposition models. As a result, field data are necessary to check for the existence of mercury ”hot spots” near coal-fired power plants.

Literature reviews examined the evidence for deposition around local sources including coal-fired power plants (Lipfert et al. 2004, Sullivan, et al., 2005) . The extant experimental data are considered at three spatial scales: local (< 30 km), regional (< ~300 km), and national (multi-state data). The reviews covered data on mercury content in soils, sediments, precipitation, and fish and mercury deposition rates in precipitation. The reviews (Lipfert et al. 2004, Sullivan, et al. 2005) lead to the overall conclusion that atmospheric deposition of Hg is affected by emissions from coal-fired power plants. However, because of the numerous assumptions required and the use of simplistic models, it is not possible to describe these relationships accurately based on these findings. Nevertheless, coal-fired plants seem to contribute less than 10% of total Hg deposition on a national scale, and the resulting effects on fish Hg appear to be even smaller. In contrast, there is strong evidence of enhanced local deposition within 3 km of the chlor-alkali plants, with elevated soil concentrations and estimated deposition rates of 10 times background. (Lodenius, 1998, Biester et al. 2002, Biester et al. 2002a, Sensen et al. 2002, Southworth et. al. 2004). Evidence for enhanced deposition near a calcining plant also exists (Abbott et al. 2003, Susong et al. 2003).

Reliable quantitative understanding of the processes of mercury emissions, deposition, and translocation through the food chain remains elusive. Complex atmospheric chemistry and dispersion models are required to predict precise concentration and deposition contributions, and aquatic process models are required to predict effects on fish. However, there is uncertainty in all of these predictions, especially with regard to the fate of reactive gaseous mercury in coal-fired power plant plumes. Therefore, at this time, the most reliable way to understand the impacts of coal-fired power plants on Hg deposition is based on empirical data. In terms of excesses over background, local soil concentration Hg increments are around 30%-60%; sediment increments are 18-30% (Anderson et al. 1977, Menounou et. al. 2003) and wet deposition increments are around 11-12% (Lipfert et. al., 2004). Soil and sediment effects are necessarily cumulative, in contrast to wet deposition. Based on the empirical finding (Lipfert et al., 2004) that fish Hg is proportional to the square root of wet deposition (after controlling for water chemistry), then the contribution of coal-fired power plants to fish Hg would be about 5-6%. Local differences in water chemistry may also help explain the absence of a relationship between state-level fish concentrations and wet deposition levels; it is possible that the absence of local impacts on fish at the Kincaid power plant (Anderson, et al. 1977) was related to water chemistry.

3. Measuring Soil and Vegetation Concentrations of Mercury around Coal-Fired Power Plants

Studies of soils, sediments, and wet deposition around coal plants typically find some evidence of enhanced deposition; however, the impact and statistical significance of the results is generally weak. Many of the coal plant studies (Klein et al. 1973, Anderson et al. 1977, Crockett et al. 1979, Kotnik, et al. 2000) were conducted in the 1970's when emission rates were higher due to fewer pollution controls and the use of coals with higher Hg content. In addition, these studies did not attempt to correlate modeled deposition with measured soil concentrations.

The Hg deposition models are based on a number of assumptions and hence there is uncertainty in the predicted deposition rates. A key assumption in the models is that the mixture of reactive gaseous mercury (RGM) to elemental mercury Hg(0) is constant in the exhaust plume. However, recent experiments suggest that reactive gaseous mercury converts to Hg(0) quickly (Edgerton et al, 2004, Laudal et al, 2004). If the hypothesis is correct, then local impact of coal-fired power plants will be greatly reduced, since Hg(0) does not deposit as quickly as reactive gaseous mercury. In studies conducted at two sites in Georgia and one in Florida (Edgerton et al., 2004), plumes from power plants were tracked using the sulfur dioxide emitted from the stacks as a tracer. For 31 plume touchdown events, mercury speciation at the ground level monitoring station, which was 14 – 150 km away from the power plants, showed levels of reactive gaseous mercury around 14 - 23% as compared to 53 - 75% when emitted from the stacks. Wet deposition cannot account for this reduction, as rain did not occur over the period of travel time from emission to the monitoring point (< a few hours). Dry deposition effects were also minimized by collecting samples early in the day when dry deposition is minimized. Studies of airborne mercury speciation in power plant plumes also showed a reduction in the percentage of reactive gaseous mercury (Laudal, 2004). An airplane was flown through the plume from the Pleasant Prairie power plant in southern Wisconsin at distances of less than ¼ mile, 5 miles, and 10 miles from the stack. Measurements of the stack gas showed that releases were 67% Hg(0) and 33% Hg^{+2}. Less than ¼ mile from the stack, the mercury speciation changed to 83% Hg(0) and by five miles approximately 88% of the mercury was in elemental form, Hg(0). This percentage of Hg(0) was maintained at the 10 mile distance. While there are uncertainties associated with measuring mercury in the plume from an aircraft, these results are consistent with the findings on the ground (Edgerton et. al, 2004) and eliminate differential deposition as the cause for the depletion of Hg^{+2}.

The extant computer modeling suggests that increased local deposition will occur on a local (2 to 10 km) to regional scale (20 to 50 km) with local increases a small percentage of background deposition on the regional scale. (EPA, 1997, Sullivan, et al., 2001, Sullivan et al., 2003). The amount of deposition depends upon many factors including emission rate, chemical form of mercury emitted (with reactive gaseous mercury depositing more readily than elemental mercury), other emission characteristics (stack height, exhaust temperature, etc), and meteorological conditions. Modeling suggests that wet deposition will lead to the highest deposition rates and that these will occurwithin a few km of the plant. The rates of dry deposition are are predicted to be less than for wet deposition, but they apply to a much greater area (Sullivan et al., 2003). Therefore, on the regional scale, dry deposition may be more important than wet. However, it is quite difficult to measure dry deposition of Hg directly.

To further understand the impacts of local deposition, soil and vegetation samples were collected around two coal-fired power plants and analyzed for mercury. One plant is a mid-size plant in the midwest that burns locally mined lignite. This site will be referred to as Plant A (for reasons of confidentiality). The second plant is the Kincaid power plant located southeast of Springfield IL that was studied in the 1970's (Anderson et al., 1977). This study combines modeling of mercury deposition patterns with soil and vegetation mercury measurements. The deposition model used emissions data, meteorological conditions, and plant data to define sample locations likely to exhibit deposition in excess of background that can be attributed to the power plant.

The data are evaulated looking for revidence of ‘hot spots’. Comparisons between the data and the models were made to test the validity of the model.

3.2 Deposition Model Paramaters

The local atmospheric transport of mercury released from the coal-fired power plants was studied to estimate the local impacts of mercury deposition. The Industrial Source Code (ISCST3) Short Term air dispersion model was used to model these processes (EPA, 1995). This code is an updated version of the computer code used by the U.S. Environmental Protection Agency to examine local deposition from combustion sources in their report to Congress in 1998 (EPA, 1997).

Mercury emissions data from the lignite-burning power plant (Plant A) and the Powder River Basin coal-burning power plant (Kincaid) were used to represent the source terms. Meteorological data from nearby weather stations were used to simulate typical weather patterns. This approach was selected to test the consistency between model results and environmental monitoring data that suggests that measured mercury levels in environmental media and biota may be elevated in areas around stationary combustion sources that emit mercury.

Modeling deposition requires three key sets of parameters: source emissions rate, deposition parameters, and meteorological data. These are described in detail in Sullivan (2005). The following sections summarize key data for deposition modeling..

3.2.1 Emissions

Three types of gaseous mercury species occur in the emissions and they behave quite differently once emitted from the stack. Elemental mercury, Hg(0), due to its high vapor pressure and low water solubility, is not expected to deposit close to the facility. In contrast, reactive gaseous mercury (RGM), Hg^{+2}, is much more soluble in water and is accommodated in rain and therefore, will deposit in greater quantities closer to the emission sources. In addition, RGM will also undergo dry deposition at a much higher rate than elemental mercury. The third type of emission is in particulate form. Previous studies (Sullivan, et al. 2003) showed that for less than 2% Hg(p), deposition was dominated by RGM with only a small contribution from Hg(0) over 30 km from the plant. The deposition is linearly proportional to source strength. Therefore, errors in the estimated amount of RGM will affect the total deposition, but not the deposition pattern. Due to emission controls at the power plants, less than 1% of the Hg is expected to be particulate form and this type of mercury was not modeled.

3.3.1.1 Plant A

Power plant A features two units with a total generation capacity of nearly 1,200 megawatts. The plant first started generating electricity in 1979. The total emissions from the two stacks at Plant A were 344 kg or $1.2\ 10^{-2}$ g/s. Measured speciation data for Plant A indicated 82.2% Hg(0) and 17.8% Hg^{+2}. Using the fractional release rate from the test data, the release rate for each mercury category is 0.01 g/s for Hg(0) and 0.002 g/a for Hg^{+2}.

In addition, there was another coal-fired power plant located approximately 15 km to the southwest of Plant A. Speciation data were not available for this plant and it was assumed that it was identical to Plant A (EPA, 2001). This plant emits 267 kg (0.0085 g/s) of mercury per year at the following rates 0.007 g/s of Hg(0) and 0.0015 g/s of Hg^{+2}.

3.3.1.2 Kincaid Plant

Total mercury emissions from the Kincaid power station were 161 kg (0.0051 g/s) in 2001. Speciation data were not available, however other plants burning Powder River Basin coal generally emit 15 – 30% reactive gaseous mercury with the remainder Hg(0). For modeling purposes, a value of 20% reactive gaseous mercury and 80% Hg(0) was selected. Based on these assumptions, emissions from Kincaid were 0.0041 g/s for Hg(0) and 0.001 g/s for Hg^{+2}.

The city of Springfield also operates a small coal-fired power plant approximately 25 kilometers to the northwest of the Kincaid power plant. Releases from this small plant were 11 kg/yr (0.00035 g/s) and were included in the modeling. Mercury release rates from the Springfield Power plant are 0.00028 g/s for Hg(0) and 0.00007 g/s for Hg^{+2}.

Due to the separation distance and small size of this plant as compared to the Kincaid plant, modeled deposition from the small plant did not change the deposition pattern around the Kincaid plant.

3.3 Modeled Deposition and Data Collection and Analysis for Plant A

3.3.1 Deposition Modeling

Meteorological data for a five year period from the nearest airport, located about 40 miles away, were reviewed to determine wind patterns under dry and wet conditions. Under dry condtions, the prevailing winds ran along an axis from the northwest towards the southeast Winds occurred regulary in each direction along this axis. Under wet conditions, winds were generally from the north and east. This leads to predictions of wet deposition near the plant and to the southwest. Deposition modeling of the emissions from the two power plants (e.g. regional background was not modeled) based on the meteorological data predicted highest deposition rates within 10 km of the plant in a southwesterly direction. The modeled deposition near (0,0) in Figure 10 is from plant A. The depositon patttern in the southwest corner (-10,000, -12,000) is from the second plant. The patterns do not show substantial overlap. Total deposition rates were 3 – 10 ug/m^2/yr. The total background deposition in this region is expected to be 10 – 20 ug/m^2/yr. Thus, the plant may produce a region of a few tens of square kilometers with depostion at 15 – 100% above background. The region with predicted deposition more than 100% above background (contour of 10 μg/m^2/y) is less than 1 km^2 in area. The region that was 15% above background deposition is less than 20 km^2. Dry depostion rates were lower than wet depostion rates and were not predicted to be a major contributor to deposition in the region. Total deposition was dominated by RGM. Over the modeled domain, deposition of Hg(0) was a small fraction of the total deposition even though 82% of the emitted mercury is in this form.

3.2.2 Sampling Design

Based on the modeled deposition analysis a soil and vegetation sampling design was selected to extend approximately 8 km to the south and west of the plant. Figure 11 shows the final sampling grid which which was modified from the general layout suggested by the modeling to account for site-specific conditions (e.g., inaccessibility of sample locations, site activities, and changes in soil type which would alter background levels of mercury). The sampling area south and west of the plant covered an approximately square region of 64 km^2. The land surrounding the power plant was either part of an active strip mine or agricultural. Although many sampling sites were within the strip mine permit area, most of the land had been reclaimed. Strip mine personnel identified sites that had been fully reclaimed, or were at least known not to have been disturbed for at least a year. Agricultural area sampling sites were chosen because they appeared undisturbed for at least one year (i.e. had not been plowed). Many of the agricultural sites were at the crest of roadside ditches, adjacent to a plowed or mowed area.

Soil and vegetation samples were collected in November 2003 at 54 selected sites around the coal-fired power station as shown in Figure 10. At each site, five samples were collected. Three surface samples from the top five centimeters of soil separated by approximately 3 m, one deep sample at a depth of 5 – 10 cm, and one sample of the vegetation.

Samples of approximately 100 grams weight were collected in watertight wide-mouth 250 mL plastic screw-top cups. Samples were collected using stainless steel trowels, which were rinsed with tap water and wiped dry between each use. Blind field duplicates were collected every 10th sample. Samples were shipped back to Brookhaven National Laboratory (BNL) for analysis. Latitude and longitude for each sample location were identified using a GPS locator system (Garmen Etrex) with a resolution of 6 meters.

3.3.3 Mercury Analysis Methods and Quality Assurance

The soil samples were analyzed using a Direct Mercury Analyzer (DMA-80, Milestone, Inc, Monroe, CT), Figure 12. Solid samples, approximately 0.5 grams, are placed on small boats that enter the DMA. The typical working range for this method is 0.05-600 ng of mercury. Since soil samples are at most about 0.5 grams, the DMA-80 easily measures levels below 1 ppb (ng/g). DMA-80 analyses were conducted on soil samples as received. Moisture content was determined separately for all samples, and mercury concentrations were adjusted to a dry weight basis.

Quality assurance was evaluated through taking blind duplicates of 10% of the samples, measurement of empty sample boats in the DMA-80, and use of one of two NIST mercury standards (SRM 2709 and SRM 2710) at every 10th measurement. The NIST soil standard SRM 2709 San Joaquin soil) was used for soil measurements. It has a mercury level of 1380 +/- 80 ng/g. The NIST standard SRM 2710, peach leaves, was used for vegetation samples. It has a mercury level of 30 +/- 5 ng/g. Empty boat samples averaged 1.4 ng/g and were all less than the minimum soil sample (10 ng/g). Blind duplicates were statistically similar to the similar soil samples. Each sample was measured in triplicate to examine the homogeneity of the sample. The range of the mercury levels in the three samples averaged +/- 12.5% of the average of the three samples.

3.3.4 Data Analysis and Interpretation

3.3.4.1 Soil Data

At each sample location, the three surface soil samples were averaged to give a composite. Analysis of the data shows that they are log normally distributed, as would be expected for soil samples (Tack, 2005). At the fifty-four locations the average value was 27.6 ng/g (dry weight basis), with a standard deviation of 6.9. The minimum value was 11.6 ng/g and the maximum value was 55.4 ng/g. All of the data were within approximately a factor of 2 of the average value thereby suggesting that ‘hot spots’ were not found in the soil near the plant, in contrast to the findings for chlor-alkali plant discussed previously. For example, the four highest values (> 36 ng/g) might be outliers, but only two are contiguous and none fall into the patterns predicted by modeling. Averaging each of these four values with their nearest neighbors, to simulate what might occur in a sizeable water body, reduces their magnitudes considerably. These averaged values are from 5-13% above the median of all samples (27.5 ng/g), and do not constitute “hot spots” in the conventional sense.

Comparison between the predicted deposition versus measured mercury concentrations in the soil was accomplished by overlaying the deposition map over the sampling map with sample results color-coded by measured concentration. Figure 3 is the graphical representation of the analysis. Soil concentrations were binned into three approximately equal size groups containing 17 or 18 samples. Sample locations with symbols representing measured mercury levels represent the measured data. The range of the groups was 11.4 – 25.2, 25.2 – 29.5 and 29.5 – 55.4 ng/g. The tight spread of the middle group shows that 1/3 of the soil Hg measurements were in this narrow range. Predicted regions of enhanced deposition are covered by the filled contours with red representing 5 ug/m^2/yr and blue representing 3 ug/m^2/yr.

The model predicts the incremental rate of deposition due to the coal plant emissions, while the measured data are soil concentrations, reflecting the effects of cumulative deposition, both local and regional, and eh natural constituents of the soil. Therefore, a direct comparison between the modeled results and the measurements is not possible. However, if excess deposition were occurring in a region, it is expected that this would be reflected by higher soil concentrations. Figure 15 shows that the patterns of modeled deposition and measured data do not match. The measured soil data suggest that the main finger of the plume is slightly south of the area predicted by modeling. Also, the measured data shows a fair degree of scatter, as expected, in contrast to the smoothly varying deposition pattern. To evaluate if there was a match between the data and the model statistically, both were ordered from high to low and a rank correlation between the soil and predicted deposition was performed. The Spearman rank correlation coefficient was -0.02 indicating no correlation between the two.

A second approach to determining if excess deposition was occurring was to compare the concentrations of surface soil (0 – 5 cm) and subsurface soil (5 – 10 cm) collected at each sampling location. The subsurface samples showed similar values and characteristics as the surface samples. The average value was 28.2 ng/g with a range of 10 – 49 ng/g for the subsurface samples. The Spearman Rank correlation coefficient between the surface and subsurface soil Hg concentrations is 0.77 indicating a high degree of correlation. Therefore, there is no evidence that local deposition increased the surface Hg soil concentrations relative to the subsurface soil.

Although there is no strong evidence of excess deposition in the soils near the plant, estimates of the potential deposition were performed for the 8 km (5 mile) quadrant to the southwest of the plant where data were collected. The following assumptions were used to estimate excess deposition.

Case a) the difference between the average surface and average subsurface soil values. This value is 0.6 ng/g.

Case b) the difference between the average surface concentration and the average of the lowest 1/3 surface concentrations. This assumes that the pre-operational background was the average of the lowest 1/3 of the samples. This ‘background’ average is 22.4 ng/g and the average of all soils was 28.8 ng/g.

Case c) the predicted modeled yearly deposition generated by ISTCST (408 g/yr) multiplied by the operational period (23 years).

Using a soil density of 1.5 g/cm^3 and limiting the excess deposition to the top 5 cm give a total volume of soil of 2.55 10^6 m^3 and a soil mass of 3.8 10^{12} g over the study area. Assuming that current mercury emission rates and speciation distribution apply over the 23-year operating period of the plant, Table 1 presents the estimated amount deposited, the % of RGM emissions, and the % of total emissions that this represents. The table shows that for any scenario, less than 2% of the RGM (less than 0.5%) of the total mercury emissions from the plant deposited within this 8 km square quadrant to the southwest of the plant. Even though modeling suggests greater deposition in the sampled region, if it is assumed that similar deposition occurs in the other 3 quadrants, then less than 7% of the RGM emitted deposits within 8 km (5 miles of the plant). This analysis also supports the contention that although mercury levels may be slightly elevated near the plant, a 'hot spot' large enough to substantially affect water quality and fish Hg does not occur in the area sampled near the plant.

3.3.4.2 Vegetation Concentrations

One vegetation sample was collected at each sampling location. The vegetation samples are a measure of mercury deposition over the current growing season. Vegetation mercury levels are known to be influenced by both wet and dry deposition of mercury. In sampling, every attempt was made to collect the same type of vegetation, grass, from each location. This was not always possible, but all samples are grasses. The samples were analyzed in duplicate and the average value was taken as a measure of the Hg content for samples that were low in mercury and provided similar readings in both measures. Many samples showed high levels of Hg (> 1ppm) and wide variability in the measured value. These samples were analyzed between 3 and 7 times to improve the accuracy of the results. The statistics from the average at each location are in Table 2.

Unlike the soil samples, the vegetation samples did show regions of elevated Hg concentration. Defining a high Hg concentration as any average value that is 3 times above the median value for all samples shows that 12 of 49 samples have ‘high’ concentrations. Individual samples were in excess of 1000 ng/g. Although, there was wide variation in the measured mercury value for the vegetation with ‘high’ levels of mercury (>50 ng/g), these samples were consistently high in all of the samples.

Figure 4 presents the measured mercury concentration in vegetation and the predicted deposition. In Figure 4, the blue contour represents excess deposition of 3 $ug/m^2/yr$ and the red represents 5 $ug/m^2/yr$. Soil vegetation samples were divided into four groups representing the range from 10.7 – 22.9; 22.9-31.5, 31.5 -84.4; and 84.4 – 691 ng/g. Each range had 12 or 13 members and therefore, they represent ¼ of all samples. Approximately ½ of the samples were within 400 m of the plant. In Figure 4, the highest values are near the plant, primarily to the north and west. This is the primary direction of wind flow during dry deposition conditions. However, deposition modeling would predict that the peak deposition rates would occur further from the plant due to the stack height and buoyancy effects. All of the values in the top ¼ of the distribution are within 4000 m of the plant with the exception of one value located approximately 6500 m east of the plant. This site was selected to be out of the predicted deposition pattern and was hoped to be representative of background. Clearly it is not. Although the frequency distribution of the entire set of samples appears to be reasonably continuous, the spatial distribution is not. Most of the very high values are located adjacent to a coal transporter that was previously used to move coal from the mine to the plant.

Table 3 presents the average concentration as a function of distance from the plant for the vegetation and soil samples. The vegetation samples show a peak between 1500 – 3000 m from the plant and a decline after that point. The soil samples do not show any significant trend with distance. This data suggests that localized samples with elevated Hg concentrations were seen in the area, but they were not clearly associated with the emissions from the power plant. Possibilities for the high Hg concentrations in vegetation include fugitive emissions (e.g. dust from coal handling, etc.) and proximity to roads. To resolve this issue would require additional sampling and analysis for other elements that are found in coal plant emissions.

3.4 Data Collection and Analysis for the Kincaid Plant

The Kincaid plant was selected because it has been studied in the 1970's (Anderson et al. 1977) for increases in mercury content in the soil, sediment, air, and fish. In addition, tracer studies were performed on the emissions from this plant and measured concentrations were compared with predictions by the ISCST computer code (Cox, et al. 1986). This provides confidence that the ISCST models reasonably represent the plume at this site.

The Kincaid plant began operation in 1967 and has two 575 MW boilers each with a 187 m exhaust stack. In addition to the Kincaid Plant, the city of Springfield operates a small plant that emits less than 9 kg (20 lbs) of mercury per year. This small plant is approximately 15 miles northwest of the Kincaid plant. Both plants were simulated in the deposition modeling.

3.4.1 Deposition Modeling

Meteorological data from the Springfield airport, located about 30 kilometers away, were reviewed for a five-year period to determine wind patterns under dry and wet conditions. Under dry condtions, the prevailing winds were primarily from the south. Winds occurred from the south 20% of the time. No other direction had winds more than 10% of the time. Winds rarely came from the Northeast. Under wet conditions, winds were generally from the south and north. However, precipitation also fell frequently when winds were out of the east.

Deposition modeling of the emissions from the two power plants based on the meteorological data considered both dry and wet depositon; however, regional background was not modeled. Simulations were performed for the 5 year period from 1986 – 1990. Ths period included annual rainfall ranging from 24 to 46 inches and was representative of the range of condtions experienced in the region. From the results annual deposition rates were calculated for both dry and wet deposition. Dry deposition is predicted to peak approximatley 18000 m north of the plant at a rate of 0.6 ug/m^2/yr. The highest wet deposition rates were predicted within 10 km of the plant in a northerly direction. The predicted peak deposition rate was 18 ug/m^2/yr at 1000 m north of the plant (this is the first calculated point). The deposition plume remains above 5 ug/m^2/yr for approximately 2 or 3 km around the plant and above 1 ug/m^2/yr for 7 or 8 km. For comparison, in the period of 1999 – 2003, wet deposition (due to all sources including the regional background) averaged 9.3 ug/m^2/yr at the nearest mercury deposition network (MDN) site in Bondville, IL which is approximatley 100 km west of the plant. Dry deposition is not well characterized but is expected to be approximately the same as wet depostion. If this is true, at the location of the predicted peak deposition near Kincaid, total Hg deposition would be approximately twice background. Depostion rates resulting from Kincaid emissions would add less than 5% of background after 8 km.

Figure 5 also illustrates the impacts of dry deposition on the overall depositon pattern. The colored contours represent the wet deposition at levels of 0.5, 1, 5, and 10 ug/m^2/yr. The dotted contour line represents the total deposition (wet and dry) and shows the extension of this region to the north of the plant due to dry deposition. At deposition rates above 1 ug/m^2/yr, dry deposition does not impact the contours because the predicted dry depostion rate in this region is much less than 1 ug/m^2/yr.

3.4.2 Sampling Design

Even though the deposition modeling predicted higher deposition to the north of the plant, due to the absence of a strong signal in the soil mercury levels at Plant A, it was decided to sample all directions uniformly around the plant. A sample grid centered around the plant and extending approximately 8 km in every direction was developed. Samples were spaced approximately 1600 m (1 mile) apart. A total of 123 sample locations were selected. In addition, 8 additional sites were chosen with the intent of defining background. These sites were from 17.5 – 38 km from the power plant. At each sampling site, 3 surface samples (0 – 5 cm), 1 subsurface sample (5 – 10 cm) and 1 vegetation sample were collected in July, 2004. With 5 samples at each of the 131 locations, this results in 655 mercury analyses for the complete set. Samples were collected approximately 15 - 20 feet from the road edge in regions that were not plowed for farming. Eight sample locations were less than 10 feet from the road due to space limitations. The sampling network near the plant is shown in Figure 6.

The region around the plant is mostly open farmland. Lake Sangchris, a state park, is located directly to the north and slightly east of the plant in the region of highest predicted deposition. Lack of access roads in this region prevented higher density sampling.

3.4.4 Data Analysis and Interpretation

3.4.4.1 Soil Data

A total of six values (3 locations and duplicate measures at each location) were used to estimate the average at each sample location. At the 124 locations around the plant, the average value was 32 ng/g (dry weight basis), with a standard deviation of 17.7 (GSD =1.34). The median value was 25.9 ng/g. The minimum value averaged over the six measurements was 16.9 ng/g and the maximum value was 155.6 ng/g. The minimum value for any single sample was 12 ng/g and the maximum for any single sample was 218 ng/g. The data showed a much wider distribution than for Plant A with a few values that were very high above the mean. Statistical analysis of the data (Sullivan, et al. 2005) showed that the data did not fit a lognormal distribution with the distribution skewed towards higher values above the mean. This could imply enhanced deposition, or it could imply differences due to different soils.

In examining the data, it appeared that there were several outliers with values much higher than the mean. For the purposes of further analysis and data interpretation an outlier was defined as any location that had a soil mercury value that was more than a factor of 2 greater than at adjacent locations (< 1 mile). A factor of 2 difference from nearest neighbors could not occur due to deposition only because the deposition rate does not change by a factor of two in less than a mile, Figure 5. Therefore, the change is likely due to having different soil properties. This approach identified outliers at locations identified as A10, B2, D12, F2, and I10 on Figure 6.

These sites had soil Hg values ranging from 56 – 155.6 ng/g. With the reduced data set, the average Hg value in the remaining 119 samples is 29.1 ng/g, a standard deviation of 8 ng/g and a range of 16.9 – 65.2 ng/g. The probability plot for the reduced data set showed that the data was closer to the lognormal distribution, however, it was still skewed toward the higher values (Sullivan, et al. 2005).

Further analysis of potential "hot spots" was undertaken by focusing on the top 3 values, in relation to their immediate neighbors. These 3 values ranged from 2.6 to 5 times the median background (without its outlier). After averaging with up to 8 neighboring samples, these excess deposition ratios were reduced to 0.8, 0.9, and 1.5. Thus, only one location showed excess deposition, when averaged over a larger area to simulate effects on watersheds.

Comparison between the predicted deposition versus measured mercury concentrations in the soil was accomplished by overlaying the deposition map over the sampling map with sample results color-coded by measured concentration. Figure 7 is the graphical representation of the analysis for the samples near the Kincaid plant. Samples collected more than 15 kilometers from the plant for purposes of determining background are discussed later. Soil concentrations were binned into four approximately equal size groups containing 29 or 30 samples. Sample locations with symbols representing measured mercury levels represent the measured data. The range of the groups was 16.9 – 24.6, 24.6 – 27.0, 27.0 – 31.3, and 31.3 – 65.2 ng/g. The narrow spread of the middle groups around the average shows that 1/2 of the soil Hg measurements were in the range of 24.6– 313 ng/g. Predicted regions of enhanced deposition are covered by the filled contours with red representing 10 ug/m^2/yr, purple representing 5 ug/m^2/yr and blue representing 1 ug/m^2/yr.

Examining Figure 7 shows that there is not a particularly good correlation between regions of predicted enhanced deposition and soil Hg concentrations. Although higher concentrations do occur near the plant. In the region of predicted deposition in excess of 5 ug/m^2/yr, 6 of 12 sample locations have soil Hg concentrations in the top quartile. The highest measured concentration (with the outliers removed) 65.2 ng/g occurs at the location closest to the power plant (G7) and approximately 0.8 miles west of the plant.

Figure 8 graphically represents the soil data, binned into the same four groups as in Figure 7 with a base map of the local roads. Examining this figure it appears that there is a correlation between soil Hg and the East-West sample locations designated with the numeral 7 (e.g. A7, B7, etc.). This sampling transect was along the road that passes immediately south of the power plant. This road was the busiest road in the sampling domain near the plant as it had all of the employee traffic, it connected the two nearest towns, and had an access ramp to the interstate to the west of the plant. In addition, samples in the lowest quartile were frequently associated with roads carrying less traffic.

Table 4 presents the average mercury content (ng/g dry) of the three soil surface samples at each location. The outliers at A10 (56 ng/g) , B2 (155.7 ng/g), D12 (78.1 ng/g), F2 (78.9 ng/g), and I10 (120.5 ng/g) are not included. The average along each transect (A- K and 1 – 12) is also presented in the table. The plant was located between G7 and H7. High concentrations (65 ng/g at G7 and 42 ng/g at H7) were measured near the plant. The highest average transect is the east-west transect labeled 7 with an average value of 41 ng/g. The average for all soils is 29.1 ng/g. There is no clear spatial pattern with distance from the plant in the transects. However, average concentrations were higher north of the plant (transects 1 -7), as compared to south of the plant.

Comparison of the surface (0- 5 cm) samples with the deep samples (5 – 10 cm) at the same locations suggests a strong correlation between the two. The highest variability occurred at high soil concentrations. However, the general trend was the same. The Spearman rank correlation coefficient between the surface and deep soils was 0.78 indicating a strong correlation. The average of the deep soils was 0.6 ppb less than for the surface soils. However, this was not statistically significant.

3.4.4.2 Soil Background Samples

Attempts to define background were based on taking soil samples from eight locations at distances of 11 – 23 miles from the plant. Each location was predicted to be well outside the domain of influence for enhanced mercury deposition from the plant. The background sites had higher mercury levels than those near the plant with 4 of the 8 sites above 40 ng/g level. The background sites were selected on fairly well traveled roads for ease of access. Near the plant, higher mercury levels did seem to be associated with higher levels of traffic. The average of all background sites was 40.5 ng/g, close to the average value along the higher traffic transect 7, 41.0 ng/g. The locations may partially explain the higher mercury values associated with the background sites. In any event, the background samples could not be used to estimate a regional background.

3.4.4.3 Vegetation Concentrations

One vegetation sample was collected at each sampling location. The vegetation samples are a measure of mercury deposition over the current growing season because the sample is from growth that occurred this year. Vegetation mercury levels are known to be influenced by both wet and dry deposition of mercury. Attempts were made to take the same type of vegetation from each location. This was not always possible, but all samples are grasses. If possible, samples from undisturbed vegetation were collected, however, in some cases, the vegetation near the edge of the road had been mowed. The samples were analyzed in duplicate and the average value was taken as a measure of the Hg content. For most samples, there was consistency between the two measurements. In contrast to Plant A, the vegetation samples did not show extremely high values and fit a log normal distribution well. Log normal distributions frequently provide a good representation of environmental data that is not impacted by outside sources. The statistics from the average at each location are in Table 5.

The vegetation samples had a different spatial distribution than the soil samples. Figure 9 presents the measured mercury concentration in vegetation and the predicted deposition. Again, there is poor agreement between the predicted deposition and the measured vegetation mercury levels. Soil vegetation samples were divided into four groups representing the range from 0.6 – 3.1, 3.1 – 4.8, 4.8 – 7.0, and 7.0 – 22.5 ng/g. Each range had approximately 30 members and therefore, the ranges represent ¼ of all samples. Examining Figure 9 there does appear to be higher deposition values along the north-south axis centered on the plant, which is the direction of the prevailing winds. However, the highest values are not near the plant. In fact, the samples with the two highest values were at locations B-8 (22.5 ng/g) and A-6 (22.3 ng/g). These locations are to the west more than 6 miles from the plant and not in the direction of the prevailing winds. Transect H, which samples directly north and south of the plant was characterized as having higher vegetation levels of mercury as compared to the other transects. Table 6 summarizes the average concentration data and provides the average along each transect. The average Hg vegetation value along transect H was 9.1 ng/g. No other transect had an average above 7.0 ng/g and the average of all samples was 5.6 ng/g. This could be interpreted as enhanced deposition during non-precipitation events. The rate of dry deposition was predicted to peak due north of the plant. Other major differences between the spatial distribution of Hg in soils and vegetation is the higher values noted along transect 7 in the soil were not found in the vegetation, and the soil concentrations tended to be higher to the northeast of the plant, while the vegetation samples had a higher average value southwest of the plant.

3.4.4.4 Vegetation Background Samples

Again, attempts to find a regional background for the site were unsuccessful. The eight background samples had a much higher average (7.0 ng/g) than the sites near the plant (5.6 ng/g). Five of the eight background samples had an average value greater than 8 ng/g. All of these samples are in the upper quarter of the distribution of samples near the plant. However, this is consistent with finding higher Hg levels in many of the background soil samples.

3.4.5 Concentrations as a function of Distance

The data were grouped as a function of distance from the plant in one-mile intervals. The sites determined to be outliers in the previous section were not used in the soil analysis. All vegetation samples were used in the analysis. There is a correlation with distance for the soil samples, with higher values measured closer to the plant. There is no apparent correlation with distance for the vegetation samples.

3.4.6 Comparison of Soil Sampling Results at the Kincaid Power Plant.

In the April 1974 the Illinois Natural History Survey (Anderson, et al. 1977) conducted a soil sampling campaign on a 3.2 km (2 mile) square grid, 38.6 km across with the Kincaid plant at the center. Two soil types Sable (Illiopolis) silty clay loam (north of the plant) and Virden silty clay loam (south of the plant) were observed. Samples were taken from the top two cm in agricultural fields that had not been disturbed for several months. These sites were at least 50 m and usually 100 m from the road. In the current study, a 1.6 km grid, 20 km wide centered around the plant was used. Soil samples were collected in undisturbed regions generally about 20 feet from the road. However, in some cases, this was not possible and 10 samples were collected at 10 feet or less from the road edge. Samples were homogenized over the top 5 cm. Moisture content was measured for all samples and mercury content was reported on a dry weight basis. The previous study reported mercury concentrations on a wet weight basis. The average correction factor for converting from wet weight to dry weight basis was 1.25. Therefore, in the following comparison, all values measured in the 1974 study were multiplied by this factor to have a uniform basis for comparison.

In the 1974 study, the Hg concentration values ranged from 1 – 50 ng/g dry weight basis. This is similar to the current study (16.9– 65 ng/g) after discounting the outliers. High values as found in the outliers (up to 155 ng/g) were not observed in the previous study. The 1974 study did not publish all of the data however, they provided average values for four quadrants (NE, NW, SE, and SW). Table 8 presents the average values in these regions from the 1974 and current studies. Both studies show the same relative ranking of quadrants with the NE being the highest followed by NW, SW and SE and both studies had approximately the same difference between the averages of the SW quadrant and the other quadrant averages. The prevailing winds are from the south to the north so this may reflect enhanced deposition. However, the 1974 study reported that there was a slight change in soil type between north and south of the plant, which also could be responsible for the difference. The values for the 2004 study were approximately 25% higher than the 1974 study. This could be due to many different factors other than deposition including analytical techniques, sampling locations, and sampling protocol. The 1974 study was taken from the agricultural fields that are plowed to a depth of 17 cm annually. This would cause mixing and presumably dilution as deposition is a surface process. Mercury content typically decreases with depth from the surface. Also, in the current study an apparent correlation with vehicular traffic along the road was observed and perhaps the difference in average values reflects the difference in sample location (roadside versus field). In any event, it is not possible to determine if the difference between average Hg values in the two studies is meaningful.

3.4.7 Estimated Mass Deposition

Mass deposition estimates were performed for the approximately 400 square kilometer sampling region around the plant. Using a soil density of 1.5 g/cm^3 and limiting the excess deposition to the top 5 cm gives a total volume of soil of 2.0 10^7 m^3 and a soil mass of 3.010^{13} g over the study area. Table 12 presents the excess estimated amount deposited for four cases:

a) using the soil surface average minus the soil subsurface average (0.6 ng/g),

b) the soil surface average minus the value of the soil concentration where 2/3 of the measured values were greater than the measured value (3.7 ng/g),

c) soil average by quadrant minus the lowest soil average by quadrant (Table 11), this approach was used in the 1974 study by Anderson (Anderson, 1977), this approach assumes that the SW quadrant (lowest average quadrant) is not impacted by plant emissions and is representative of background.

d) the estimate based on deposition modeling using ISCST (790 g/yr) multiplied by the operational period (37 years).

Table 9 contains the estimate of deposited mass, the % of RGM emissions represented by this mass, and the % of total emissions that this represents. Total emissions were assumed to be 160 kg/yr multiplied by the 37 year operational period of the plant. Twenty percent of emissions were assumed to be RGM. Total emissions are likely to be higher as an emission estimate of 530 kg was suggested during the early 1970's (Anderson, 1977). Also, prior to 1994 when locally mined coal was burned, the percentage of RGM was likely to be higher than 20%. Both of these assumptions skew the estimate of the % of mercury deposited to higher values. The table shows that for any scenario, less than 9 % of the RGM and less than 2% of the total mercury emissions from the plant deposited within a 20 km square centered on the plant. This analysis also supports the contention that although mercury levels may be slightly elevated near the plant, a 'hot spot' with substantially elevated levels does not occur. The analysis also suggests that greater than 90% of the mercury emitted from the plant enters the regional or global mercury cycle.

The previous study (Anderson, 1977) estimated that "more than 26% and possibly as high as 70% of the mercury emitted by the Kincaid plant" was deposited within a 19.3 km radius of the plant. The cause for this discrepancy in this estimate and the estimates in Table 9 lies in the differing assumptions made in the two analyses. In the 1977 study, the volume of soil was estimated using a depth of 17 cm, the plow depth, even though the samples were only from the top 2 cm. In addition, in that study, the background value was set to the lowest value in the four sampled quadrants, similar to Case B in Table 12.

In any event, the 1977 estimates appear to be biased high. If the current emission rate of 160 kg/yr is used to estimate the lifetime of the plant emissions, deposition of 26% of the Hg within 19.3 km radius would lead to an increase in average soil concentration in the top 5 cm of 17.6 ng/g. If this occurred, the average soil Hg concentrations on a dry weight basis would be expected to be around 40 ng/g, which is substantially higher than observed. In addition, Hg emissions rates in the early part of the operating history of the plant tended to be as much as a factor of three higher than the value used to estimate the amount of Hg that could be deposited. If the total emissions were higher than estimated, the increase in soil concentration would be expected to be even higher.

5. Conclusions

A thorough quantitative understanding of the processes of mercury emissions, deposition, and translocation through the food chain is currently not available. Complex atmospheric chemistry and dispersion models are required to predict concentration and deposition contributions, and aquatic process models are required to predict effects on fish. There are uncertainties in all of these predictions. Therefore, the most reliable method of understanding impacts of coal-fired power plants on Hg deposition is from empirical data.

A review of the literature on mercury deposition around sources including coal-fired power plants found studies covering local mercury concentrations in soil, vegetation, and animals (fish and cows (Lopez et al. 2003). There is strong evidence of enhanced local deposition within 3 km of the chlor-alkali plants, with elevated soil concentrations and estimated deposition rates of 10 times background. For coal-fired power plants, the data show that atmospheric deposition of Hg may be slightly enhanced. On the scale of a few km, modeling suggests that wet deposition may be increased by a factor of two or three over background. The measured data suggest lower increases of 15% or less. The effects of coal-fired plants seem to be less than 10% of total deposition on a national scale, based on emissions and global modeling.

The following summarizes our findings from published reports on the impacts of local deposition. In terms of excesses over background the following increments have been observed within a few km of the plant:

- local soil concentration Hg increments of 30%-60%,
- sediment increments of 18-30%, and
- wet deposition increments of 11-12%.
- fish Hg increments of about 5-6%, based on an empirical finding that fish concentrations are proportional to the square root of deposition.

Important uncertainties include possible reductions of RGM to Hg(0) in power plant plumes and the role of water chemistry in the relationship between Hg deposition and fish content.

Soil and vegetation sampling programs were performed around two mid-size coal fired power plants. The objectives were to determine if local mercury hot spots exist, to determine if they could be attributed to deposition of coal-fired power plant emissions, and to determine if they correlated with model predictions. These programs found the following:

- At both sites, there was no correlation between modeled mercury deposition and either soil concentrations or vegetation concentrations. At the Kincaid plant, there was excess soil Hg along heavily traveled roads. The spatial pattern of soil mercury concentrations did not match the pattern of vegetation Hg concentrations at either plant.

- At both sites, the subsurface (5 – 10 cm) samples the Hg concentration correlated strongly with the surface samples (0-5 cm). Average subsurface sample concentrations were slightly less than the surface samples, however, the difference was not statistically significant.
- An unequivocal definition of background Hg was not possible at either site. Using various assumed background soil mercury concentrations, the percentage of mercury deposited within 10 km of the plant ranged between 1.4 and 8.5% of the RGM emissions. Based on computer modeling, Hg deposition was primarily RGM with much lower deposition from elemental mercury. Estimates of the percentage of total Hg deposition ranged between 0.3 and 1.7%. These small percentages of deposition are consistent with the empirical findings of only minor perturbations in environmental levels, as opposed to "hot spots", near the plants.

The major objective of this study was to determine if there was evidence for "hot spots" of mercury deposition around coal-fired power plants. Although the term has been used extensively, it has never been defined. From a public health perspective, such a "hot spot" must be large enough to insure that it did not occur by chance, and it must affect water bodies large enough to support a population of subsistence fishers. The results of this study support the hypothesis that neither of these conditions have been met.

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6. References

Abbott ML, Susong DD, Olson M, Krabbenhoft DP. Mercury in soil near a long-term air emission source in southeastern Idaho, Environmental Geology (2003) 43:352-356.

Anderson WL, Smith KE. Dynamics of Mercury at a Coal-Fired Power Plant and Adjacent Cooling Lake, Environ Sci Tech,(1977),11:75-80.

Biester H, Muller G, Scholer HF. Estimating distribution and retention of mercury in three different soils contaminated by emissions from chlor-alkali plants, Science of Tot Env (2002),284:177-189.

Biester H, Muller G, Scholer HF. Binding and mobility of mercury in soils contaminated by emissions from chlor-alkali plants, Science of Tot Env(2002a) 284: 191-203.

Cox WM., Rorex HW, Moss GK. *Evaluation of Rural Air Quality Simulation models, Addendum C: Kincaid SO_2 Data Base*, EPA-450/83-003c, U.S. Environmental Protection Agency, Research Triangle Park, NC, March 1986.

Crockett A., Kinnison RH. Mercury Residues in Soil around a Large Coal-Fired Power Plant, Environ Sci Tech (1979) 13:712-5.

Edgerton E, Hartsell B, Jansen J. "Hg Speciation in Coal-Fired Power Plant Plumes: Evidence for Reduction of RGM to HG(0)," 7th Annual Conference on Mercury as a Global Pollutant, Ljubljana, Slovenia, July 2, 2004.

U.S. Environmental Protection Agency, User's Guide for the Industrial Source Complex (ISC3) Dispersion Models, Volume I – User Instructions, EPA-454/B-95-003A, Office of Air Quality Planning and Standards Emissions, Monitoring, and Analysis Division, Research Triangle Park, NC. 1995

U.S. Environmental Protection Agency, Fate and Transport of Mercury in the Environment, Mercury Study Report to Congress, vol. III, EPA-452/R-970995, 1997.

U.S. Environmental Protection Agency, State Mercury Emissions Estimates from Power plants, June 2001, http://www.epa.gov/ttn/atw/combust/utiltox/utoxpg.html#DA1.

Environmental Science and Technology Science Page, "Controversial Results Downplay Power Plant Mercury Emissions," Nov. 3, 2004. http://pubs.acs.org/subscribe/journals/esthag-w/2004/nov/science/rr_mercury.html

Environmental Science and Technology Science Page, "Mercury Trading Scheme Raises Concern," March 4, 2004. http://pubs.acs.org/subscribe/journals/esthag-w/2004/mar/science/kc_mercury.html

Environmental Science and Technology Policy Page, "Mercury Policy Debate Heats Up," July 28, 2004. http://pubs.acs.org/subscribe/journals/esthag-w/2004/jul/policy/kc_mercury.html

Klein DH, Russell P. Heavy Metals: Fallout around a Power Plant, Environ Sci Tech (1973) 7:357-8.

Kotnik J, Horvat M., Mandic V, Logar M. Influence of the Sostanj coal-fired thermal power plant on mercury and methyl mercury concentrations in Lake Velenje, Slovenia, Science Total Environ (2000), 259:85-95.

Laudal D, Dunham G, Levin L. Mercury Control Technology R&D Review, Pittsburgh, PA, July 14 – 15, 2004. http://www.netl.doe.gov/publications/proceedings/04/HgReview/Laudal2.Presentation%20for%20Project%20Review%20Meeting%20Plume%20Study%20-%20Pittsburgh%20July%2014-15%202004%20-%202.pdf

Lodenius M. Dry and wet deposition of mercury near a chlor-alkali plant, Science Tot Environ (1998), 213: 53-56.

Lopez Alonzo M., Benedito JL, Miranda M, Fernandez JL, Castillo C, Hernandez J. Shore, R.F., Environ Pollut (2003), 125:173-81.

Mercury Deposition Network, http://nadp.sws.uiuc.edu/mdn.

Menounou N, Presley BJ. Mercury and other trace elements in sediment cores from central Texas lakes, Arch Environ Contam Toxicol (2003), 45:11-29.

Sensen M, Richardson DHS. Mercury levels in lichens from different host trees around a chlor-alkali plant in New Brunswick, Canada. , Science Tot Environ (2002),293:31-45.

Southworth GR, Lindberg SE, Zhang H. Anscombe, F.R., Fugitive mercury emissions from a chlor-alkali factory; sources and fluxes to the atmosphere, Atmospheric Environment (2004) 38:597-611.

Sullivan TM, Lipfert FD, Morris SC, Moskowitz PD. Potential Health Risk Reduction Arising From Reduced Mercury Emissions from Coal-Fired Power Plants, FY 2001 Report, September, 2001, BNL-71538-2003.

Sullivan TM, Lipfert FD, Morris SM. The Local Impacts of Mercury Emissions from Coal Fired Power Plants on Human Health Risk, Progress Report for the Period of March 2002 – March 2003, BNL-71554-2003, May 2003.

Sullivan TM, Bowerman B, Adams J, Lipfert FD, Morris SM, Bando A, Pena R, Blake R. Mercury Emissions from Coal Fired Power Plants: Local Impacts on Human Health Risk, BNL topical report in press (2005).

Susong DD, Abbott ML, Krabbenhoft DP. “Mercury accumulation in snow on the Idaho National Engineering and Environmental Laboratory and surrounding region, southeast Idaho, USA, Environmental Geology (2003), 43:357-363.

Tack FMG, Vanhaesebroeck T, Verloo MG, Van Rompaey K, Van Ranst E. Mercury baseline levels in Flemish soils (Belgium), " Environmental Pollution (2005),134:173-179.

Tobias FJ, Bech J, Sanchez Algarra P. Statistical Approach to Discriminate Background and Anthropogenic Input of Trace Elements in Soils of Catalonia, Spain, Water Air and Soil Pollution (1997), 100:63-78.

Wang D, Shi X., Wei S. Accumulation and transformation of atmospheric mercury in soil, Science Tot Environ (2003), 300:209-214.

Xiao Z, Smar J, Lindqvist O, Giouleka E. Atmospheric mercury deposition to grass in southern Sweden, Science Tot Environ (1998),213: 85-94.

Table 1 Estimates of mass deposition and fraction of mercury emitted that was deposited for Plant A

Case	Total Mass Deposited (g) over 23 year plant life	% of RGM	% of Hg(total)
Surface vs. Subsurface Avg	2200	0.16	0.023
Lowest 1/3 Avg as background	24500	1.7	0.3
ISCST Model	9400	0.66	0.12

Table 2 Statistical parameters for Vegetation Samples at Plant A.

	Value (ng/g)
Min	10.7
Max	653.3
Median	31.8
Average	96.7 (log mean 1.7)
Standard Deviation	143.5 (Geometric Standard Deviation 0.47)

Table 3 Average vegetation and soil concentrations as a function of distance from the plant.

Distance	# of vegetation samples	Average Hg concentration in vegetation (ng/g)	# of soil samples	Average Hg concentration in soil (ng/g)
160 – 1500 m	5	37.4	5	28.8
1500 – 3000 m	10	179.4	10	27.0
3000 – 4500 m	8	153.4	8	29.9
4500 – 6000 m	10	106.8	10	31.1
6000 –7500 m	13	29.2	13	24.9
7500 – 9000 m	6	23.2	6	30.0

Table 4 Average soil concentration (ng/g) dry at each sample location. Average values for the transects are the bottom and last rows of the table.

ID	A	B	C	D	E	F	G	H	I	J	K	Row Avg.
1						28.5	26.2	33.4	45.3	48.1		36.3
2	27.3		26.4	21.0	36.4		26.2	27.6		38.4	26.0	28.7
3	28.8	35.3	57.4	20.7	30.7	28.5	35.1	26.2	35.0	31.0	36.0	33.2
4	33.1	25.4	31.1	23.2		29.1	27.7	26.1		35.4	27.9	28.8
5	25.5	22.8	37.4	19.3	24.9	27.0	24.9	27.5		25.6	18.5	25.3
6	23.1	33.6	30.9	23.6		29.5	23.0	22.5		23.6	27.0	26.3

7A				27.0	30.6	27.0	32.9		26.9			28.9
7	34.1	24.6	39.2	35.3	63.9	33.8	65.2	42.1	35.4	37.8	39.7	41.0
8	23.9	30.9	23.3	27.4	23.6	43.7	26.3	47.8	25.7	25.0	23.9	29.2
9	23.7	26.1	25.9	20.9	20.0	26.7	20.7	27.4	24.5	25.3	28.5	24.5
10		20.8	24.6	30.8	31.3	27.8	29.6	31.3		26.3	21.1	27.1
11	27.9	22.3	21.8	27.1	26.1	24.5	29.4	25.6	25.9	26.2	22.8	25.4
12			16.9		25.2	27.7	23.8	31.8	23.9	26.1	25.3	25.1
Avg.	27.5	26.9	30.5	25.1	31.3	29.5	30.1	30.8	30.3	30.7	27.0	

Table 5 Mercury concentration statistics for vegetation samples at Kincaid

	Value (ng/g)
Min	0.6
Max	22.5
Median	4.9
Average	5.6
Standard Deviation	3.8
Geometric Standard Deviation	2.0

Table 6 Average Hg concentration in vegetation near the Kincaid Power Plant.

Location	A	B	C	D	E	F	G	H	I	J	K	Row Avg.
1						7.0	3.4	14.1	5.8	0.6		6.2
2	3.7	4.2	6.7	8.6	4.0	4.7	5.5	4.3		0.6	1.4	4.4
3	6.0	4.7	5.5	4.1	9.9	3.3	6.4	8.1	1.5	1.3	8.1	5.4
4	3.1	5.0	2.2	5.0		5.7	8.2	19.6		3.5	3.3	6.2
5	2.2	2.4	5.1	2.5	10.5	2.9	7.0	5.8		2.0	2.0	4.2
6	22.3	3.9	6.6	2.7		6.9	4.8	5.4		3.4	6.3	6.9
7A				1.0	5.1	5.5	3.2		4.7			3.9
7	8.4	7.7	3.8	3.8	4.7	6.1	1.7	4.2	6.5	5.6	4.5	5.2
8	0.9	22.5	1.2	10.6	3.9	6.1	3.6	11.3	5.0	7.6	1.5	6.7
9	2.4	3.8	1.8	5.9	2.3	6.8	2.4	14.8	6.5	2.4	5.9	5.0
10	13.0	4.7	5.0	8.0	2.0	7.2	7.4	8.8	5.2	1.9	7.9	6.5
11	8.3	2.9	4.4	9.8	7.0	6.6	11.3	10.7	4.2	3.7	7.9	7.0
12				4.4	9.2	1.9	7.6	2.6	4.6	10.7	2.9	5.5
Average	7.0	6.2	4.2	5.5	5.8	5.4	5.6	9.1	4.9	3.6	4.7	

Table 7 Soil and vegetation mercury concentrations as a function of distance.

Distance (miles)	# Soil Samples	Average Soil Hg (ng/g)	# Vegetation Samples	Average Vegetation Hg (ng/g)
0 – 1	2	38.7	2	5.3
1 – 2	7	35.3	9	5.2
2 – 3	15	30.4	13	6.1
3 – 4	12	27.1	14	6.1
4 - 5	25	27.4	26	5.4
5 - 6	19	28.3	23	5.2
6 - 7	22	30.4	22	5.8
7 - 8	12	26.9	12	6.2
8 - 9	3	29.3	3	6.0

Table 8 Average soil mercury concentrations by quadrant.

Quadrant	Average Soil Hg (ng/g dry) 2004 Study	Average Soil Hg (ng/g dry) 1974 Study	Difference from SW Quadrant (ng/g)	
			1974	2004
SW	25.8	18.8	----	----
SE	26.8	20.0	1.2	1.0
NW	30.9	23.8	5.0	5.1
NE	31.8	27.5	8.7	6.0

Table 9 Mercury mass deposition estimates around the Kincaid Plant based on soil data and ISCST modeling

	Excess (ng/g)	Total Mass Deposited (kg)	% of RGM	% of Hg(total)
Case A: Surface avg. minus subsurface avg.	0.6	16.4	1.4	0.28
Case B: Surface average minus soil concentration at 1/3 of the distribution	3.7	101	8.5	1.7
Case C: Quadrant average minus lowest quadrant average	1.0 (SE-SW) 5.1 (NW-SW) 6.0 (NE-SW)	6.8 34.8 41.0 Total – 82.6	7.0	1.4
ISCST Deposition	N/A	29.2	2.5	0.5

Figure 1 Modeled total Hg deposition (µg/m^2/y) pattern around plant A, located at (0,0). Second plant in the lower left corner is responsible for the deposition pattern in this region.

Figure 2 Soil and vegetation sample locations around the power plant.

Figure 3 Comparison of predicted deposition (ug/m^2/yr) (filled contours) and measured soil Hg concentrations (ng/g) (symbols)

Figure 4 Measured mercury levels in vegetation compared to deposition modeling predictions.

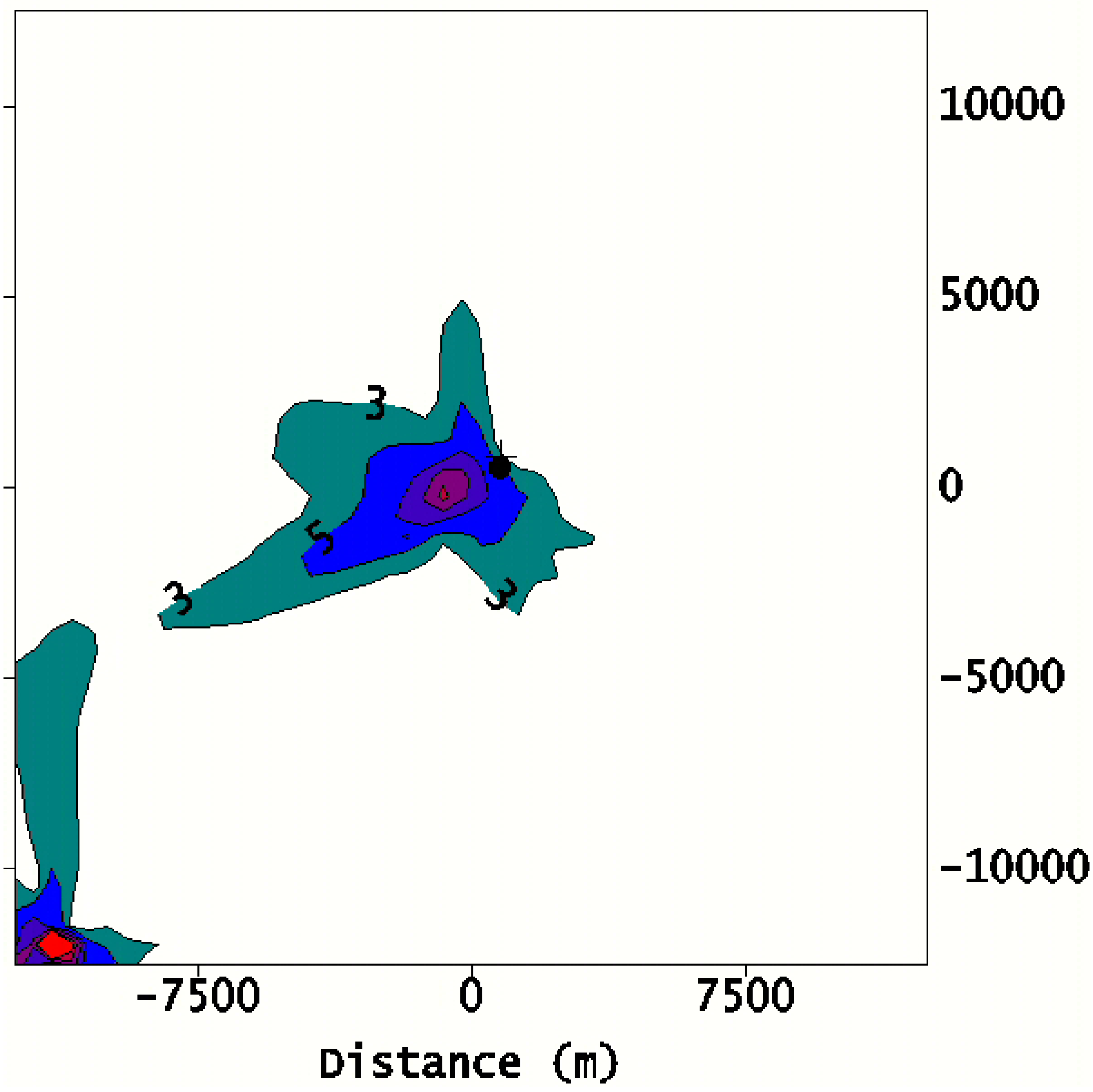
Figure 5 Predicted mercury deposition wet and total deposition contours from emissions from the Kincaid power plant.

Figure 6 Sample grid around the Kincaid Power Plant.

Figure 7 Comparison of measured soil Hg data and predicted Hg deposition (filled contours).

Figure 8 Soil Hg levels (ng/g dry) posted on local area map. Plant located between G7 and H7.

Figure 9 Vegetation Hg levels around the Kincaid Power Plant.



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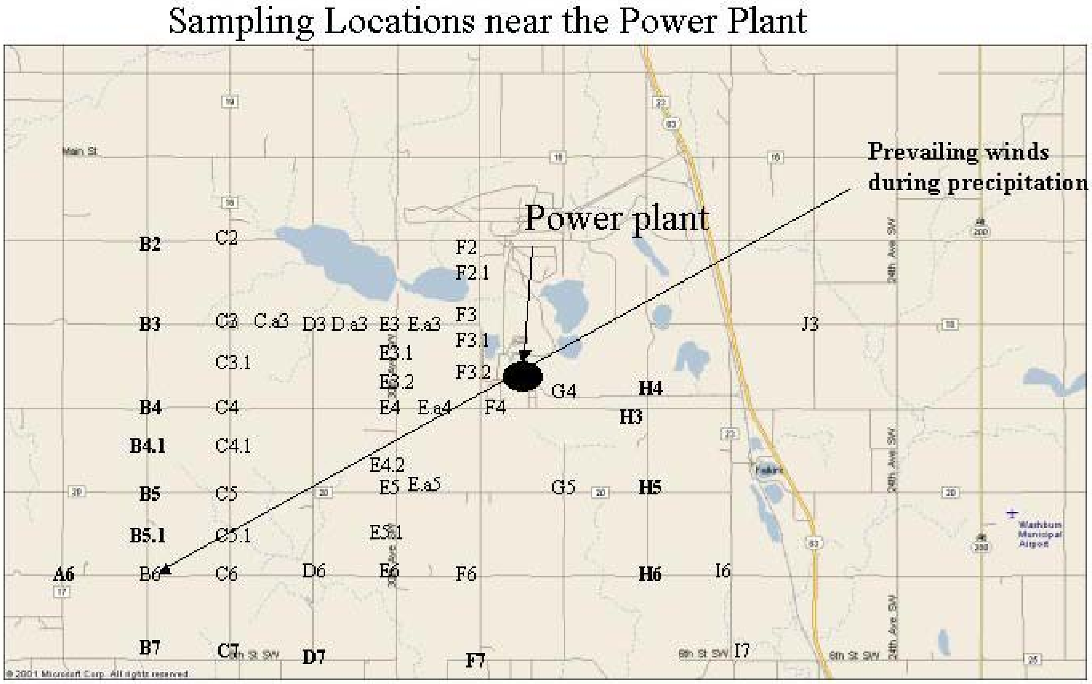
Sampling Locations near the Power Plant

Figure 2 Soil and vegetation sample locations around the power plant.

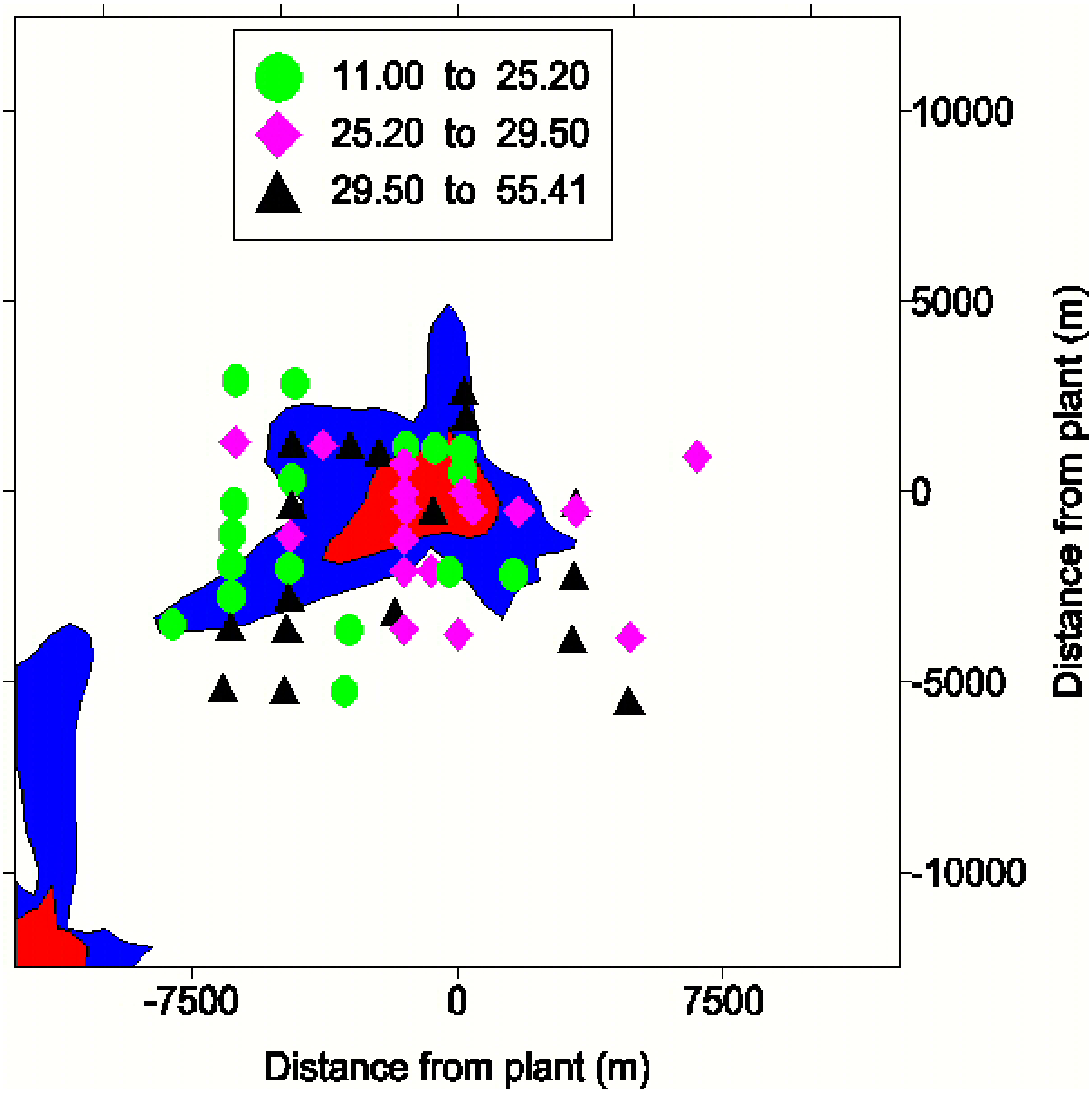

Figure 3 Comparison of predicted deposition (ug/m^2/yr) (filled contours) and measured soil Hg concentrations (ng/g) (symbols)

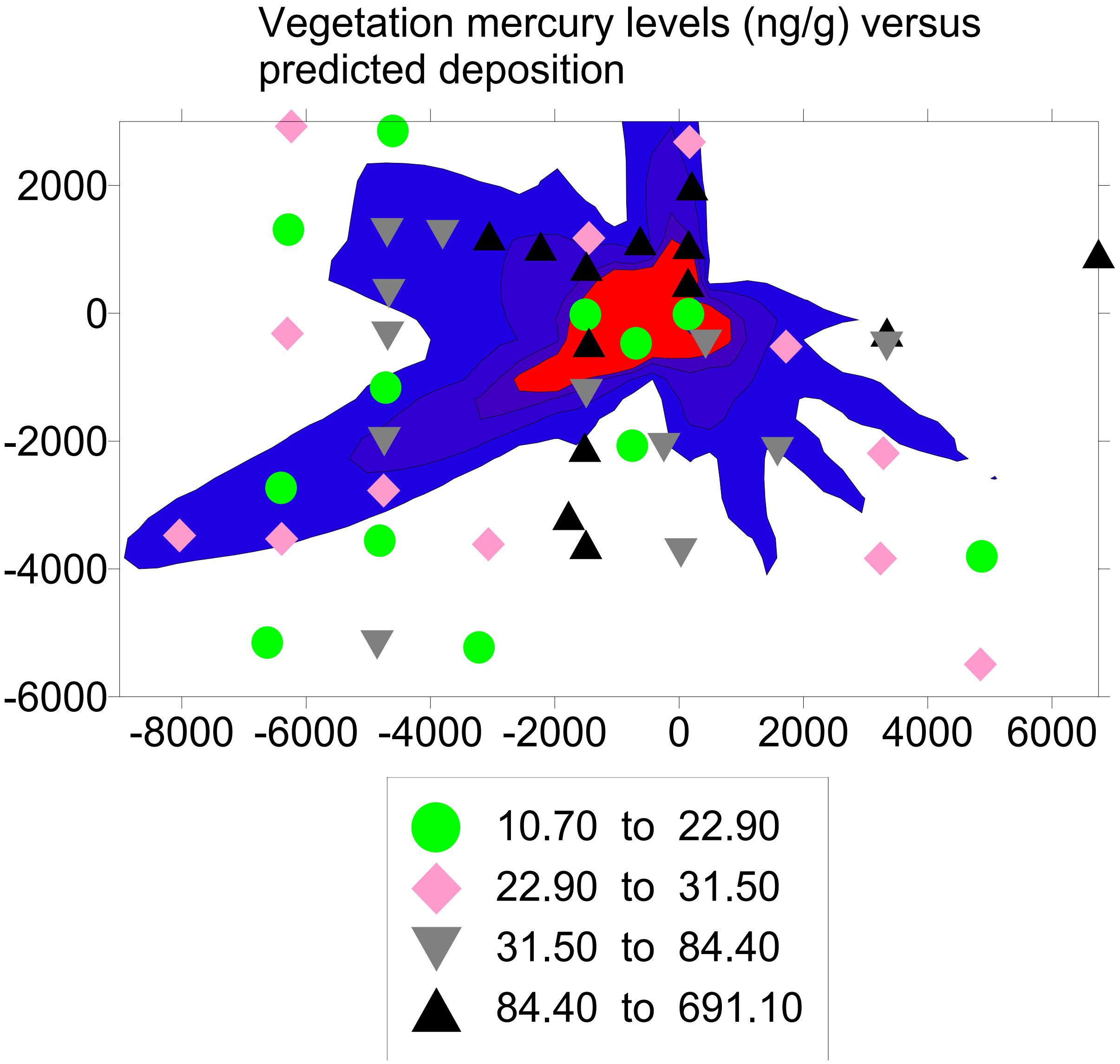

Figure 4 Measured mercury levels in vegetation compared to deposition modeling predictions.

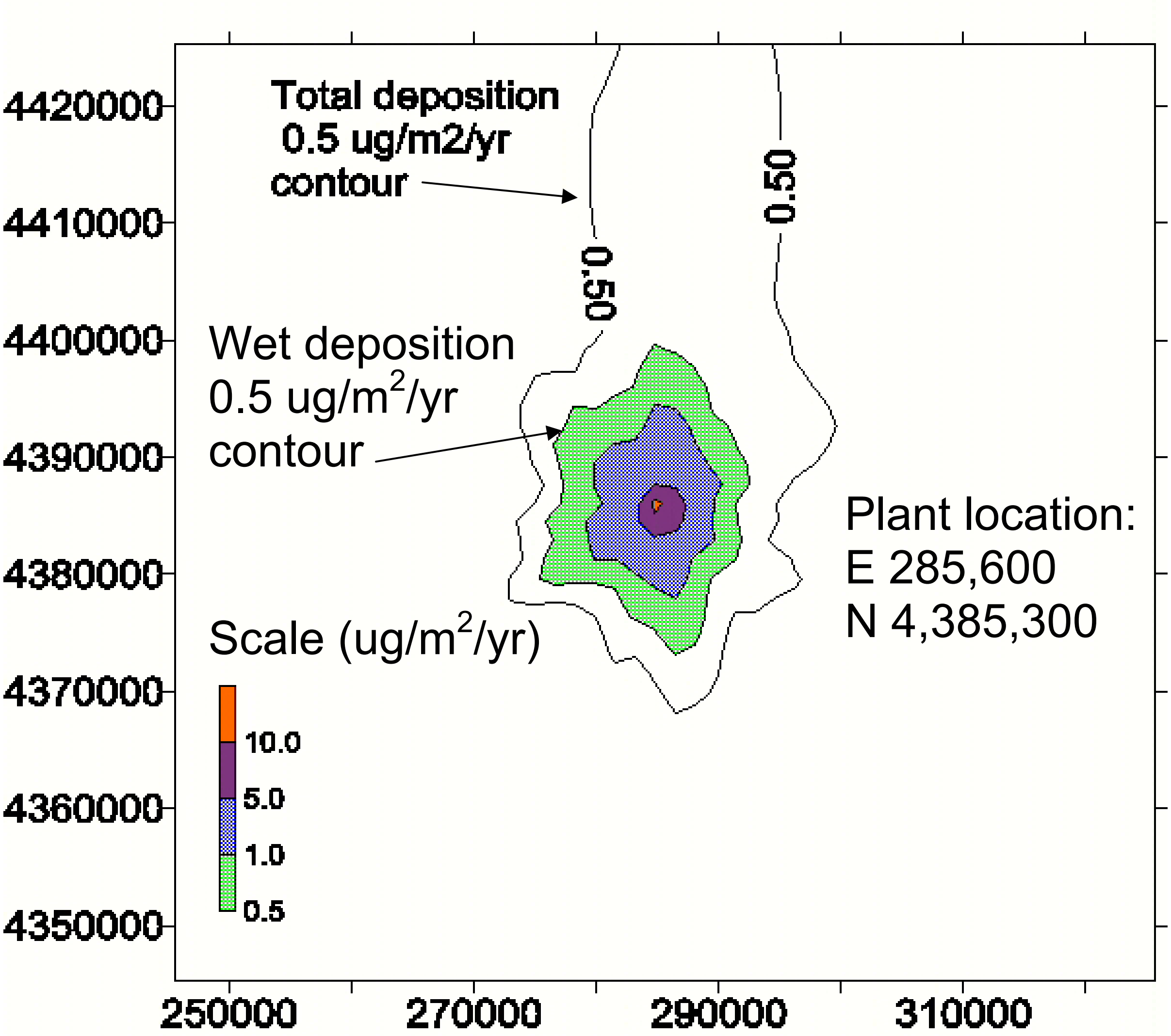

Figure 5 Predicted mercury deposition wet and total deposition contours from emissions from the Kincaid power plant.

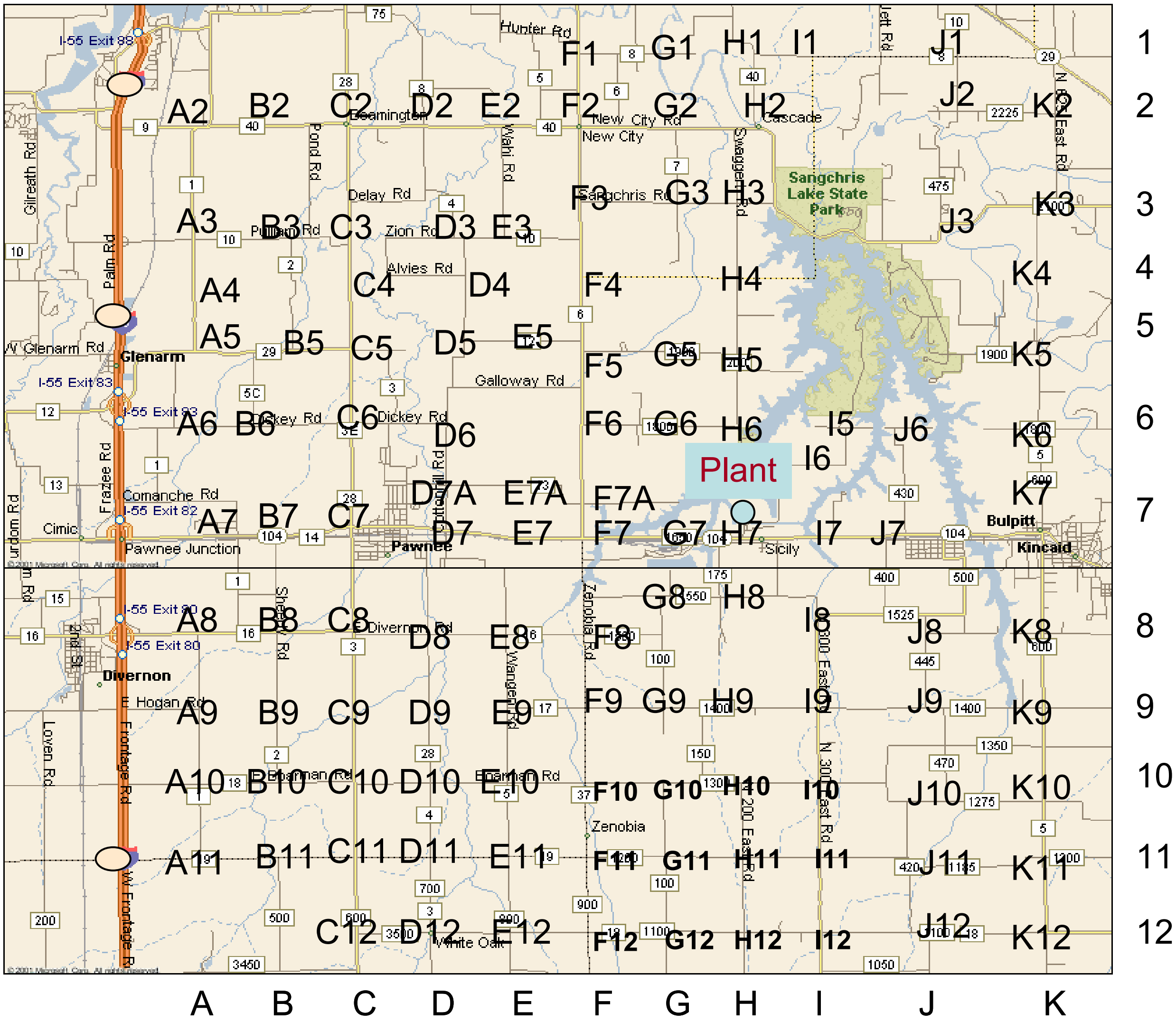

Figure 6 Sample grid around the Kincaid Power Plant.

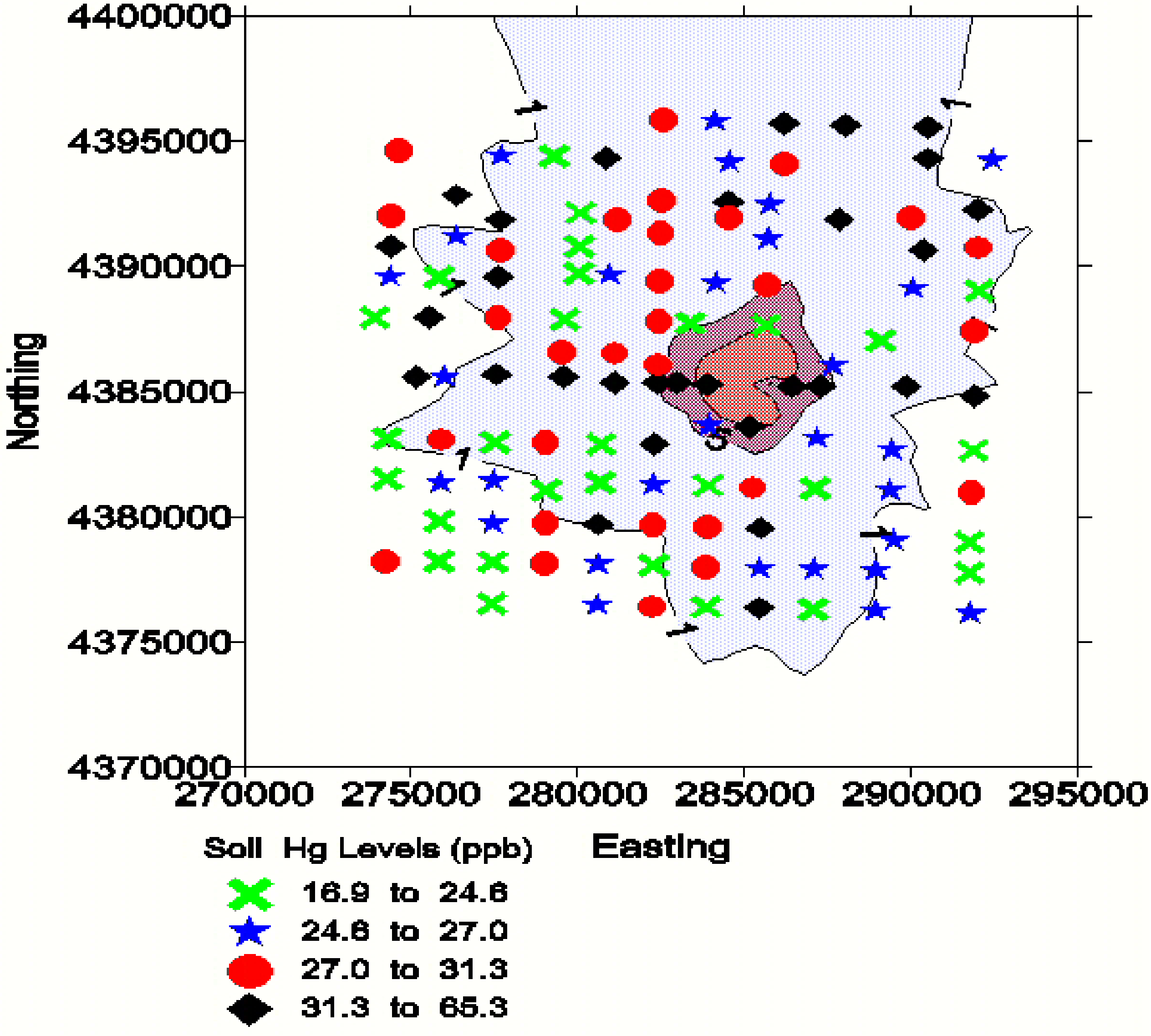

Figure 7 Comparison of measured soil Hg data and predicted Hg deposition (filled contours).

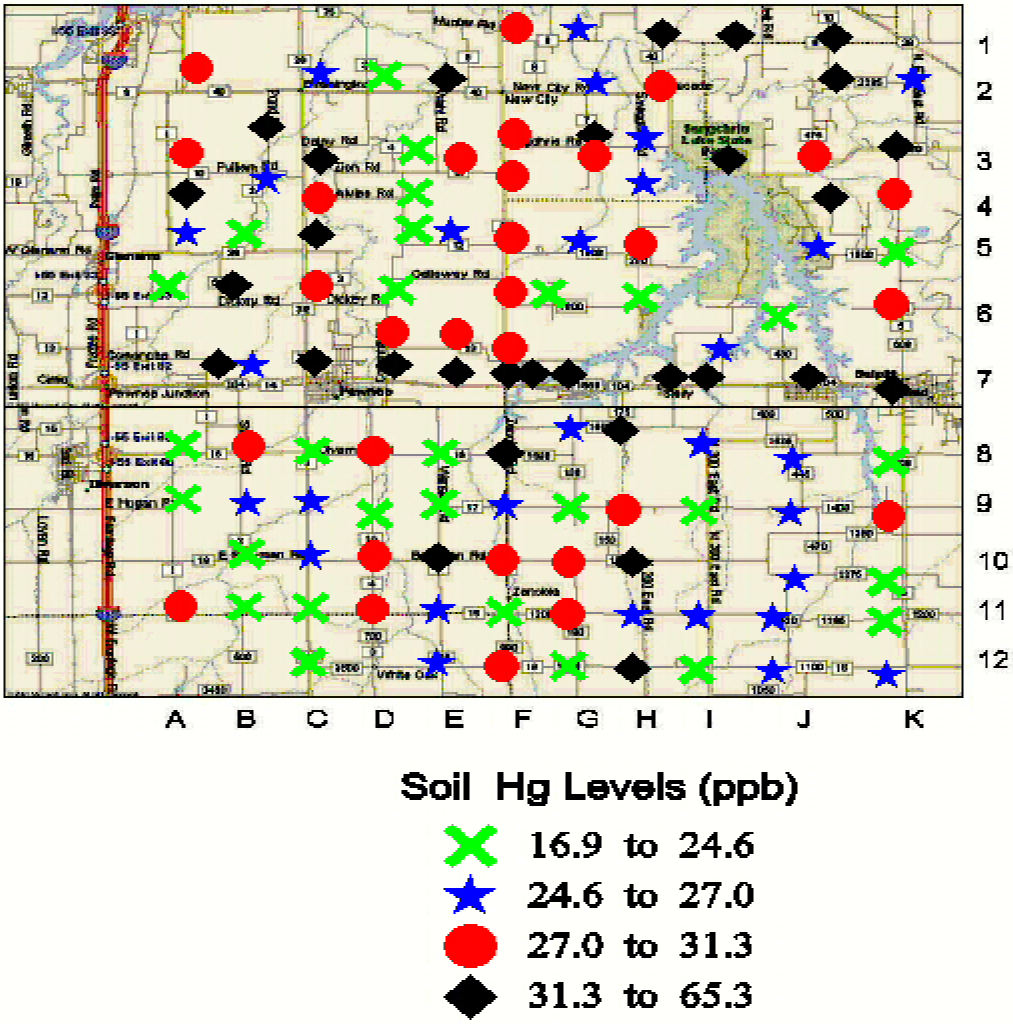

Figure 8 Soil Hg levels (ng/g dry) posted on local area map. Plant located between G7 and H7.

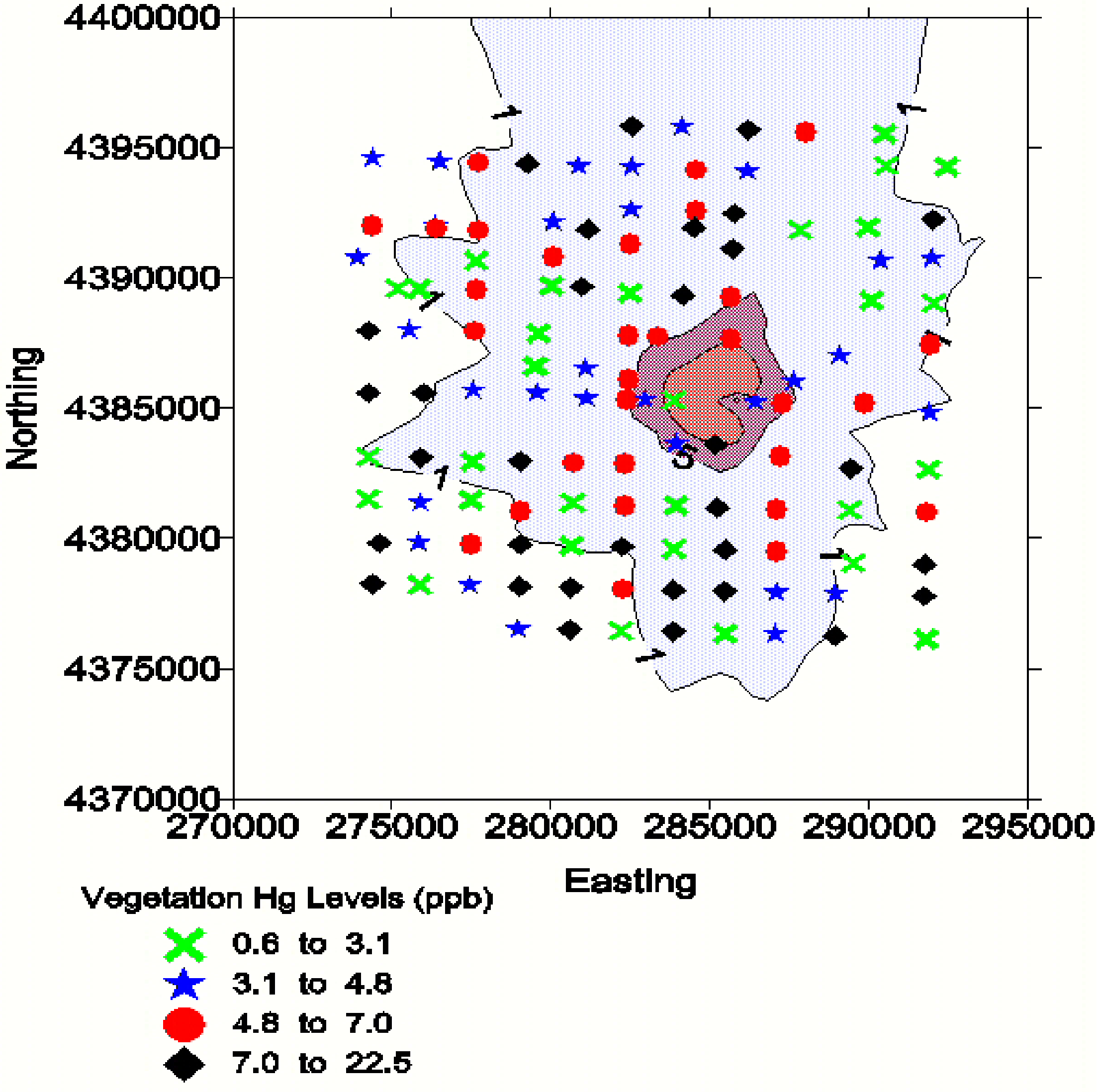

Figure 9 Vegetation Hg levels around the Kincaid Power Plant.