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MERCURY DISTRIBUTION IN SOIL AROUND A LARGE COAL-FIRED POWER PLANT

Environmental Monitoring and Support Laboratory
Office of Research and Development
U.S. Environmental Protection Agency
Las Vegas, Nevada 89114

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FOREWORD

Protection of the environment requires effective regulatory actions which are based on sound technical and scientific information. This information must include the quantitative description and linking of pollutant sources, transport mechanisms, interactions, and resulting effects on man and his environment. Because of the complexities involved, assessment of specific pollutants in the environment requires a total systems approach which transcends the media of air, water, and land. The Environmental Monitoring and Support Laboratory-Las Vegas contributes to the formation and enhancement of a sound integrated monitoring data base through multidisciplinary, multimedia programs designed to:

- develop and optimize systems and strategies for monitoring pollutants and their impact on the environment
- demonstrate new monitoring systems and technologies by applying them to fulfill special monitoring needs of the Agency's operating programs

The report presents the data collected in a study of mercury residues in soil around a large coal-fired power plant. Detailed soil sampling and analyses indicate low residue levels and no statistically significant differences in mercury distribution patterns within 30 kilometers (km) of the plant. These data will be considered by EPA and others in making decisions concerning regulations of mercury emissions from coal-fired utilities. For additional information, please contact the Pollutant Pathways Branch, Environmental Monitoring and Support Laboratory-LV, P. O. Box 15027, Las Vegas, Nevada 89114.

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INTRODUCTION

The toxicity and hazards of mercury are well established and its distribution throughout the environment has been documented (Peakall and Lovett, 1972). Due to the known hazards, a number of reports have focused on the sources of mercury resulting from man's activities. The combustion of coal is often cited as a significant mercury source and various emission estimates have been calculated (Horn, 1975; Joensuu, 1971; Billings and Matson, 1972).

The amount of mercury released to the atmosphere by coal-fired electric utilities can be estimated from the amount of coal consumed using the average mercury content of the coal and a release factor. Coal combustion by the electric utilities in the United States was about 365 X 10^9 kilograms (kg) in 1975, and is projected to increase to 854 X 10^9 kg by the year 2000 (Merrit, 1976). The mercury level in U.S. coal has been estimated from different data bases to average 1,000 parts per billion (ppb) (Joensuu, 1971), 300 ppb (Billings and Matson, 1972), 200 ppb (Horn, 1975), and 150 ppb (Magee et al., 1973). The latter figure, representing the most recent data, was compiled from the largest data base and is probably the more accurate. Not all mercury in coal is released during coal combustion and a reasonable release factor appears to be 0.9 (Billings et al., 1973; Kalb, 1975; Bolton et al., 1973). Therefore, the 1975 estimated mercury release from U.S. coal-fired electric utilities was about 49,000 kg.

The fate of this released mercury has not been extensively studied. Klein and Russell (1973) studied heavy metal fallout around a 650 megawatts (MW) coal-fired power plant equipped with a 120-meter (m) stack. They state that the soils around the plant are enriched in mercury, but present no statistical proof. Bolton et al. (1973) conducted a major study of trace elements at the 870 MW coal-fired Allen Steam Plant and found no significant evidence of elemental accumulation. Cannon and Anderson (1972) reported on elements around the Four Corners Power Plant in New Mexico, but showed no significant elevation of mercury levels in plants or soil. The lack of statistical treatment, the small number of samples involved, and the terrain and background levels of mercury limit the usefulness of these studies. For these reasons, and as a part of a project to select a biological monitor, an investigation was conducted to determine whether a mercury gradient exists around coal-fired power plants.

The selection of a power plant was based upon the estimated potential for contamination. This report focuses on the soil sampling portion of this study. All samples were collected in December 1974.

The Four Corners Power Plant was selected because of its size (>2,150 MW), its coal consumption (6.3 X 10^9 kilograms per year (kg/y)), its length of operation (since 1963), and its relatively short stacks (two are 76 m and two are 91 m). In addition, the surrounding arid terrain (15 to 20 centimeters per year (cm/y) percipitation) is undisturbed by cultivation and the soils are low in most elements including mercury.

Estimates of the mercury concentration of the coal burned by the plant vary considerably. Based upon a very limited sampling, Billings et al. (1973) estimated the mercury concentration to be 300 ppb. Green and Robinson (1971) sampled five coal blending piles as well as the mine and found the average mercury concentration to be 375 ppb. However, the U.S. Geological Survey analyzed 21 coal samples from the mine and reported an average value of 90 ppb (Swanson, 1972). The latter analyses were made using proven modern methods in an attempt to provide precise quantitative data under a strong quality control program. Assuming the figure of 90 ppb to be the most accurate, it can be calculated that the Four Corners Power Plant emitted over 1 percent of all mercury released by coal-fired power plants in the United States for 1974.

CONCLUSIONS

Detailed soil sampling and analyses for mercury residues indicate no significant differences in mercury residue levels within 30 km of a large coal-fired power plant. Other recent data indicated emissions of 589 and 1,372 kg of mercury by a coal-fired power plant and a smelter, respectively, and pose no health hazard relative to air contamination (Horn, 1975). These data should be considered in making decisions concerning regulation of mercury emissions from coal-fired power plants. Horn (1975) indicates that U.S. coal-fired utilities emit 9 percent of all human-related releases to the atmosphere in the U.S. In 1974, utility emissions amounted to only 4 percent of the natural degassing mercury loss in conterminous United States (calculated from Horn, 1975).

SAMPLING AND ANALYSIS

The sampling design selected was a radial grid employing 16 evenly spaced radii and five logarithmically spaced circles, concentric around the power plant (Figure 1). The radii of the circles (A-E) were 1.0, 2.9, 6.8, 14.6, and 30 kg. From the 80 sites on the grid, only 70 composite soil samples were collected since some sites fell on stripped land or in the cooling pond. Each sample was a composite of 10 subsamples collected 15 m apart. The upper 1 to 2 cm of soil was collected by trowel and stored in pint glass jars.

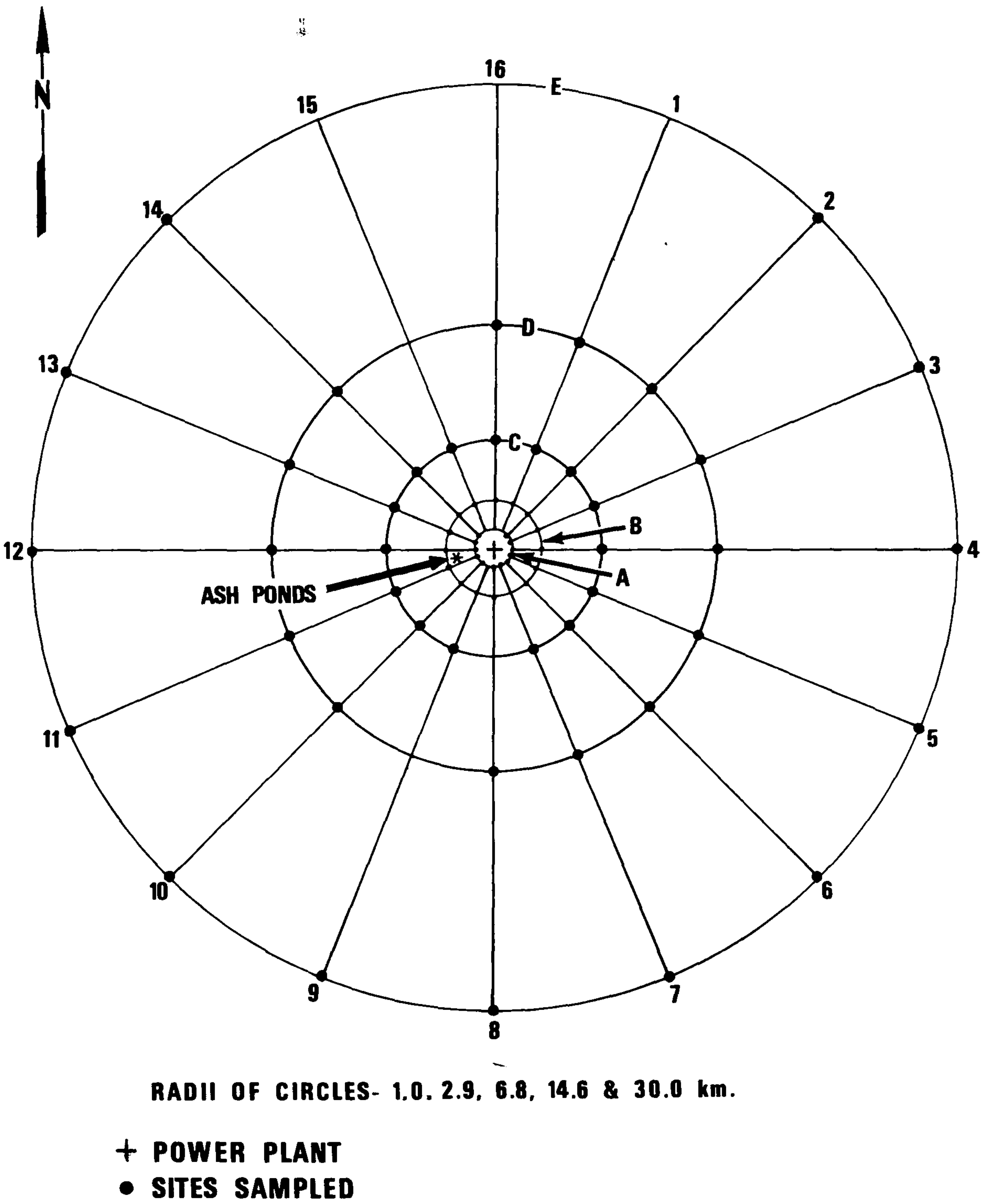

Figure 1. Soil sampling sites around the Four Corners Power Plant

The samples were dried at 60°C, sieved to 10 mesh, and 5 replicates of 150 to 200 milligrams (mg) were analyzed using an Isotope Atomic Absorption spectrometer (Hadeiski and McLaughlin, 1975). The geometric mean of the replicate standard deviations was 1.9 ppb with a range of 0.2 to 21 ppb. The standard deviation of the replicates could probably have been reduced if the samples had been pulverized to homogenize the sample. Eleven percent of the samples was randomly selected and reanalyzed for quality control purposes. Of the eight reruns, seven were within ±1 ppb of the original value while the eighth was off by 7 ppb.

The standard regression line of nanograms of mercury versus the digital millivolt reading was calculated using 60 analyses of five liquid standards and a blank. The regression was verified using a U.S. Environmental Protection Agency liquid standard and two National Bureau of Standards (NBS) Standard Reference Materials, orchard leaves (#1571) and coal (#1632). The lower limit of detection was 5 ppb.

RESULTS

The overall arithmetic mean (the data did not exhibit significant departure from normality) for the 70 samples was 16 ppb with a range of 6 to 45 ppb and an overall standard deviation of 6.7 ppb (see Table 1). Initially the data were subjected to a two-way analysis of variance test using the analytical replicates. The residual error was therefore a measure of laboratory precision only. Since the program used cannot accommodate missing samples, six of the radii were dropped from the data analysis. The analysis of variance on the ten remaining radii showed the following:

1. A highly significant difference between circles (i.e., soil mercury levels are related to distance from the power plant).

2. A highly significant difference between radii (i.e., soil mercury levels are related to compass direction from the power plant).

3. A highly significant interaction effect (i.e., the type of effect due to radii varies significantly from circle to circle, or the type of effect due to circles varies significantly from radii to radii).

Attempts at subdividing the data to eliminate the interaction effects were not successful.

Since it was possible that the analytical error was insignificant relative to field sampling error, a second approach was attempted. A nested two-way analysis of variance was employed in which radii were grouped in twos or fours which provided a residual error term composed of both analytical and field sampling errors. The grouping of radii was conducted since it was felt that small changes in compass direction would have an insignificant effect on residue levels. The results of these analyses of variance were

TABLE 1. MEAN MERCURY RESIDUES IN SOIL BY SITE (PPB)

Radius		Concentric Circle						
Number	Direction	A	B	C	D	E	Mean	Standard Deviation
1	NNE	--	6	12	10	--	9.3	3.1
2	NE	45	9	12	13	19	20	15
3	ENE	30	13	13	11	9	15	8.4
4	E	22	11	10	11	9	13	5.3
5	ESE	21	13	10	10	12	13	4.5
6	SE	16	14	10	14	11	13	2.4
7	SSE	23	--	9	6	14	13	7.4
8	S	12	20	--	14	11	14	4.0
9	SSW	14	20	12	--	12	14	3.8
10	SW	15	26	14	21	14	18	5.3
11	WSW	27	22	23	14	21	21	4.7
12	W	20	24	15	13	12	17	5.1
13	WNW	24	23	18	22	17	21	3.1
14	NW	16	15	16	26	11	17	5.5
15	NNW	--	14	13	--	--	14	0.7
16	N	--	7	23	30	--	20	1.2
MEAN		22	16	14	15	13	16	
STANDARD DEVIATION		8.7	6.3	4.4	6.8	3.7		6.7

-- = no sample.

Radii of circles A - E are 1.0, 2.9, 6.8, 15 and 30 km, respectively.

similar to those encountered previously. Highly significant effects due to circles, radii and interaction were again discovered. Since the interaction effect could not be eliminated, it must be concluded that the distribution of mercury residues around the plant is very complex. Therefore, no statement based on statistical analysis can be made concerning the relationship of mercury residue levels relative to direction or distance from the plant.

In an attempt to discern a distribution pattern of mercury, a plot was made of mean mercury residues by site. Lines of iso-mercury residues were then plotted as shown in Figure 2. Aside from a couple of small scattered sites exceeding 20 ppb, there are three areas with levels exceeding 20 ppb. The area due west of the plant may be related to the ash ponds located in this section. The 25 ppb high to the north-northwest cannot be accounted for by the wind direction, but another power plant (350 MW, 13 k north-northeast) is located in that area. The samples with the highest mercury levels detected were collected from an undisturbed island located in the cooling pond, sites 2A and 3A.

DISCUSSION

The 70 soil samples collected on a radial grid around the Four Corners Power Plant probably represent the most intensive sampling effort to date at that site. No statistically significant differences in mercury residue levels were detected although some distribution patterns are evident. The levels of mercury detected are quite low, but in good agreement with soil data compiled by Cannon and Anderson (1972). They reported an average value of 0.02 (20 ppb) ppm compared to 16 ppb in this study. Shacklette et al. (1971) reported the average mercury concentration of the Western United States to be 83 ppb which is nearly twice as high as the maximum and five times higher than the mean value reported here.

The question then arises as to the fate of mercury emissions from the plant. The estimated annual release of mercury through stack emissions is 510 kg based upon a release factor of 0.9, a coal consumption of 6.3×10^9 kg/y and a mercury concentration of 90 ppb. This value is in agreement with Arizona Public Services' (1975) emission estimate of 76 grams per hour (g/h) plus or minus a factor of two for the plant operating at full load. Converting and using an operating factor of 70%, the estimate becomes 466 kg of mercury per year. The total amount of mercury emitted by the plant since it began operating is about 3,600 kg.

If all this mercury was evenly deposited within 5 or 10 km of the plant and remained in the top 2.54 cm of soil, the average soil concentration would have increased by 122 ppb or 30 ppb, respectively. The average mercury levels in soils around the power plant are considerably below these figures. Obviously, the emitted mercury is ultimately being deposited over a much larger area. It is possible, however, that mercury is being deposited and rapidly revolatilized in aerobic terrestrial environments (Rogers, 1975).

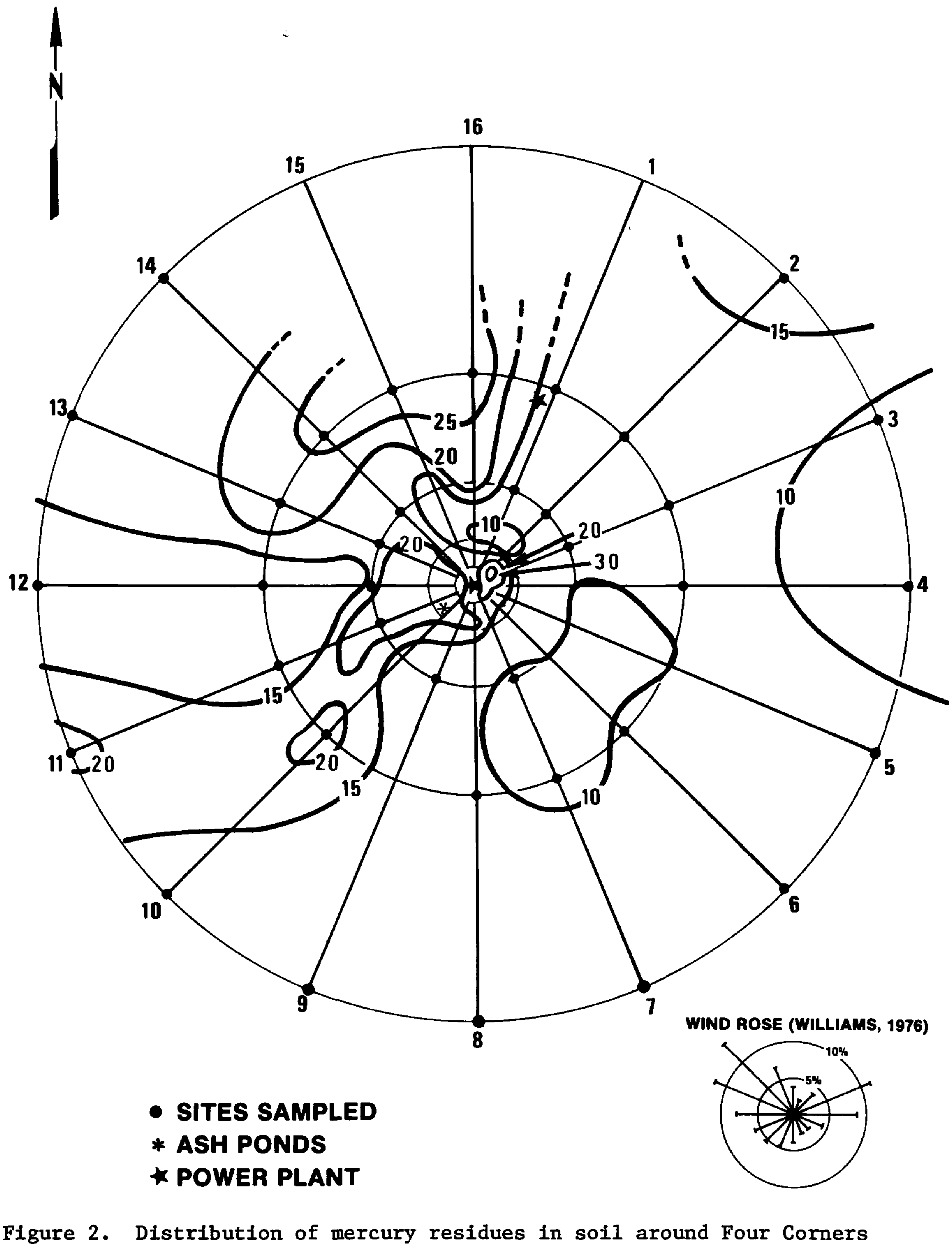

Figure 2. Distribution of mercury residues in soil around Four Corners Power Plant (PPB)

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16. ABSTRACT

Seventy soil samples were collected on a radial grid employing sixteen evenly spaced radii and five logarithmically spaced circles, concentric around the Four Corners power plant. The soil samples were analyzed for total mercury using a Zeeman Atomic Absorption spectrophotometer. Residue levels were quite low compared to average soil residues and no statistically valid differences in mercury residue levels were detected between circles or radii using two-way analysis of variance techniques. F-ratios indicated: significant differences between circles, significant differences between radii, and significant complex interaction which could not be eliminated. Contours of iso-mercury concentrations show a relative high west of the plant near the ash ponds and another just east of the plant. The fate of the 510 kg of mercury emitted per year is not known, but it is not accumulating near the plant. Mercury emissions by U.S. coal-fired power plants amount to only 4% of the natural degassing loss in the U.S., and levels near power plants appear low. The significance of mercury emissions by power plants should be evaluated on a regional basis since the evidence shows no significant local elevation of mercury in soils or air.

17. KEY WORDS AND DOCUMENT ANALYSIS

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