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**TRACE ELEMENTS IN THE TERRESTRIAL ENVIRONMENT
OF A COAL-FIRED POWERHOUSE**

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PREPARED FOR THE U.S. ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION UNDER CONTRACT AT(07-2) 1

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Printed in the United States of America

Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road
Springfield, Virginia 22161

Price: Printed Copy \$5.00; Microfiche \$3.00

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OF A COAL-FIRED POWERHOUSE**

by

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Publication Date: September 1977

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ABSTRACT

A coal-fired powerhouse at the Savannah River Plant, Aiken, South Carolina operated for more than 20 years without electrostatic precipitators and consumed about 360,000,000 kilograms of coal per year. Twenty-nine trace elements were measured in fly ash, and in samples of soil, vegetation, and ground water collected along a 29-km traverse centered on the powerhouse. There were statistically significant effects of the stack releases upon the concentration of Ba, Be, Cu, Hg, Mn, Se, and Sr in soil; Be, Co, Mo, Sr, and V in vegetation; and Co and Mn in ground water. The Ba, Hg, Sr, and Mn in fly ash are less available to plants than the same elements in soil.

This report was submitted in fulfillment of ERDA/EPA Interagency Agreement EPA-IA GD5-B681 (Subagreement 78BCC). Work was completed as of September 30, 1977.

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INTRODUCTION

The Savannah River Plant (SRP) at Aiken, South Carolina, has operated seven coal-fired powerhouses since 1952. The largest of these powerhouses, located in 400-D Area (Figure 1), consumes about 360,000,000 kilograms of coal per year. This powerhouse consists of four units of the pulverized-wet bottom type and burns bituminous coal. Each unit has a 38-meter stack. From startup until late 1975, this powerhouse operated without electrostatic precipitators and depended upon mechanical collectors to remove fly ash before combustion products were released to the air. The mechanical collectors were at best about 75% efficient. Electrostatic precipitators, installed on each of the four stacks during late 1975 and early 1976, are greater than 99% efficient for removing fly ash. This study estimated the quantities of trace elements released in the fly ash from this powerhouse before the electrostatic precipitators were added and determined the influence of these elements upon the surrounding terrestrial ecosystems. These ecosystems are typical for much of the coastal plains of the southeastern United States.

The trace elements measured were antimony (Sb), arsenic (As), barium (Ba), beryllium (Be), bromine (Br), cadmium (Cd), cerium (Ce), cesium (Cs), chromium (Cr), cobalt (Co), copper (Cu), europium (Eu), gallium (Ga), lanthanum (La), lead (Pb), manganese (Mn), mercury (Hg), molybdenum (Mo), nickel (Ni), samarium (Sm), scandium (Sc), selenium (Se), strontium (Sr), terbium (Tb), thorium (Th), uranium (U), vanadium (V), ytterbium (Yb), and zinc (Zn).

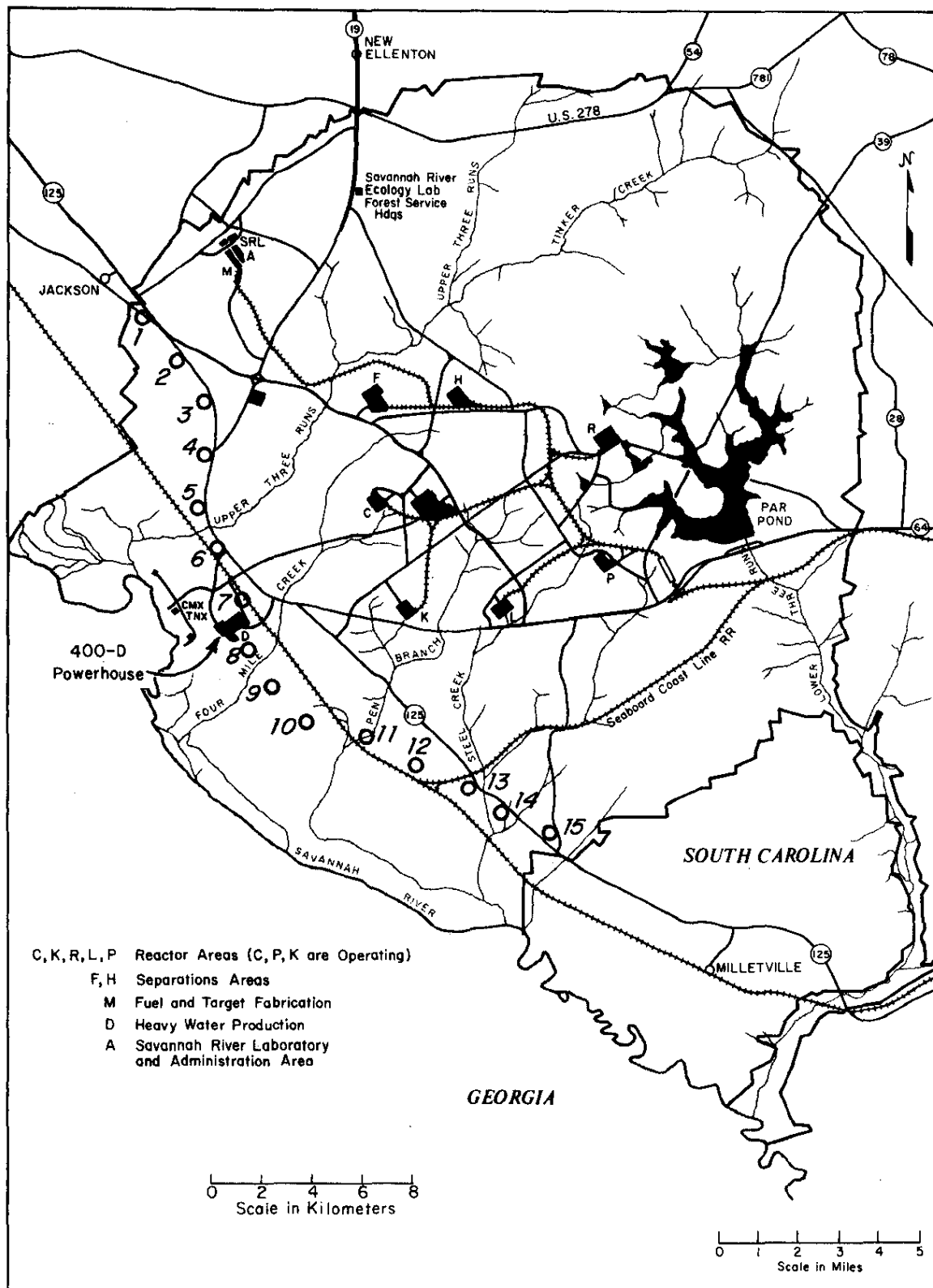


Figure 1. Environmental sampling locations

CONCLUSIONS

Of 29 trace elements measurable in fly ash, 10 produced statistically significant effects upon the concentration in soil, vegetation, or ground water. These were:

Ba, Be, Cu, Hg, Mn, Se, and Sr in soil

Be, Co, Mo, Sr, and V in vegetation

Co and Mn in ground water

Of the trace elements released from the powerhouse stacks, only two are present in the environment in concentrations that could require further consideration. These are Mn in ground water and Se in soil. Both of these are due to naturally high environmental concentrations that have been accentuated by the powerhouse releases of fly ash.

About one-half of the Hg in coal burned in the 400-D Area powerhouse is in the fly ash removed by the electrostatic precipitators. Apparently, the conditions in this powerhouse are conducive to the condensation of Hg on the fly ash because studies at other sites generally conclude that more than 90% of the Hg in coal is released from the stacks as a vapor.

Ba, Hg, Sr, and Mn in the fly ash released from the 400-D Area powerhouse and deposited on the surrounding soil are less available to vegetation than are the same elements naturally occurring in Savannah River Plant soils. Many of the other elements may fall in this same category, but the availability of these elements was not evident because the quantities deposited were so much smaller than those naturally occurring in the soil.

RECOMMENDATIONS

Further efforts are needed to determine the fraction of Hg in coal that is removed from the flue gases with fly ash. The conclusion from other studies that nearly all Hg is released as a vapor may be based more on the known volatility of Hg than upon data collected in those studies because flue gases are very difficult to sample. If some power plants remove a much greater fraction of the Hg in fly ash than others, determining the conditions that cause this difference may be useful.

The leachability of trace elements in fly ash in soil and their availability to vegetation should be studied more extensively. This information must be determined before the consequences of trace element deposition in soil can be evaluated for stack releases or for disposal from electrostatic precipitators or other collectors.

MATERIALS AND METHODS

ESTIMATION OF TRACE ELEMENTS RELEASED

The fly ash content of gases emitted from the mechanical collectors was measured twice during August 1969, with a dust collection sample train manufactured by Glass Innovations, Inc. Corresponding measurements were made with gases entering the electrostatic precipitators during October and December 1975. There were a total of seven tests. Nine samples of fly ash from the electrostatic precipitators, fly ash from the mechanical collectors, bottom ash, and coal entering the boilers were collected during November and December 1975 for trace element analysis.

COLLECTION OF ENVIRONMENTAL SAMPLES

Environmental samples were collected at 15 locations shown in Figure 1. Because there are six smaller coal-fired powerhouses on the Savannah River Plant site, samples were collected at locations affected most by the powerhouse being studied and least by the others. Atmospheric dispersion and deposition calculations were used to select sampling locations and to estimate the 400-D Area powerhouse effects on these locations (Appendix A). At Location 15, the most distant from the 400-D Area powerhouse, the contribution by the 400-D Area powerhouse to total fly ash deposition was 71.5%. At Location 7, the nearest to the 400-D Area powerhouse, the contribution of the 400-D Area powerhouse was greater than 99%.

Vegetation samples included broom sedge (*Andropogon virginicus*), dog fennel (*Eupatorium compositifolium*), and panic grass (*Panicum aciculare*) collected at each of the 15 locations on September 2 and 3, 1975. Each sample was collected over an area of 0.25 to 0.5 hectare. The vegetation was dried at 110°C and ground to pass a 1-mm screen. The vegetation was resampled for Hg analysis on October 19, 1976, and was air dried before grinding to pass a 1-mm screen.

Three soil core samples were collected about 15 m apart at each location on September 9 and 10, 1975. With a split-barrel sampler, cores were taken in segments of 0 to 7.5 cm, 7.5 to 15 cm, and 15 to 30 cm. The samples were air dried and sieved with a 2-mm sieve.

To collect ground water samples, holes were drilled (during the period August 11 and 19, 1976) to a depth of one or two meters below the water table at each of the 15 sampling locations. A spiral steel auger was used, and no casing or screen was placed into the holes. After the water became clear, 15 liters were removed from each hole with a steel bucket. The hole at Location 15 was drilled in very sandy soil and caved so quickly that water could not be obtained. The depth drilled to reach ground water ranged from 1.8 to 10.5 m. The depth at each location is shown in Table 1.

Table 1. DRILLING DEPTHS TO REACH GROUND WATER

| <i>Location^a</i> | <i>Depth, m</i> |
|-----------------------------|-----------------|
| 1 | 10.5 |
| 2 | 2.9 |
| 3 | 1.8 |
| 4 | 2.9 |
| 5 | 8.8 |
| 6 | 3.5 |
| 7 | 1.9 |
| 8 | 3.5 |
| 9 | 2.0 |
| 10 | 5.1 |
| 11 | 1.8 |
| 12 | 5.8 |
| 13 | 5.6 |
| 14 | 3.5 |
| 15 | Hole caved |

^a Locations are shown in Figure 1.

TRACE ELEMENT ANALYSIS METHODS

Analytical methods required to measure the elements of interest in the different types of samples collected included plasma source emission spectrometry, ^{252}Cf neutron activation, flameless atomic absorption spectrometry, conventional atomic absorption spectrometry, and conventional spectrophotometric methods. These methods and their applicability to the various types of samples are described below.

Plasma Source Emission Spectrometry

In plasma source emission spectrometry, an inductively coupled plasma torch provides an ultra-stable heat source for the vaporization and excitation of metallic elements in aqueous solution. The excited atoms of the respective elements subsequently emit their excitation energy as discrete wavelengths of electromagnetic radiation (light) characteristic of the emitting atom. The emitted light enters the spectrometer through a narrow slit and is separated into its component wavelengths by a diffraction grating. The diffracted light is brought to focus at the focal plane of the optical system — and in this way — is detected and measured by photocells.

Water samples were analyzed directly by the plasma source emission spectrometer. Solid samples (vegetation, soil, coal, and coal ash) were completely dissolved prior to analysis in an acid digestion bomb that provides for rapid dissolution at increased pressures. The digestion solution used in the acid bomb to effect dissolution was $\text{HF-HClO}_4\text{-HNO}_3$.

Plasma source emission spectrometry analyses were provided by Barringer Research of Ontario, Canada. The following elements were determined in all samples: Ba, Sr, Mn, Zn, V, Cr, Cu, Ni, Co, and Be.

^{252}Cf Neutron Activation

The neutron activation was performed with an approximately 100-milligram ^{252}Cf source in the Savannah River Laboratory Neutron Activation Facility. Analyses were limited to solid samples. In the analyses, 10-gram aliquots were weighed into high-density polyethylene irradiation containers and irradiated for about 2 weeks at a neutron flux of 10^7 . After irradiation, samples were allowed to decay and were then measured by gamma spectrometry. The following elements were determined: Ce, Ga, La, Br, Sc, Th, Sb, Sm, Cs, U, Eu, Yb, and Tb.

Conventional Atomic Absorption Spectrometry

In atomic absorption spectrometry, characteristic radiation of the element to be determined is produced by a hollow cathode or other type of vapor discharge lamp. Light from the lamp passes through a flame into a monochromator where the particular wavelengths of interest are isolated and focused onto a photodetector. Metallic elements in solution are injected into the flame where they are vaporized and their compounds are dissociated into atoms. The free unexcited atoms in the flame absorb the characteristic radiation from the vapor discharge lamp and attenuate its intensity. The degree of attenuation is a function of the concentration of the element in solution.

All conventional atomic absorption analyses were performed by Barringer Research. For water samples, Cr, Co, Cu, Mo, Ni, Cd, Zn, Pb, and Se were determined. The determinations were by direct analysis except for Se, which required preconcentration by precipitation followed by dissolution and atomic absorption analysis. For solid samples, dissolution was effected by the acid digestion bomb as described earlier, and atomic absorption analyses were made for Pb, Sr, Cd, and Mo.

Flameless Atomic Absorption Spectrometry

Mercury in all samples was measured by flameless atomic absorption spectrometry at the Savannah River Laboratory. In this analysis, samples were subjected to a dual oxidation step — permanganate oxidation in the presence of nitric acid and sulfuric acid and persulfite oxidation — to solubilize and oxidize the different Hg species. After oxidation and conversion to mercuric ion, excess permanganate was reduced by hydroxylamine and mercuric ion was reduced to metallic mercury with stannous chloride. Mercury vapor was aerated through an optical cell in the spectrometer sample beam where it was observed and measured.

Conventional Spectrophotometry

In conventional spectrophotometry, the concentration of a constituent in solution is determined by measuring the relative absorbance or transmittance of light of a definite wavelength through a solution containing the constituent. At Barringer Research, arsenic was measured by a spectrophotometric procedure based on the red complex that is formed when arsenic is reduced to arsine and is subsequently absorbed in a solution of silver diethyldithiocarbamate and pyridene. Water samples as well as dissolved solid samples were measured for arsenic by this technique.

STATISTICAL ANALYSES

The calculated fly ash deposition at each of the environmental sampling locations versus distance from the powerhouse stacks approximates a straight line as shown in Figure 2. Curvilinear regression and correlation analysis show a correlation coefficient r of -0.99 . Therefore, if fly ash deposition of trace elements has been sufficient to produce a statistically significant effect in environmental samples, then a significant negative correlation coefficient should be obtained by curvilinear regression and correlation analysis. An extremely large change in the concentration of a given trace element with fly ash particle size could prevent a significant correlation between the logarithm of concentration and the logarithm of distance. A comparison of the concentrations in the two types of fly ash (mechanical collector and electrostatic precipitator) indicates that very few of the trace elements are likely to exhibit extremely large concentration differences with particle size.

Analysis of variance was used to determine differences in the concentration of the trace elements in the different types of ashes, different depths of soil, and different species of vegetation. When a significant difference existed, a least significant difference (LSD) at the 0.05 level of significance was calculated.

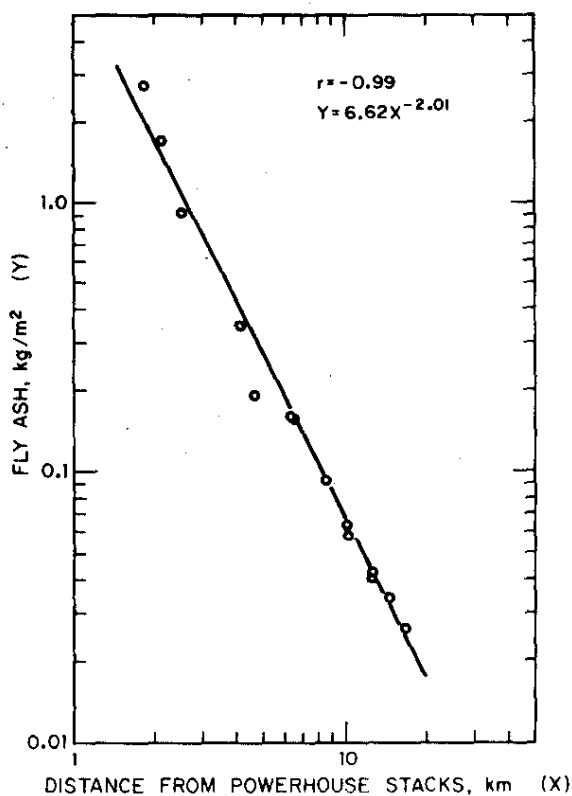


Figure 2. Fly ash deposition

RESULTS

TRACE ELEMENTS IN ASHES AND COAL

A mass balance for the trace elements in the powerhouse was not made because the main emphasis of this study was to determine the influence of the powerhouse stack releases upon the environment. However, the concentrations of trace elements in coal, bottom ash, fly ash from the mechanical collectors, and fly ash from the electrostatic precipitators were determined so that the data would be available if needed to interpret the environmental concentrations. The concentrations of trace elements in coal and different types of ashes are listed in Table 2.

Concentrations of Ba, Cd, Ce, Eu, La, Ni, Sm, and Th in the three types of ashes were statistically the same. Similar results were reported by Klein et al.¹ for Ba, Ce, Eu, La, Sm, Th, Co, Sc, Sr, and Mn. However, our study showed that Co, Sc, and Sr concentrations were higher in fly ash than in bottom ash, and Mn was higher in bottom ash than in fly ash. The following elements were significantly higher in the electrostatic precipitator fly ash than in either the mechanical collector fly ash or bottom ash: As, Be, Br, Co, Cu, Ga, Hg, Mo, Pb, Sb, Se, Sc, U, V, and Zn. Similar results were reported by Klein et al.¹ for As, Cu, Ga, Pb, Sb, Se, Zn, and Cd; in our study, Cd concentration did not differ significantly between the different ashes.

The concentrations of Hg in coal and bottom ash are similar to those reported by Kaakinen et al.² and Kalb,³ but the concentrations in fly ash are higher than those that they reported. Generally the Hg in coal is considered to be emitted from the stack as a vapor.¹⁻⁶

The results of our study indicate that about 50% of the Hg is in the fly ash collected by the electrostatic precipitator; this is a much larger percentage than is generally believed. However, because in other studies a good mass balance was not obtained^{1,2,5} and because flue-gas streams are difficult to sample, our results are probably accurate. If our results are accurate, Hg releases have been substantially reduced at the Savannah River Plant 400-D Area powerhouse with the installation of the electrostatic precipitators.

Table 2. TRACE ELEMENTS IN COAL AND THE DIFFERENT TYPE OF ASHES
(ppm)

| Element | Fly ash | | Bottom ash | LSD ^a | Coal |
|---------|----------------------------|----------------------|------------|------------------|------|
| | Electrostatic precipitator | Mechanical collector | | | |
| Ba | 889 | 792 | 808 | NSD | 130 |
| Sr | 579 | 589 | 333 | 170 | 71 |
| Mn | 352 | 275 | 811 | 376 | 40 |
| Zn | 280 | 116 | 95 | 86 | 23 |
| V | 218 | 166 | 140 | 51 | 27 |
| Ce | 189 | 251 | 150 | NSD | 22 |
| Cr | 171 | 140 | 160 | 20 | 35 |
| As | 164 | 55 | 4 | 44 | 3 |
| Cu | 130 | 93 | 67 | 29 | 16 |
| Ni | 89 | 87 | 77 | NSD | 18 |
| Ga | 72 | 32 | 20 | 12 | 7 |
| La | 69 | 61 | 61 | NSD | 9 |
| Co | 67 | 47 | 40 | 14 | 6 |
| Pb | 60 | 28 | 5 | 14 | 5 |
| Br | 47 | 12 | 3 | 19 | 15 |
| Sc | 32 | 28 | 20 | 5 | 4 |
| Th | 23 | 24 | 25 | NSD | 3 |
| Sb | 19 | 6 | 3 | 5 | 2 |
| Mo | 18 | 11 | 7 | 5.6 | 2 |
| Be | 16 | 12 | 9 | 4 | 2 |
| Sm | 15 | 13 | 12 | NSD | 2 |
| Se | 15 | 6 | 3 | 6 | 2 |
| Cs | 14 | 13 | 10 | 3 | 2 |
| U | 13 | 8 | 8 | 4 | 2 |
| Eu | 11 | 12 | 8 | NSD | 6 |
| Yb | 12 | 8 | 10 | 2 | 1.4 |
| Tb | 2.5 | 2.1 | 2.0 | NSD | 1.5 |
| Hg | 0.84 | 0.33 | 0.08 | 0.37 | 0.06 |
| Cd | 0.71 | 0.39 | 0.50 | NSD | <0.5 |

^a

LSD — least significant difference at the 0.05 level of significance.
NSD — no significant difference at the 0.05 level of significance.

STACK RELEASES AND GROUND DEPOSITION

During the seven stack-gas sample tests, the fly ash released averaged 40 ± 15 grams per kilogram of coal burned. From these data and the annual coal consumption, the annual fly ash releases shown in Figure 3 were calculated. After all four boilers in the 400-D Area powerhouse began operating in late 1952, fly ash releases were reasonably constant at 20.9×10^6 kg/yr through 1957; from 1957 through 1975, releases have averaged about 14.5×10^6 kg/yr. During the 23 years of operation without electrostatic precipitators, an estimated total of 3.6×10^8 kg of fly ash were released. The total deposition during this period at each of the 15 environmental sampling locations was estimated with the deposition rates shown in Appendix A and the total fly ash released. Results of these estimates are shown in Figure 4. The maximum fly ash deposition, 2.75 kg/m^2 , was at Location 7, and the minimum, 0.026 kg/m^2 , was at Location 15. From these depositions, the effect of the powerhouse stacks upon trace element concentrations in the environment would be expected to decrease rapidly as distance from the stack increases.

Twenty-nine trace elements were measured in the fly ash as shown in Table 2. The concentrations in fly ash ranged from 890 ppm for Ba to 0.7 ppm for Cd. The total release from 1952 through 1975 as shown in Table 3 ranged from 320,400 kg for Ba to 250 kg for Cd. The Hg released in fly ash was 290 kg, but if 90% of the Hg in the coal burned was released as is generally thought, then the total release would be 500 kg.

Calculated total depositions for each trace element at each environmental sampling location are listed in Table 4. The depositions at Location 7 ranged from 2440 mg/m^2 for Ba to 1.95 mg/m^2 for Cd. At Location 15, the depositions ranged from 24 mg/m^2 for Ba to 0.02 mg/m^2 for Cd.

Table 3. TRACE ELEMENTS RELEASED FROM POWERHOUSE STACKS IN FLY ASH

| <i>Element</i> | <i>Stack releases, kg</i> | <i>Element</i> | <i>Stack releases, kg</i> | <i>Element</i> | <i>Stack releases, kg</i> |
|----------------|---------------------------|----------------|---------------------------|----------------|---------------------------|
| Ba | 320,400 | Ga | 25,900 | Sm | 5,400 |
| Sr | 208,800 | La | 24,800 | Se | 5,400 |
| Mn | 126,000 | Co | 24,100 | Cs | 5,000 |
| Zn | 100,800 | Pb | 21,600 | U | 4,700 |
| V | 79,200 | Br | 16,900 | Eu | 4,000 |
| Ce | 68,400 | Sc | 11,500 | Yb | 4,000 |
| Cr | 61,200 | Th | 8,300 | Tb | 960 |
| As | 57,600 | Sb | 6,500 | Hg | 290 |
| Cu | 46,800 | Mo | 6,500 | Cd | 250 |
| Ni | 32,000 | Be | 5,800 | | |

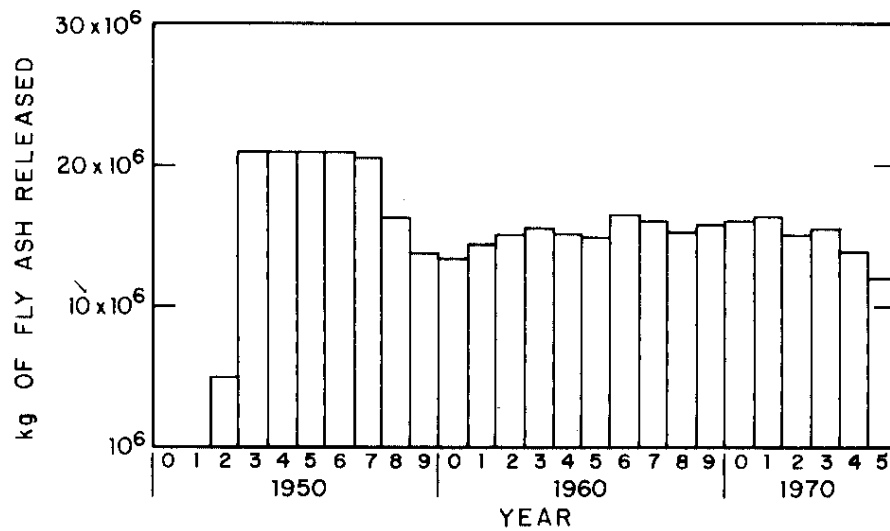


Figure 3. Annual fly ash releases by powerhouse

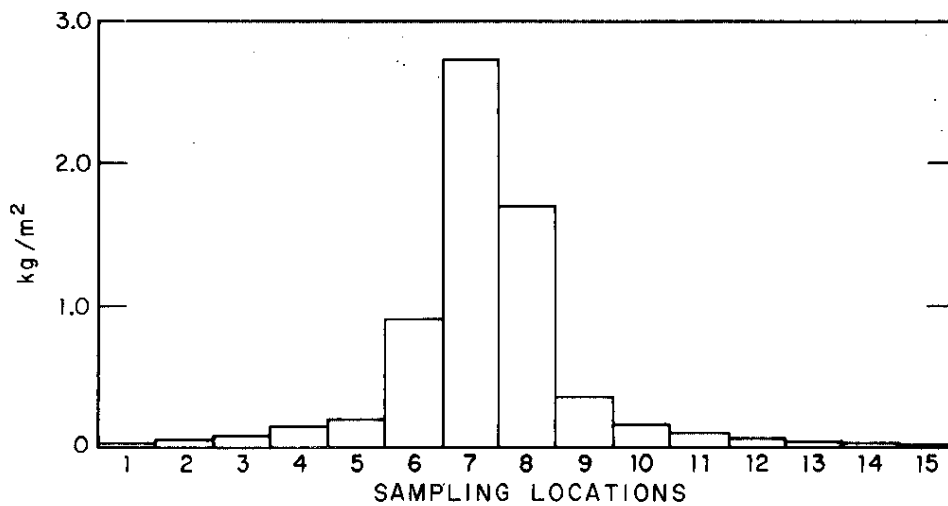


Figure 4. Total fly ash deposition at each environmental sampling location (1952 through 1975)

Table 4. TOTAL DEPOSITION OF EACH TRACE ELEMENT IN FLY ASH AT EACH ENVIRONMENTAL SAMPLING LOCATION
(ng/m²)

| <i>Sampling Locations</i> | | | | | | | | | | | | | | | |
|---------------------------------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|------|
| <i>Element</i> | 1 | 2 | 3 | 4 | 5 | 6 | 7 | 8 | 9 | 10 | 11 | 12 | 13 | 14 | 15 |
| Ba | 38 | 52 | 83 | 139 | 171 | 814 | 2440 | 1510 | 310 | 147 | 89 | 57 | 36 | 30 | 24 |
| Sr | 25 | 34 | 54 | 90 | 112 | 530 | 1590 | 980 | 200 | 96 | 58 | 37 | 24 | 20 | 15 |
| Mn | 15 | 21 | 33 | 55 | 68 | 320 | 960 | 600 | 123 | 58 | 35 | 23 | 14 | 12 | 9 |
| Zn | 12 | 16 | 26 | 44 | 54 | 256 | 770 | 475 | 98 | 46 | 28 | 18 | 12 | 9 | 7 |
| V | 9 | 13 | 20 | 34 | 42 | 200 | 600 | 370 | 76 | 36 | 22 | 14 | 9 | 7 | 6 |
| Ce | 8 | 11 | 18 | 29 | 36 | 173 | 520 | 320 | 66 | 31 | 19 | 12 | 8 | 6 | 5 |
| Cr | 7 | 10 | 16 | 27 | 33 | 157 | 470 | 290 | 60 | 28 | 17 | 11 | 7 | 6 | 5 |
| As | 7 | 10 | 15 | 26 | 32 | 150 | 451 | 278 | 57 | 27 | 17 | 11 | 7 | 6 | 4 |
| Cu | 6 | 8 | 12 | 20 | 25 | 119 | 357 | 220 | 45 | 22 | 13 | 8 | 5 | 4 | 3 |
| Ni | 4 | 5 | 8 | 14 | 17 | 82 | 245 | 151 | 31 | 15 | 9 | 6 | 4 | 3 | 2 |
| Ga | 3.1 | 4.2 | 6.7 | 11 | 14 | 66 | 198 | 122 | 25 | 12 | 7.2 | 4.6 | 3.0 | 2.4 | 1.9 |
| La | 3 | 4 | 6.4 | 11 | 13 | 63 | 190 | 117 | 24 | 11 | 6.9 | 4.4 | 2.8 | 2.3 | 1.8 |
| Co | 3 | 4 | 6 | 10 | 13 | 61 | 184 | 114 | 23 | 11 | 7 | 4 | 3 | 2.3 | 1.8 |
| Pb | 2.6 | 3.5 | 6 | 9 | 12 | 55 | 165 | 102 | 21 | 10 | 6 | 4 | 3 | 2 | 1.6 |
| Br | 2.0 | 2.8 | 4.4 | 7.3 | 9 | 43 | 129 | 80 | 16 | 7.8 | 4.7 | 3.0 | 1.9 | 1.6 | 1.2 |
| Sc | 1.4 | 1.9 | 3.0 | 5.0 | 6.2 | 29 | 88 | 54 | 11 | 5.3 | 3.2 | 2.0 | 1.3 | 1.1 | 0.8 |
| Th | 1.0 | 1.3 | 2.1 | 3.6 | 4.4 | 21 | 63 | 39 | 8 | 3.8 | 2.3 | 1.5 | 0.9 | 0.8 | 0.6 |
| Sb | 0.8 | 1.1 | 1.7 | 2.8 | 3.5 | 16 | 49 | 31 | 6.3 | 3.0 | 1.8 | 1.2 | 0.7 | 0.6 | 0.5 |
| Mo | 0.8 | 1.1 | 1.7 | 2.8 | 3.5 | 17 | 50 | 31 | 6.3 | 3.0 | 1.8 | 1.2 | 0.7 | 0.6 | 0.5 |
| Be | 0.7 | 0.9 | 1.5 | 2.5 | 3.1 | 15 | 44 | 27 | 5.6 | 2.6 | 1.6 | 1.0 | 0.7 | 0.5 | 0.4 |
| Sn | 0.6 | 0.9 | 1.4 | 2.3 | 2.9 | 14 | 41 | 25 | 5.0 | 2.5 | 1.5 | 1.0 | 0.6 | 0.5 | 0.4 |
| Se | 0.6 | 0.8 | 1.4 | 2.3 | 2.9 | 14 | 41 | 25 | 5.2 | 2.5 | 1.5 | 1.0 | 0.6 | 0.5 | 0.4 |
| Cs | 0.6 | 0.8 | 1.3 | 2.2 | 2.7 | 13 | 38 | 24 | 4.9 | 2.3 | 1.4 | 0.9 | 0.6 | 0.5 | 0.4 |
| U | 0.6 | 0.8 | 1.2 | 2.0 | 2.5 | 12 | 36 | 22 | 4.5 | 2.1 | 1.3 | 0.8 | 0.5 | 0.4 | 0.3 |
| Eu | 0.5 | 0.6 | 1.0 | 1.7 | 2.1 | 10 | 30 | 19 | 3.8 | 1.8 | 1.1 | 0.7 | 0.5 | 0.4 | 0.3 |
| Yb | 0.5 | 0.6 | 1.0 | 1.7 | 2.1 | 10 | 30 | 19 | 3.8 | 1.8 | 1.1 | 0.7 | 0.5 | 0.4 | 0.3 |
| Tb | 0.1 | 0.15 | 0.2 | 0.4 | 0.5 | 2.3 | 6.9 | 4.2 | 0.9 | 0.4 | 0.3 | 0.2 | 0.1 | 0.08 | 0.07 |
| Hg | 0.03 | 0.05 | 0.07 | 0.12 | 0.15 | 0.74 | 2.20 | 1.35 | 0.28 | 0.13 | 0.08 | 0.05 | 0.03 | 0.03 | 0.02 |
| Cd | 0.03 | 0.04 | 0.07 | 0.11 | 0.14 | 0.65 | 1.95 | 1.20 | 0.24 | 0.12 | 0.07 | 0.05 | 0.03 | 0.02 | 0.02 |
| <i>Fly ash, g/m²</i> | 43 | 59 | 93 | 156 | 193 | 920 | 2750 | 1690 | 348 | 165 | 101 | 64 | 41 | 34 | 26 |

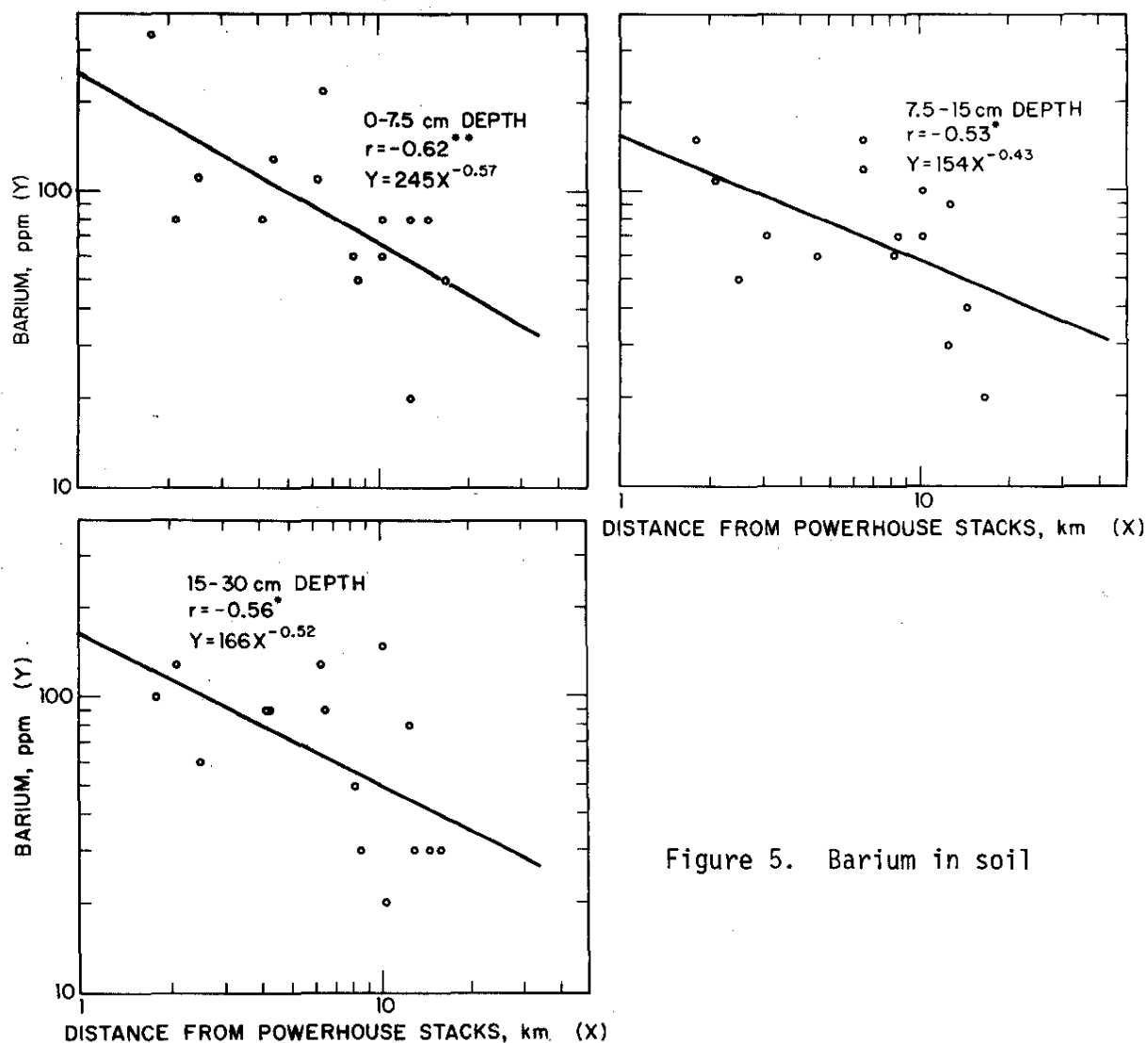
TRACE ELEMENTS IN SOIL

Of the 29 trace elements measurable in fly ash, all could be measured in most soil samples except Cd, which was <0.1 ppm in 43 of the 45 soil samples. At Locations 3 and 14, the 7.5-15 cm depth of soil contained 0.2 ppm of Cd. Only 7 of the trace elements showed a significant correlation between the logarithm of the concentration in soil and the logarithm of the distance from the powerhouse. These were Ba, Sr, and Mn in all depths of soil (Figures 5-7), Cu and Hg in the 0-7.5 cm depth (Figure 8), and Be and Se in the 7.5-15 cm depth (Figure 9). As shown in Table 3, Ba, Sr, and Mn are the trace elements released in the largest quantities and would be more likely to cause a measurable effect in soil. Also, the mobility of these elements in soil⁶⁻⁸ accounts for the measurable effects at all three depths.

Although Hg has been released in very small quantities (Table 3), it is considered very immobile in the soil⁹⁻¹⁰ and thus would be expected to accumulate in the surface soil as was observed. Cu is also not readily leachable.¹¹ The only apparent reason for the significant correlation between Be and Se in only the 7.5-15 cm depth of soil is that they may have been released in larger quantities at some time in the past when different coal was burned. To evaluate the importance of these measurable effects of the powerhouse stack releases upon the concentration of trace elements in soil, the concentrations at Location 7 (1.8 km from the stacks) were compared with those at Location 15 (16.5 km from the stacks); the concentrations at Location 15 are probably very near the natural levels. For this comparison, estimated concentrations calculated with the equations determined by linear regression provide a statistically better comparison than the measured results. The ratio of the estimated concentration at these two locations for each trace element having a significant relation with distance from the stacks is shown below:

| <i>Element</i> | <i>Soil depth, cm</i> | <i>Ratio of concentration at 1.8 km to concentration at 16.5 km</i> |
|----------------|---------------------------|---|
| Ba | 0-7.5 | 3.7 |
| Ba | 7.5-15 | 2.6 |
| Ba | 15-30 | 3.1 |
| Mn | 0-7.5 | 2.6 |
| Mn | 7.5-15 | 3.6 |
| Mn | 15-30 | 4.1 |
| Sr | 0-7.5 | 3.8 |
| Sr | 7.5-15 | 4.3 |
| Sr | 15-30 | 2.1 |
| Cu | 0-7.5 | 2.4 |
| Hg | 0-7.5 | 2.1 |
| Be | 7.5-15 | 2.4 |
| Se | 7.5-15 | 2.8 |

These ratios indicate that the powerhouse stack releases have increased the concentrations of these elements in soil by a factor of about 2 to 4 at the sampling location nearest to the stacks.



For Figures 5-23, the * indicates a significant correlation at the 0.05 level of significance and the ** indicates a significant correlation at the 0.01 level of significance.

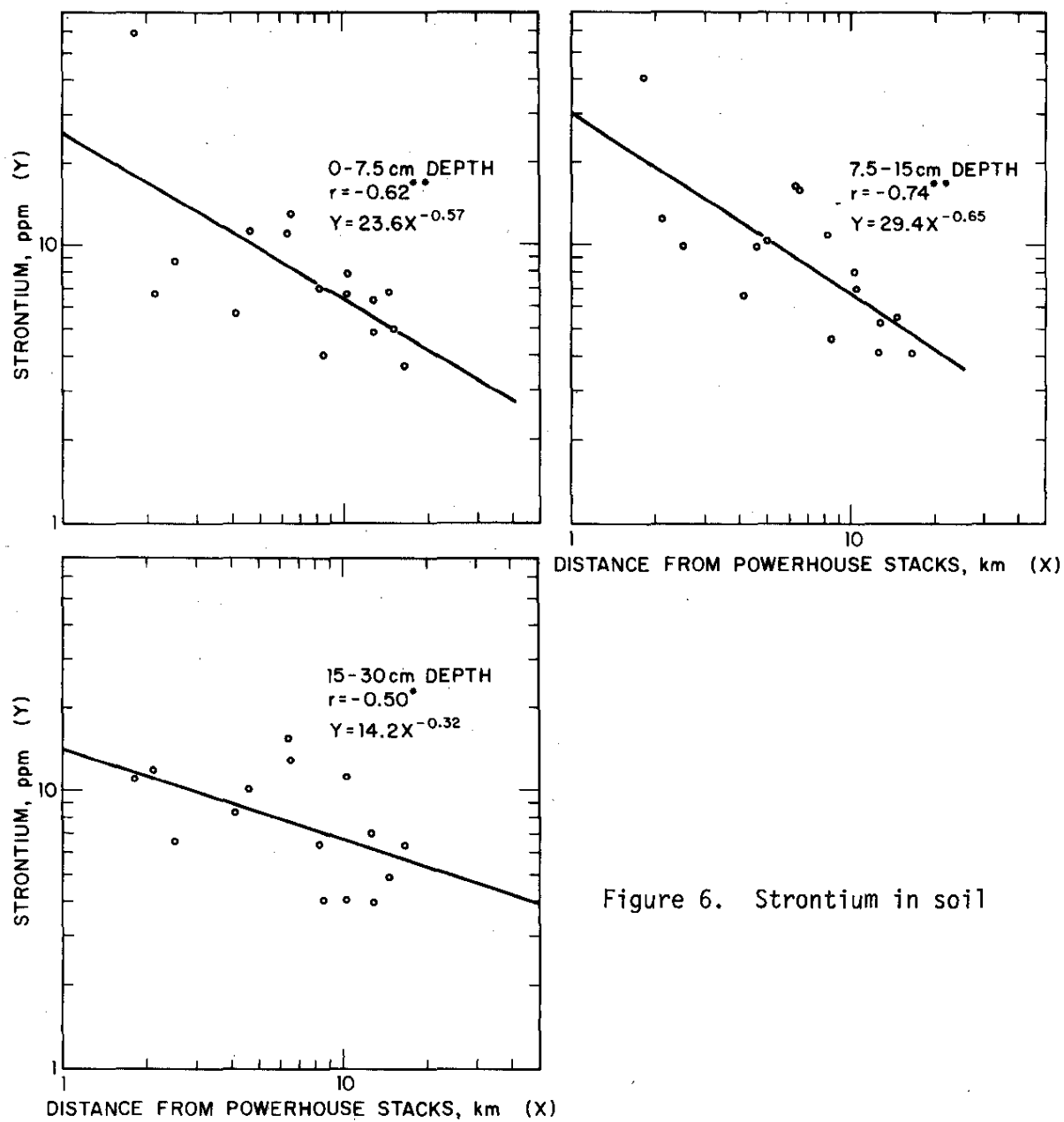


Figure 6. Strontium in soil

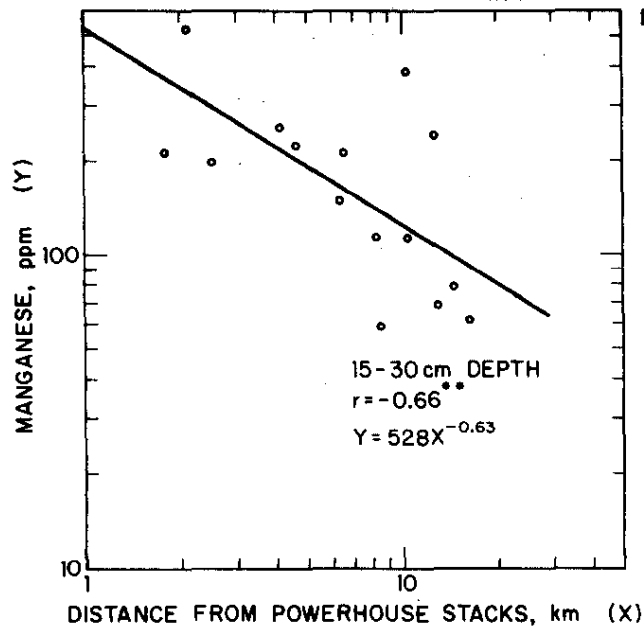
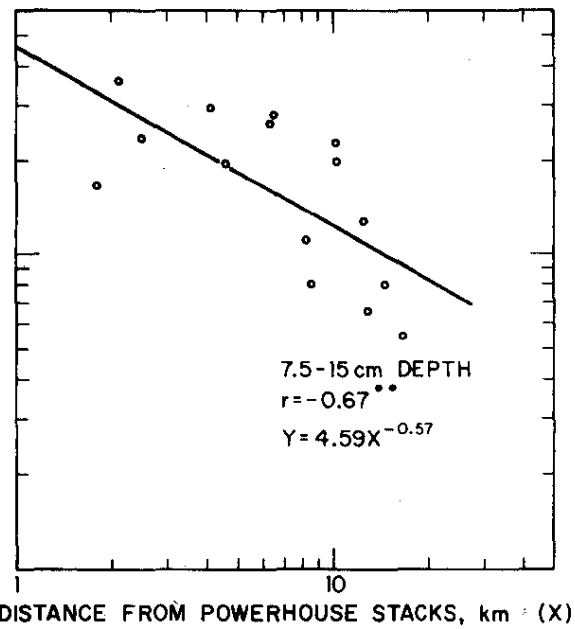
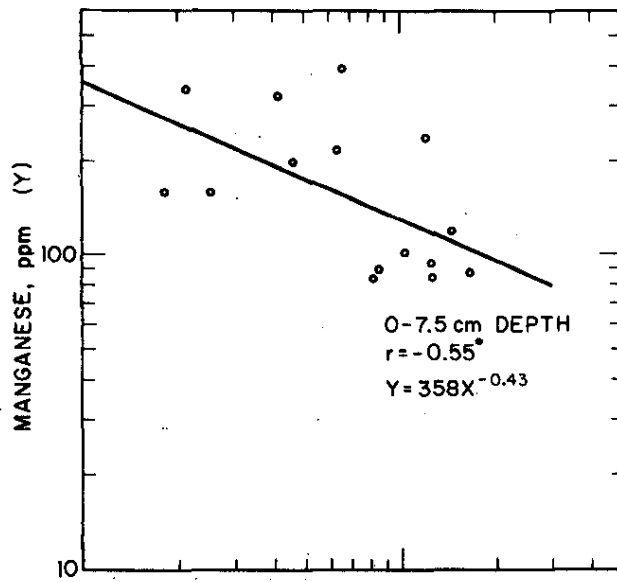


Figure 7. Manganese in soil

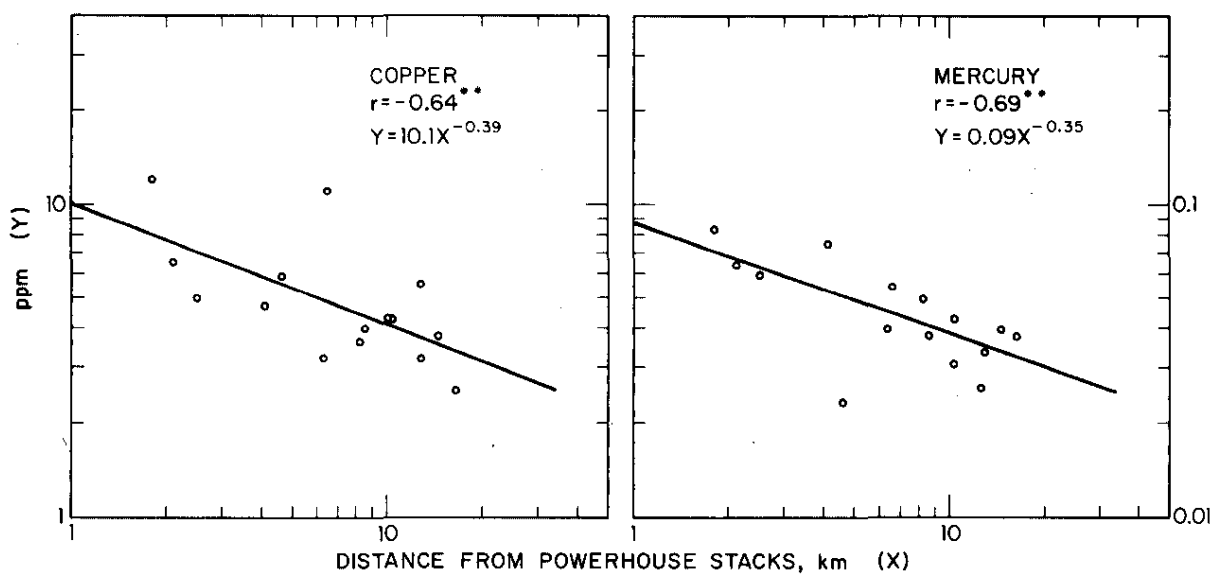


Figure 8. Copper and mercury in 0-7.5 cm depth of soil

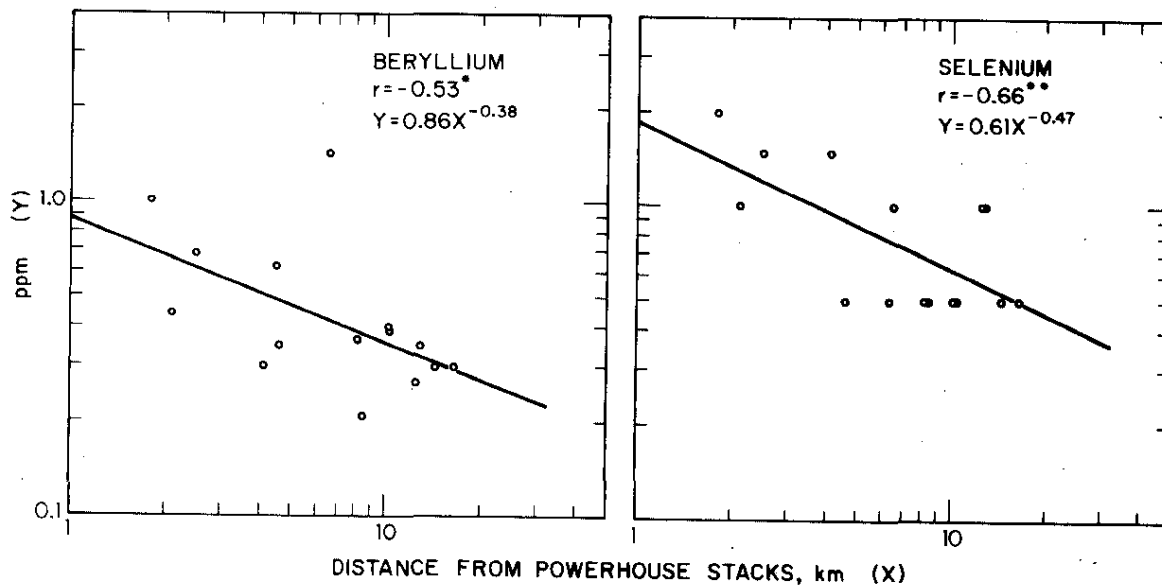


Figure 9. Beryllium and selenium in 7.5-15 cm depth of soil

As shown in Table 5, the concentrations of Ce, Co, Cr, Hg, Ni, La, Pb, Se, Sm, U, and Yb differed significantly in the different depths of soil. Of these elements, Co, Cr, Ni, Pb, and Hg concentrations were higher in the 0-7.5 cm depth than in the greater depths, and thus indicated deposition from the atmosphere. Eu, Se, Sm, U, and Yb concentrations were highest at the 15-30 cm depth and indicated either natural occurrence at this depth, precipitation as they were leached from above, or greater atmospheric deposition in the past than has occurred more recently.

Table 5. TRACE ELEMENTS IN THE DIFFERENT DEPTHS OF SOIL
(average ppm for all locations)

| Element | Soil depth | | | LSD ^a |
|---------|------------|-----------|----------|------------------|
| | 0-7.5 cm | 7.5-15 cm | 15-30 cm | |
| Ba | 103 | 79 | 73 | NSD ^b |
| Sr | 10.8 | 10.7 | 8.4 | NSD |
| Mn | 182 | 183 | 194 | NSD |
| Zn | 16.4 | 17.0 | 15.3 | NSD |
| V | 18.5 | 16.2 | 16.1 | NSD |
| Ce | 48 | 51 | 32 | 15 |
| Cr | 22 | 14 | 14 | 4 |
| As | 1.5 | 1.3 | 1.7 | NSD |
| Cu | 5.3 | 4.9 | 4.6 | NSD |
| Ni | 6.4 | 4.8 | 4.4 | 0.9 |
| Ga | 3.0 | 2.6 | 2.9 | NSD |
| La | 19.1 | 19.5 | 17.4 | 2.1 |
| Co | 5.5 | 2.6 | 2.4 | 1.4 |
| Pb | 12.7 | 8.1 | 8.6 | 3.1 |
| Br | 1.3 | 1.2 | 1.5 | NSD |
| Sc | 1.9 | 1.9 | 1.8 | NSD |
| Th | 9.5 | 8.9 | 7.7 | NSD |
| Sb | 0.4 | 0.3 | 1.4 | NSD |
| Mo | 1.3 | 1.1 | 1.2 | NSD |
| Be | 0.67 | 0.48 | 0.44 | NSD |
| Sm | 3.3 | 5.3 | 15.6 | 5.2 |
| Se | 0.9 | 0.9 | 1.3 | 0.4 |
| Cs | 1.3 | 1.2 | 1.3 | NSD |
| U | 2.2 | 3.2 | 14.8 | 8.1 |
| Eu | 0.3 | 0.5 | 7.6 | 3.6 |
| Yb | 1.5 | 1.3 | 3.6 | 1.3 |
| Tb | 0.5 | 0.6 | 0.5 | NSD |
| Hg | 0.048 | 0.017 | 0.032 | 0.011 |

^a LSD - least significant difference at the 0.05 level of significance.

^b NSD - no significant difference at the 0.05 level of significance.

TRACE ELEMENTS IN VEGETATION

Of the 29 trace elements measured in fly ash, 17 were measurable in the three species of vegetation: Ba, Be, Br, Cd, Co, Cr, Cu, Hg, Mn, Mo, Ni, Pb, Sc, Se, Sr, V, and Zn. Only 5 of these trace elements showed a significant correlation in one of the species of vegetation: Be, Co, Mo, and V in dog fennel (Figures 10-13) and Sr in panic grass

(Figure 14). Also, Br in dog fennel (Figure 15) and Hg in broom sedge (Figure 16) showed a significant positive correlation. This positive correlation indicated that these species are obtaining Br and Hg from some source other than the powerhouse stacks or that something released from the 400-D Area to the atmosphere is influencing the availability of Br and Hg in soil.

To evaluate the importance of these measurable effects of powerhouse stack releases upon the concentration of trace elements in vegetation, the ratio of the estimated concentration calculated with the linear regression equation at Location 7 (1.8 km from the stacks) and Location 15 (16.5 km from the stacks) was determined. These are shown below:

| <i>Element</i> | <i>Species</i> | <i>Ratio of concentration at 1.8 km to concentration at 16.5 km</i> |
|----------------|----------------|---|
| Sr | Panic grass | 2.0 |
| V | Dog fennel | 2.0 |
| Co | Dog fennel | 2.2 |
| Mo | Dog fennel | 3.2 |
| Be | Dog fennel | 1.4 |

These ratios indicate that the powerhouse stack releases have increased the concentration of these trace elements in vegetation by a factor of about 1.5 to 3.

The concentrations of Ba, Be, Cd, Co, Cr, Cu, Hg, Mn, Ni, Pb, Sc, Sr, V, and Zn differed significantly in the different species of vegetation as shown in Table 6. Ba, Be, Cu, Ni, and Sr concentrations were higher in dog fennel than in the other species, but Hg, Mn, Pb, and Zn concentrations were higher in panic grass than in the other species.

Table 6. TRACE ELEMENTS IN THE DIFFERENT SPECIES OF VEGETATION (ppm)

| <i>Element</i> | <i>Dog fennel</i> | <i>Panic grass</i> | <i>Broom sedge</i> | <i>LSD^a</i> |
|----------------|-------------------|--------------------|--------------------|------------------------|
| Ba | 89 | 50 | 36 | 14 |
| Sr | 40 | 9 | 10 | 5 |
| Mn | 194 | 310 | 214 | 58 |
| Zn | 46 | 105 | 33 | 21 |
| V | 1.1 | 1.3 | 0.5 | 0.3 |
| Cr | 21 | 14 | 20 | 1.5 |
| Cu | 8.6 | 5.6 | 4.7 | 1.2 |
| Ni | 4.9 | 2.9 | 2.6 | 1.0 |
| Co | 1.1 | 1.1 | 0.6 | 0.2 |
| Pb | 5 | 23 | 3 | 9 |
| Br | 34 | 30 | 27 | NSD ^b |
| Sc | 0.19 | 0.17 | 0.07 | 0.08 |
| Mo | 0.5 | 0.6 | 0.4 | NSD |
| Be | 0.3 | 0.1 | 0.1 | 0.03 |
| Se | 1.7 | 1.3 | 1.2 | NSD |
| Hg | 0.048 | 0.093 | 0.049 | 0.017 |
| Cd | 0.10 | 0.14 | 0.03 | 0.05 |

^a LSD - least significant difference at the 0.05 level of significance.

^b NSD - no significant difference at the 0.05 level of significance.

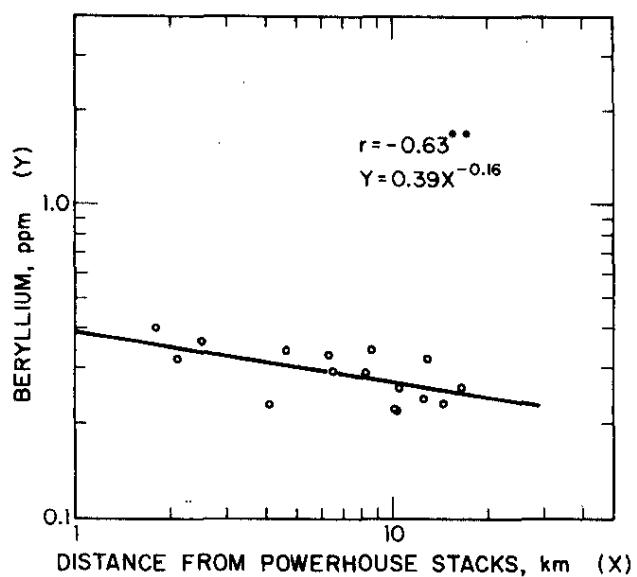


Figure 10. Beryllium in dog fennel

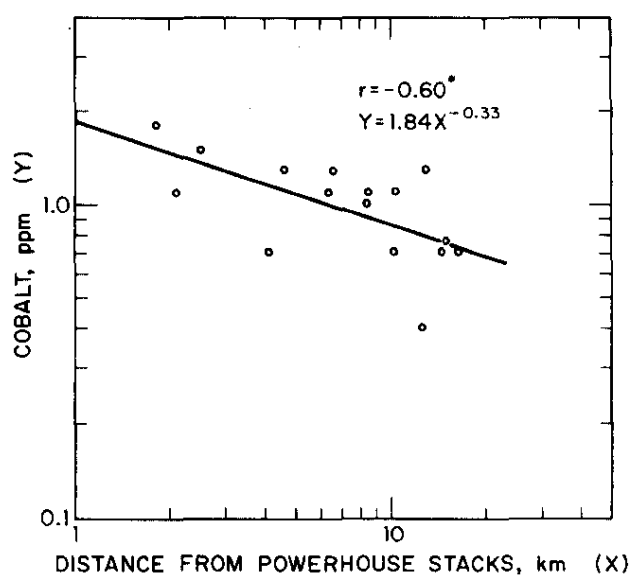


Figure 11. Cobalt in dog fennel

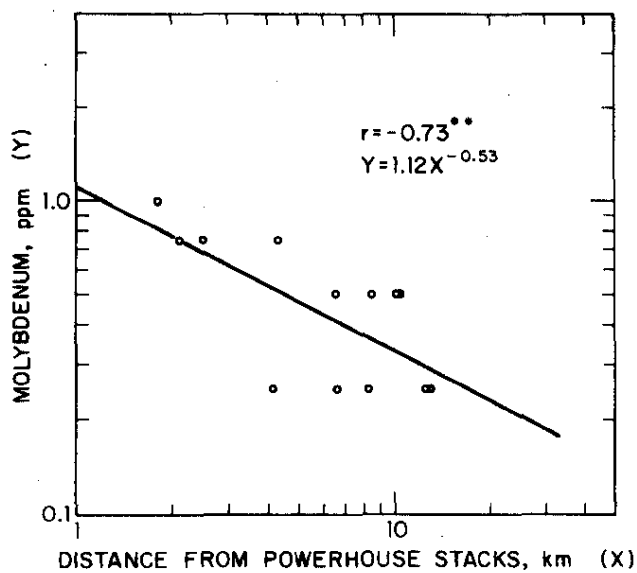


Figure 12. Molybdenum in dog fennel

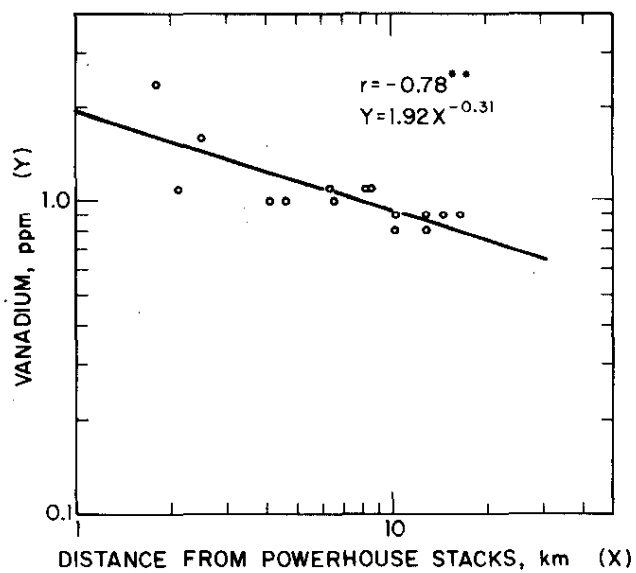


Figure 13. Vanadium in dog fennel

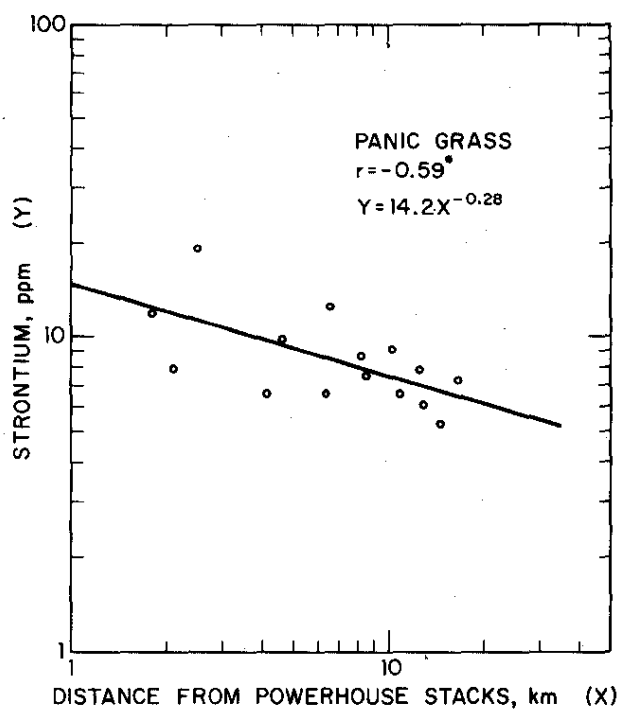


Figure 14. Strontium in panic grass

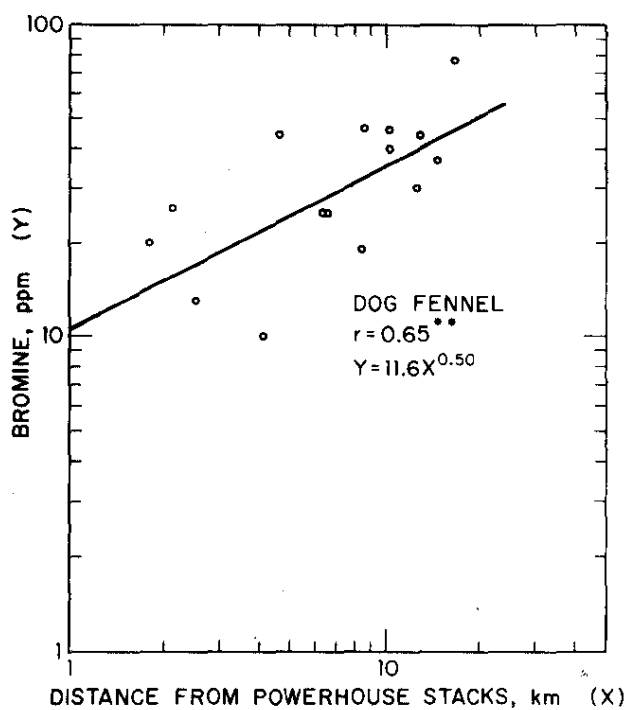


Figure 15. Bromine in dog fennel

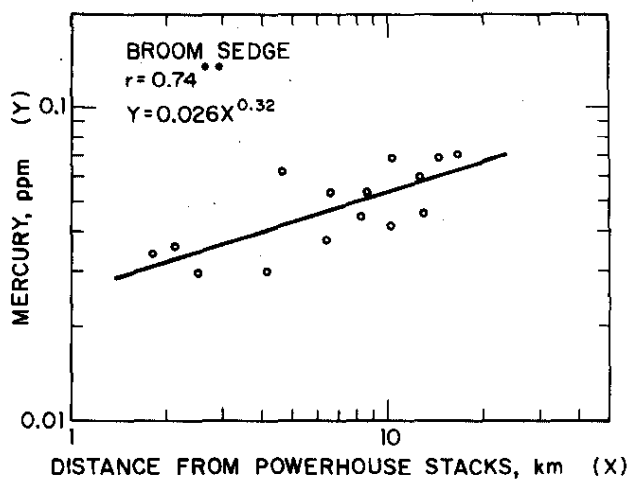


Figure 16. Mercury in broom sedge

TRACE ELEMENTS IN GROUND WATER

Of the 29 trace elements measured in fly ash, only 7 were measured in most of the 14 ground-water samples. These include Cu, Co, Cr, Hg, Mn, Sr, and Zn. Co and Mn showed a significant negative correlation between the logarithm of the concentration and the logarithm of the distance from the powerhouse stack (Figures 17 and 18). The negative correlation indicated an influence on concentration by the stack releases. The significant positive correlation for Cu (Figure 19) indicated a source other than the powerhouse stacks or that something released from the 400-D Area to the atmosphere is reducing the leachability of Cu in soil.

To evaluate the importance of these measurable effects of powerhouse stack releases upon the concentration of trace elements in ground water, the ratio of the estimated concentration calculated with the linear regression equation at Location 7 (1.8 km from the stacks) and Location 15 (16.5 km from the stacks) was determined:

| <i>Element</i> | <i>Ratio of concentration at 1.8 km to concentration at 16.5 km</i> |
|----------------|---|
| Co | 2.1 |
| Mn | 13.4 |

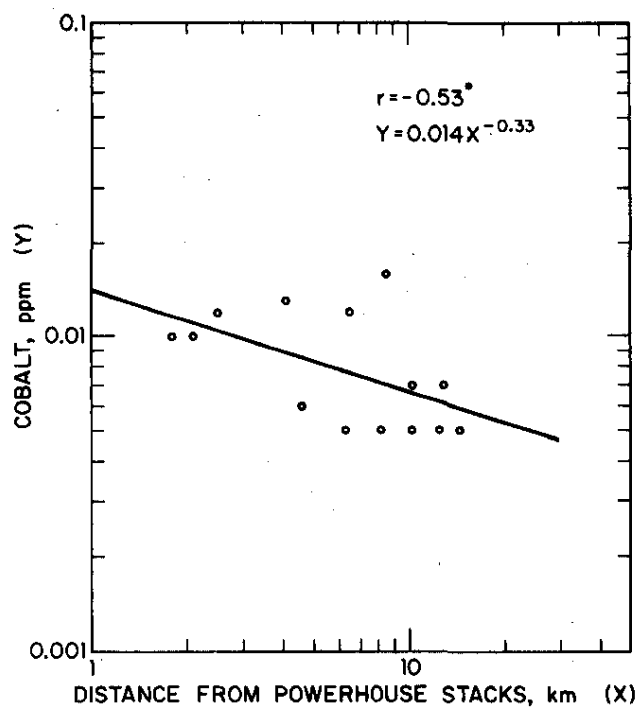


Figure 17. Cobalt in ground water

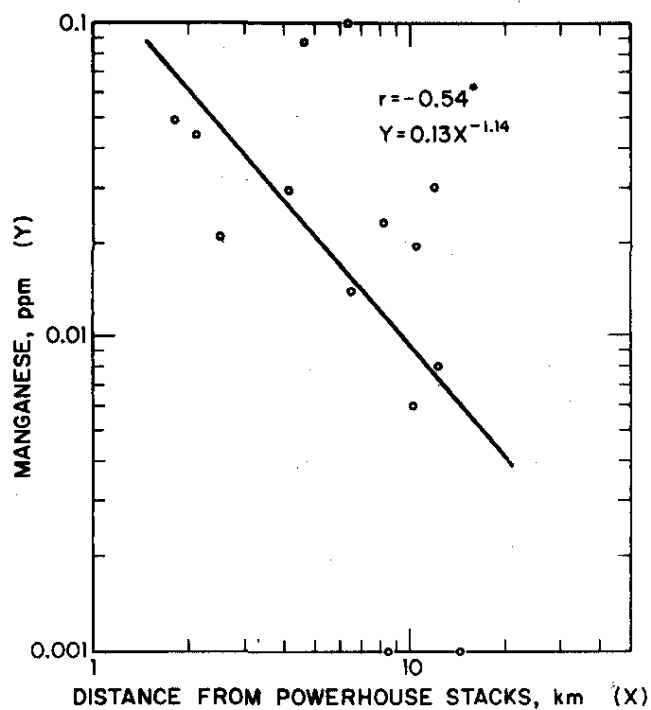


Figure 18. Manganese in ground water

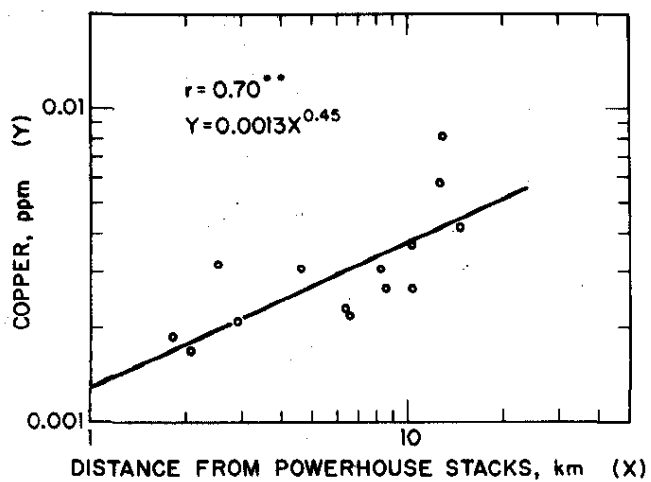


Figure 19. Copper in ground water

AVAILABILITY TO VEGETATION OF TRACE ELEMENTS IN FLY ASH

If the trace elements in fly ash deposited on soil are to influence the terrestrial ecosystem, the elements must enter the food chain through absorption by vegetation. Very little is known concerning the availability of the trace elements in fly ash to plants.¹² Some research has shown that additions of fly ash to soil increases the available Mo¹³ and B.¹⁴ However, some laboratory studies indicate that a very large percentage of the trace elements in fly ash may not be readily available to plants.¹⁵

To determine the comparative availability of trace elements in fly ash released from the powerhouse stacks and the trace elements naturally present in Savannah River Plant soil, the ratio of the concentration of each trace element in vegetation to the concentration in soil at each sampling location was calculated. The average concentration for the three species of vegetation and three depths of soil was used. If a trace element is more available in fly ash than in soil and if the fly ash deposition has been sufficient to influence the concentrations, then the ratio should decrease as distance from the powerhouse stacks increases. On the other hand, if the trace element is less available in fly ash than in the soil, then the ratio should increase as distance from the powerhouse stacks increases. Curvilinear regression and correlation analyses were used to determine whether a significant relation exists between the logarithm of the ratio of trace elements in vegetation to trace elements in soil and the logarithm of the distance from the powerhouse stacks. Only four significant correlations were determined. These were for Ba, Sr, Mn, and Hg. All of these correlations were positive, as shown in Figures 20-23. Thus, the Ba, Sr, Mn, and Hg present in the deposited fly ash are less available than in Savannah River Plant soil.

Because Ba, Sr, and Mn are the trace elements of highest concentrations in fly ash, similar results may have been attained with other elements if these elements had been deposited in larger quantities.

Fly ash consists of small glass-like spheres.¹⁶ Some elements condense on the surface of the spheres and others are incorporated. Because glass is very resistant to leaching,¹⁷ the elements incorporated in the glass should probably remain unavailable to plants for very long periods of time.

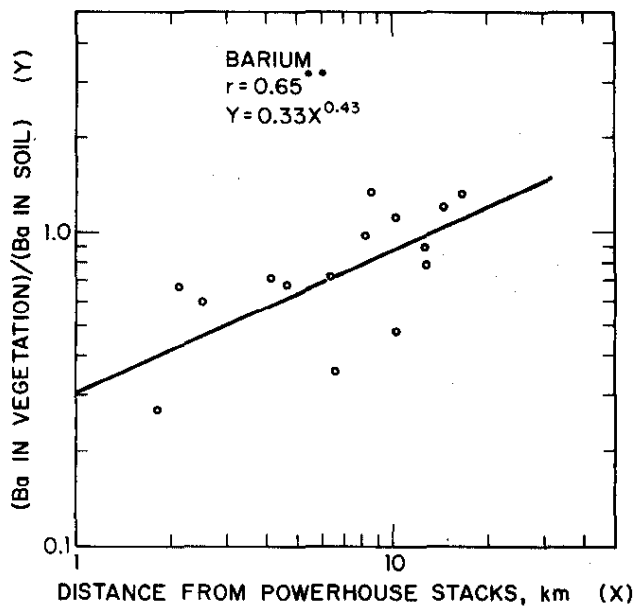


Figure 20. Ratio of barium in vegetation to barium in soil

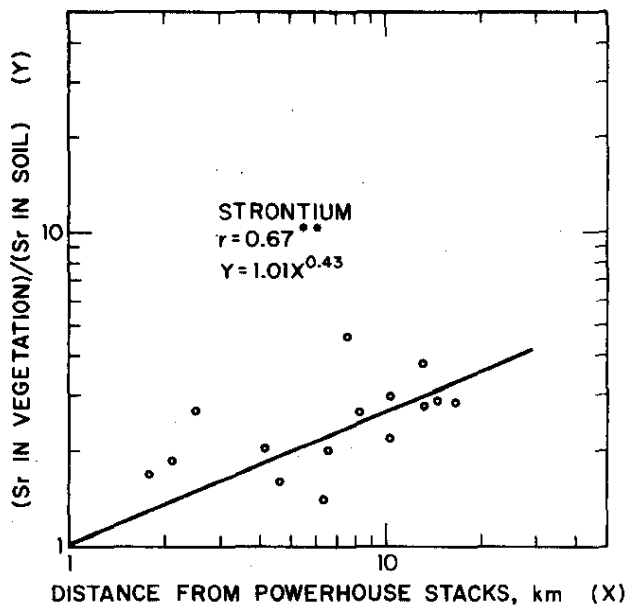


Figure 21. Ratio of strontium in vegetation to strontium in soil

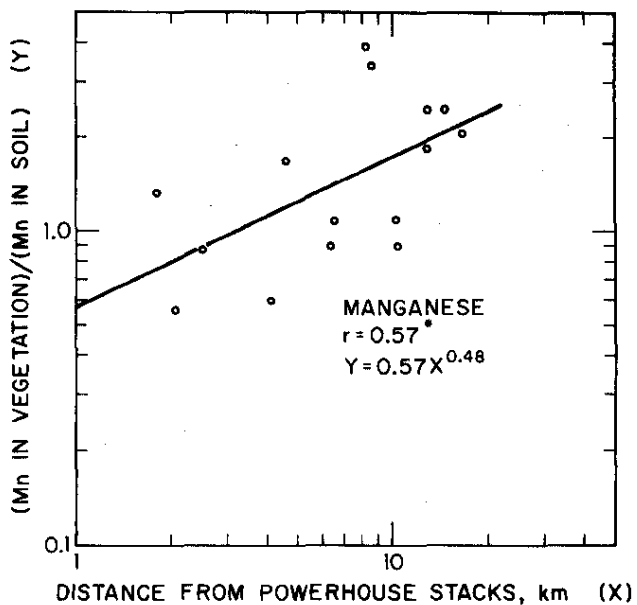


Figure 22. Ratio of manganese in vegetation to manganese in soil

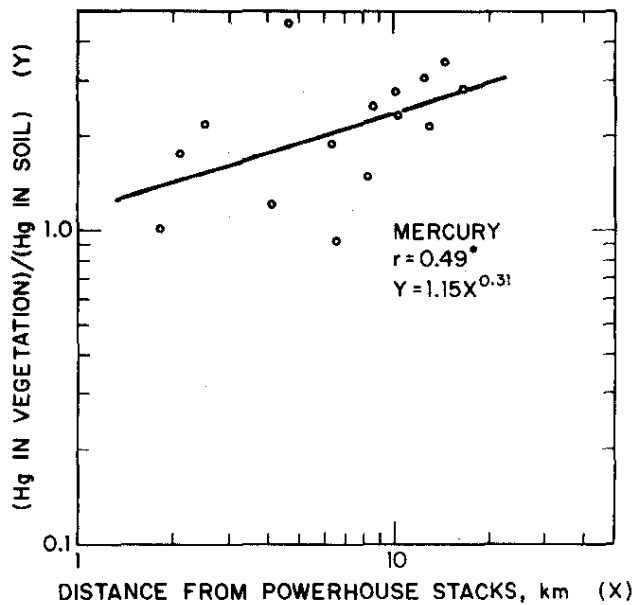


Figure 23. Ratio of mercury in vegetation to mercury in soil

DISCUSSION

The variation in soils at the 15 environmental sampling locations certainly influenced the results of this study. All were sandy and had a very low pH as shown in Table 7.

The consequences of the measurable effects of powerhouse stack releases upon the concentrations of trace elements in soil depend upon several factors. These factors include the magnitude of the concentrations measured as compared with the concentrations found in most soils, the availability to plants of the trace elements in fly ash as compared with the same trace element in soil, and the concentrations produced in vegetation as compared to toxic concentrations. Each factor is considered in the following sections for each trace element for which the powerhouse stack releases produced a measurable effect in the environmental samples.

Table 7. pH OF SOILS

| Location | Soil depth | | |
|----------|------------|-----------|----------|
| | 0-7.5 cm | 7.5-15 cm | 15-30 cm |
| 1 | 4.5 | 4.5 | 4.8 |
| 2 | 5.0 | 5.1 | 5.3 |
| 3 | 4.6 | 4.9 | 5.0 |
| 4 | 4.6 | 4.9 | 4.5 |
| 5 | 4.7 | 5.1 | 4.9 |
| 6 | 5.0 | 4.9 | 5.3 |
| 7 | 5.0 | 4.7 | 5.2 |
| 8 | 4.8 | 5.2 | 5.3 |
| 9 | 5.1 | 5.5 | 5.6 |
| 10 | 5.0 | 5.3 | 5.2 |
| 11 | 4.4 | 4.5 | 4.8 |
| 12 | 4.6 | 4.7 | 4.8 |
| 13 | 4.7 | 5.2 | 5.3 |
| 14 | 4.4 | 4.9 | 4.9 |
| 15 | 4.7 | 4.8 | 4.9 |

BARIUM

Ba is not essential for either plants or animals. Evidence regarding the toxicity of Ba to plants is conflicting, but generally the total quantity in soils is of little significance provided it does not precipitate SO_4^{2-} and cause a sulfur deficiency.⁶ The Ba content of most soils ranges from 100 to about 3000 ppm. The highest concentration measured in this study was 340 ppm in the 0-7.5 cm depth of soil at Location 7. This concentration is not unusually high, and the influence of the stack releases was not measurable in vegetation. The results in Figure 20 show that Ba in fly ash is less available to vegetation than is the Ba naturally occurring in soil. Any possible influence upon sulfur availability should be compensated for by stack releases of SO_2 .

STRONTIUM

Sr does not appear to be essential for plant growth⁸ although it is readily absorbed by plants. Because of its chemical similarity to Ca, the two are absorbed by plants in the same ratio as they are present in the soil solution. No evidence of naturally occurring toxicity has been reported. Most soils contain from 50 to 1000 ppm Sr with an average abundance of about 150 ppm. The highest concentration measured in this study, 590 ppm in the 0-7.5 cm depth of soil at Location 7, is higher than the average in most soils. Although Sr is readily adsorbed by plants, results in Figure 21 indicate that Sr in fly ash is less available to plants than the Sr in Savannah River Plant soils. The effects of the powerhouse stack releases upon the Sr concentration in vegetation could be measured only in panic grass. The maximum measured concentration was 19 ppm at Location 6 (2.5 km from the powerhouse stacks). This concentration is very similar to the normal concentrations reported in grasses.⁸

MANGANESE

Mn is an essential element for plant growth and can be toxic in high concentrations.⁷ The Mn content of most soils ranges from 200 to 3000 ppm with an average of about 600 ppm. The highest concentration in soil measured in this study was 530 ppm in the 15-30 cm depth of soil at Location 8 (2.1 km from powerhouse stacks). This concentration is near the average for most soils. Although Mn is readily absorbed by plants, the measured effects of powerhouse stack releases in soil are not reflected in the concentrations measured in vegetation. Also, the results in Figure 22 indicate that Mn in fly ash is less available

to plants than is the Mn naturally present in Savannah River Plant soils. As shown in Figure 18, the powerhouse stack releases of Mn have measurably affected the concentration of Mn in ground water. Two water samples in this study contain more than the water quality criteria for surface waters¹⁸ of 0.05 ppm. These were 0.088 ppm at Location 5 (4.6 km from the powerhouse stacks) and 0.101 ppm at Location 11 (8.2 km from the powerhouse stacks). At Location 7 (1.8 km from the powerhouse stacks), the concentration was 0.049 ppm. Thus, naturally high concentrations have been increased by the powerhouse stack releases. The principal reason for limiting the concentration of Mn in water is to reduce esthetic and economic problems,¹⁹ because a high concentration in laundry water can cause stains and if used to clean milk containers can cause undesirable flavors.²⁰

COPPER

Cu is essential for plant growth and can also be toxic in high concentrations.^{11,21} The concentration of Cu in most soils ranges between 2 and 100 ppm. In sandy soils such as the Atlantic coastal plains, the range is 1 to 30 ppm. The highest concentration measured in this study was 14 ppm in the 7.5-15 cm depth of soil at Location 4 (6.5 km from the powerhouse stacks), and because of its distance from the stack, appears to be largely naturally occurring. At Location 7 (1.8 km from the powerhouse stacks), the maximum concentration was 12 ppm in the 0-7.5 cm depth of soil. Both of these concentrations are similar to those usually found in soils. The measurable effects of the stack releases upon the concentrations of Cu in soils were not reflected in the concentrations in vegetation. The statistical correlation of the logarithm of the concentrations in ground water versus the logarithm of the distance from the powerhouse stacks showed a significant positive correlation (Figure 19), and is probably due to the insolubility of Cu_2S and CuS . The 400-D Area in which the powerhouse is located produces heavy water, D_2O . This process releases large quantities of H_2S to the atmosphere.²² Thus, the leachability of Cu in soil could be reduced near the area, and thereby less Cu would be leached into the ground water near the stacks than at greater distances.

BERYLLIUM

The literature gives insufficient data to evaluate the effects of Be released by the powerhouse stacks. Be is very toxic to man.²³ The upper lithosphere contains an average of 2 ppm. The concentrations in all samples of soil in this study were less than 2 ppm except the 0-7.5 cm depth at Location 9, which contained 3.7 ppm. Most of the soil contained considerably less than 1 ppm. The measurable effect of powerhouse stacks releases in soil did not extend to vegetation or ground water.

SELENIUM

The need for Se by growing plants is uncertain, but if it is essential, only very small concentrations are required.²⁴ Very small quantities are essential for animals.^{21,23} In both plants and animals, Se can be highly toxic when present in sufficient quantities. Most soils contain 0.1 to 2 ppm. The highest concentration measured in soil in this study was 2 ppm in both the 0-7.5 cm and 7.5-15 cm depths of soil at Location 7 (1.8 km from the powerhouse stacks). This concentration is at the upper limit found in most soils. The measurable effect of powerhouse stack releases upon the Se content of soil was not reflected in vegetation or ground water. The highest concentration measured in vegetation was 4 ppm in dog fennel at Location 8 (2.1 km from the powerhouse stacks). This concentration is very low compared to the 100 ppm or greater, which have been reported to be toxic to plants²⁴ but could be toxic to animals since 2 to 4 ppm in their diet is considered toxic.²¹ Thus what appears to be naturally high Se concentrations in the Savannah River Plant environs have been accentuated by the powerhouse stack releases.

COBALT

Evidence has not been offered to show that Co is essential to higher plants, but Co is essential to animals.²⁵ Also, it can be toxic to plants. The Co content of most soils is in the range of 1 to 40 ppm. The maximum concentration measured in soil in this study was 13 ppm in the 0-7.5 cm depth of soil at Location 7 (1.8 km from the powerhouse stacks). This is within the range of most soils. Although the highest concentration was in soil nearest to the powerhouse stacks, concentrations of 8.5 to 10 ppm at locations much more distant from the stack prevented a measurable effect of stack releases. The effect of powerhouse stack releases upon the Co content of dog fennel was measurable, and the highest concentration, 1.8 ppm, was at Location 7. This concentration is within the range reported for vegetation not showing toxic symptoms. For corn, 4 to 8 ppm in leaves is required to produce toxic symptoms.²⁵

The effect of Co released by the powerhouse stacks upon the concentration in ground water was also measurable, as shown in Figure 17. No drinking water criteria are available for comparison, but all measured concentrations are considerably less than the criteria for irrigation water which is 0.2 ppm.¹⁸ The maximum concentration measured in this study was 0.016 ppm at Location 3 (8.5 km from the powerhouse stacks). At Location 7 (1.8 km from the powerhouse stacks), the concentration was 0.010 ppm.

MOLYBDENUM

Mo is an essential element for plant growth^{21,26} and can be toxic to plants, but only in very high concentrations. It is also essential for animals, but can be toxic especially to ruminants. Soils generally contain 0.6 to 3.5 ppm. The concentration in all soil samples analyzed in this study was between 1 and 2 ppm and there was no measurable effect of stack releases upon concentrations in soil, but there was a measurable effect of powerhouse stack releases upon the concentration in dog fennel (Figure 12). The highest concentration in dog fennel was 1.0 ppm at Location 7 (1.8 km from the powerhouse stacks). The concentration required to be toxic to plants is not well defined, but hundreds or thousands of ppm appear to be required. The concentration toxic to animals is even more poorly defined.²¹

VANADIUM

There is no conclusive evidence that V is essential for the growth of higher plants, but there is evidence that it is essential to bacteria and algae. Also, the level below which there is no toxic effect is difficult to predict.²⁷ The concentration in soils is usually between 20 and 500 ppm. The highest concentration measured in soil in this study was 62 ppm in the 15-30 cm depth of soil at Location 4 (6.5 km from the powerhouse stacks). The effect of the stack releases upon concentrations in soil was not measurable, but there was a measurable effect upon the concentration in dog fennel (Figure 13). The highest concentration measured in dog fennel was 2.4 ppm at Location 7 (1.8 km from the stack). There are no criteria in the literature for determining toxicity levels.

BROMINE

Br has not been shown to be essential to plant growth, nor has it been regarded as particularly toxic.²⁸ Soils usually contain 10 to 40 ppm. In comparison, the soils in this study contained only 0.5 to 2.1 ppm and are thus very low in Br. The effect of Br releases from the powerhouse stacks upon the Br content of soil was not measurable. With vegetation, the statistical correlation of the logarithm of the Br concentration dog fennel versus the logarithm of the distance from the powerhouse stacks was significant but positive as shown in Figure 15. Thus the concentration of Br in dog fennel increased as the distance from the stack increased. This increase was found in both directions from the stacks thus eliminating another fixed airborne source.

The most likely explanation of this increase with distance is that the fly ash contains enough elements such as Hg and Ag which form highly insoluble bromides to reduce the already small quantity of Br available to plants. This reduction of availability would diminish as the distance from the stacks increases and because the Br content of a particular species of vegetation is related to the available Br in soil,²⁸ the amount of Br in the vegetation would increase as distance from the stacks increases.

MERCURY

Hg is not essential to plant growth but can be toxic to both plants and animals. The concentration in soils ranges from 0.01 to 10,000 ppm⁹ and, the average concentration has been reported to be 0.1 ppm¹⁰ and also 0.07 ppm.²⁹ The highest concentration measured in this study was 0.083 ppm in the 0 to 7.5 cm depth of soil at Location 7 (1.8 km from the powerhouse stacks), and most values were considerably less. Hg is generally not very soluble in soil, and therefore not readily leached or readily available to plants.¹⁰ The measurable effect of stack releases upon the concentrations of Hg in soil was not reflected in the concentrations in vegetation. The statistical correlation between the logarithm of the concentration of Hg in vegetation/concentration of Hg in soil and the logarithm of the distance from the powerhouse stacks was positive and indicated that the Hg deposited in fly ash is less available to vegetation than is Hg in the soil (Figure 23). The decreased availability of Hg near the powerhouse stacks is probably also influenced by the releases of H₂S to the atmosphere from the 400-D Area heavy water (D₂O) plant.²² The H₂S could convert some of the Hg to the very insoluble sulfides. The effect would be largest near the area and diminish with distance.

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APPENDIX A

FLY ASH DEPOSITIONS ATTRIBUTABLE TO 484-D POWERHOUSE

To determine the influence of the 484-D powerhouse stack releases upon trace element concentrations in the environment, soil, vegetation, and ground water samples must be studied. However, the samples are influenced by long-term depositions from other coal-burning facilities in the area. Therefore, the contribution from just the 484-D powerhouse must be estimated at each sampling location. The Savannah River Laboratory two-year meteorological data base¹ was processed to provide a basis for this estimate.

Deposition calculations are based on a Gaussian plume dispersion model as modified by Chamberlain to account for particle fall velocities. Chamberlain's modification produces a downward tilt in the plume according to the fall velocity of each grouping of particles to be considered. An aggregate of particles of different size must therefore be divided into groupings each with a particular fall velocity to properly treat the aggregate. The Gaussian plume dispersion model for calculating downwind ground level concentrations χ from a continuous release is

$$\chi = \frac{Q}{\pi \sigma_y \sigma_z \bar{u}} \exp \left[-\frac{1}{2} \left(\frac{y^2}{\sigma_y^2} + \frac{H^2}{\sigma_z^2} \right) \right] \quad (1)$$

where Q = source, lb/sec

σ_y, σ_z = horizontal and vertical Gaussian dispersion parameters, m

\bar{u} = wind speed, m/sec

y = crosswind distance, m

H = height of plume centerline, m

To account for particle settling, the modified height term in the exponent is

$$H = h - \frac{xV_g}{\bar{u}}$$

where x = downwind distance, m

V_g = particle terminal fall velocity, m/sec

h = calculated plume height, m

Deposition rate is determined from the product $V_g \chi$. The equation for plume centerline deposition rate w for distance x becomes

$$w = \frac{QV_g}{\pi \bar{u} \sigma_y \sigma_z} \exp \left[-\frac{1}{2} \left(\frac{\bar{u}h - xV_g}{\bar{u} \sigma_z} \right)^2 \right] \quad (2)$$

The calculated plume height h is a function of release height, effluent velocity, buoyancy effects, and meteorological conditions. Many procedures to calculate h are given in the literature. The method chosen for this work is discussed in Reference 2 and briefly discussed below.

The stack effluent is divided into two distinct stages: a transitional stage and a final stage when the plume has reached a limiting height. The limiting height is also a function of meteorological stability conditions so that a minimum of three equations must be defined to calculate h when stable and unstable atmospheric conditions are considered. These equations are:

Transitional: $\Delta h_0 = (2.0F^{1/3} x^{2/3}) / \bar{u}$

Stable limiting: $\Delta h_1 = 2.6(F/\bar{u}s)^{1/3}$

Unstable limiting: $\Delta h_2 = \frac{400F}{\bar{u}^3} + 3r \frac{W}{\bar{u}}$

where F = buoyancy term approximated by $F = 3.8 \times 10^{-5} Q_H$

Q_H = effluent heat emission rate, cal/sec

W = stack gas effluent velocity, m/sec

r = inside stack radius, m

s = stability parameter, assumed constant (0.445) for these calculations

The calculated plume height h is now defined as

$$h = S + \text{minimum} (\Delta h_0, \Delta h_i) * \begin{cases} i = 1 \text{ for stable atmosphere} \\ i = 2 \text{ for unstable atmosphere} \end{cases}$$

S = stack height, m

* Minimum $(\Delta h_0, \Delta h_i)$ implies the smallest value of the two arguments is to be used in evaluating the expression.

Sector-averaged depositions are calculated by integrating Equation 1 with respect to y , multiplying by the settling velocity, and dividing by the sector width as a function of distance:

$$\begin{aligned}\bar{w}_\theta &= \int_y \frac{XV_g}{2\pi x/N} \\ &= \frac{NQV_g}{2\pi x \sigma_z \bar{u}} \sqrt{\frac{2}{\pi}} \exp \left[-\frac{1}{2} \left(\frac{\bar{u}h - xV_g}{\bar{u}\sigma_z} \right)^2 \right]\end{aligned}\quad (3)$$

where N = number of sectors of equal angular width within 360°

Calculations must account for the different particle settling velocities as a function of particle size and the frequency of occurrence of wind velocities in the sector being considered. To facilitate these calculations, wind velocities were assigned to seven speed groups about an average wind speed, and four particle size groups were considered. The equation for total deposition rate then becomes

$$\bar{w}_\theta = \sum_{j=1}^m \sum_{g=1}^n \frac{\alpha_j \beta_g NQV_g}{2\pi x \sigma_z \bar{u}_j} \sqrt{\frac{2}{\pi}} \exp \left[-\frac{1}{2} \left(\frac{\bar{u}_j h - xV_g}{\bar{u}_j \sigma_z} \right)^2 \right]\quad (4)$$

where α_j = sector frequency of occurrence for \bar{u}_j
 β_g = fraction of total particle weight represented by group g
 m = number of wind speed groups
 n = number of particle size groups

The total stack fly ash emission rate, particle sizes, and the weight fraction of each particle size group were determined from emission tests as given in Reference 2. Table 1 lists the results of these tests along with other data used in these calculations. The listed settling velocities were obtained from Reference 3 and are based on experimentally determined terminal velocities for spherical particles as a function of particle density. Calculations were made for an assumed particle density of 3 g/cm^3 . Table 2 lists the wind speed groupings used in these calculations and the frequency of occurrence of these wind speeds for a typical sector. Frequencies are given for both stable and unstable atmospheric conditions since each condition must be calculated separately. Dispersion parameters were calculated according to procedures developed from dispersion tests at Brookhaven National Laboratory.⁴

Table 1. 484-D PARAMETERS FOR DEPOSITION CALCULATIONS

| | |
|----------------------------|---------------------------|
| Total fly ash emission | 122,000 lb/day |
| Stack height | 38.1 m |
| Inside stack diameter | 3.05 m |
| Stack effluent velocity | 7.62 m/sec |
| Stack effluent temperature | 450°K |
| Heat emission | 1.4×10^6 cal/sec |

FLY ASH PARTICLE DISTRIBUTION

| Group | Particle size, μ | Weight fraction of total emission | Settling velocity, m/sec |
|-------|----------------------|-----------------------------------|--------------------------|
| 1 | 5 | 0.555 | 0.0015 |
| 2 | 15 | 0.16 | 0.015 |
| 3 | 30 | 0.095 | 0.061 |
| 4 | 60 | 0.19 | 0.213 |

Table 2. WIND SPEED GROUPING FOR DISPERSION CALCULATIONS

| Wind speed, m/sec | Stable frequency of occurrence | Unstable frequency of occurrence |
|-------------------|--------------------------------|----------------------------------|
| 2 | 0.00381 | 0.00916 |
| 4 | 0.00880 | 0.0219 |
| 6 | 0.0150 | 0.0246 |
| 8 | 0.0131 | 0.0116 |
| 10 | 0.0018 | 0.00224 |
| 12 | 0.000058 | 0.00041 |
| 16 | 0.000012 | 0.000105 |

Estimates of fly ash deposition as a function of distance were made with 45° sector widths. Figure 1 shows the results of a typical sector. Because there was no strong variation between sectors, this figure was assumed to apply for all eight sectors at each Savannah River Plant emission point.

The total fly ash deposition at a sampling location is the sum of contributions from all sources:

$$\omega_i = \sum_{n=1}^N \psi_{n,i} S_n \quad (5)$$

where ω_i = surface deposition, $g \cdot m^{-2}$, at sample point i

ψ_n = grams deposited at sample point i per gram emitted from source point n

S_n = total emissions from source point n , grams

All source points that make a significant contribution to deposition at the sampling location are within the Savannah River Plant boundary and are indicated in Figure 2. Table 3 is a matrix indicating the distance from each source point to each sampling location. The ψ_n of Equation 5 are then obtained for each source point according to the distances given in Table 3.

Because the objective in this initial effort is to estimate the percentage contribution from 484-D to the total deposition at the various sampling locations, the S_n may be expressed in relative terms such as annual average emission or fraction of total emissions from all sources combined. Fractional source terms are given in Table 4 based on annual average emission during 1974. Fractional contributions F_i are then determined for 484-D as

$$F_i = \psi_{D,i} S_D / \omega_i \quad (6)$$

where the subscript D denotes 484-D parameters. These values are plotted as the desired percentages in Figure 3 and indicate a range of about 99% for the sampling locations nearest the 400 Area to about 71% at Location 15.

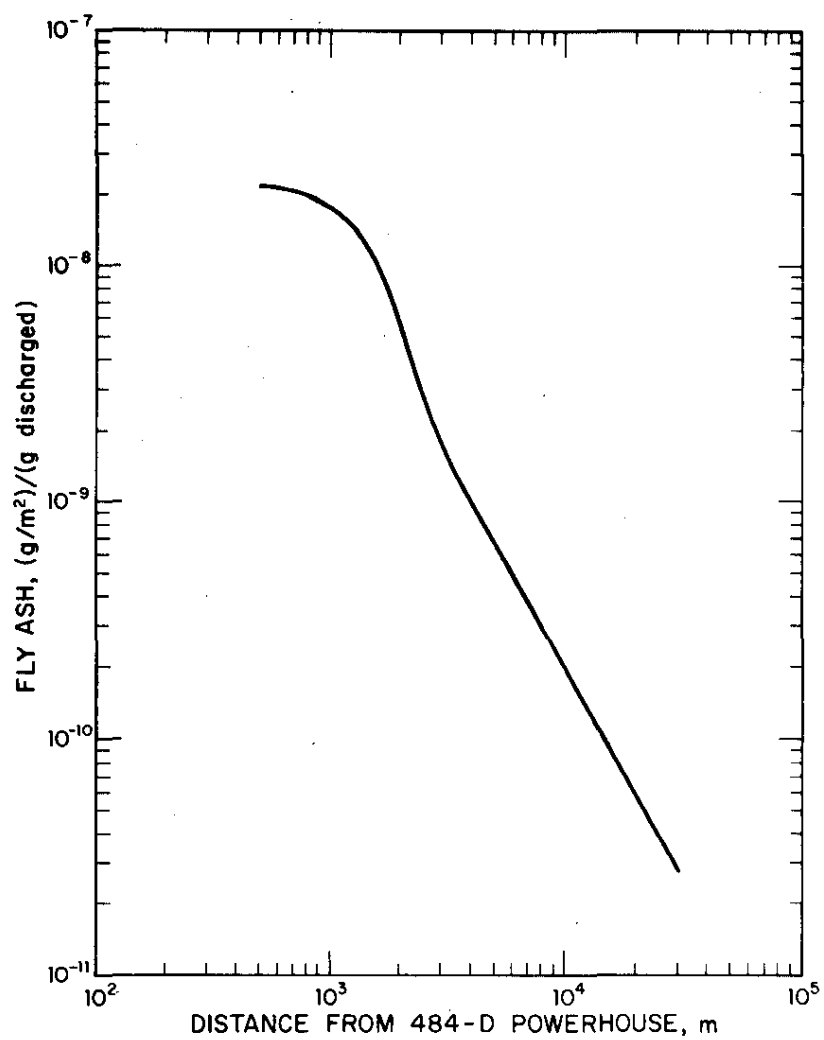


Figure 1. Estimated deposition of fly ash

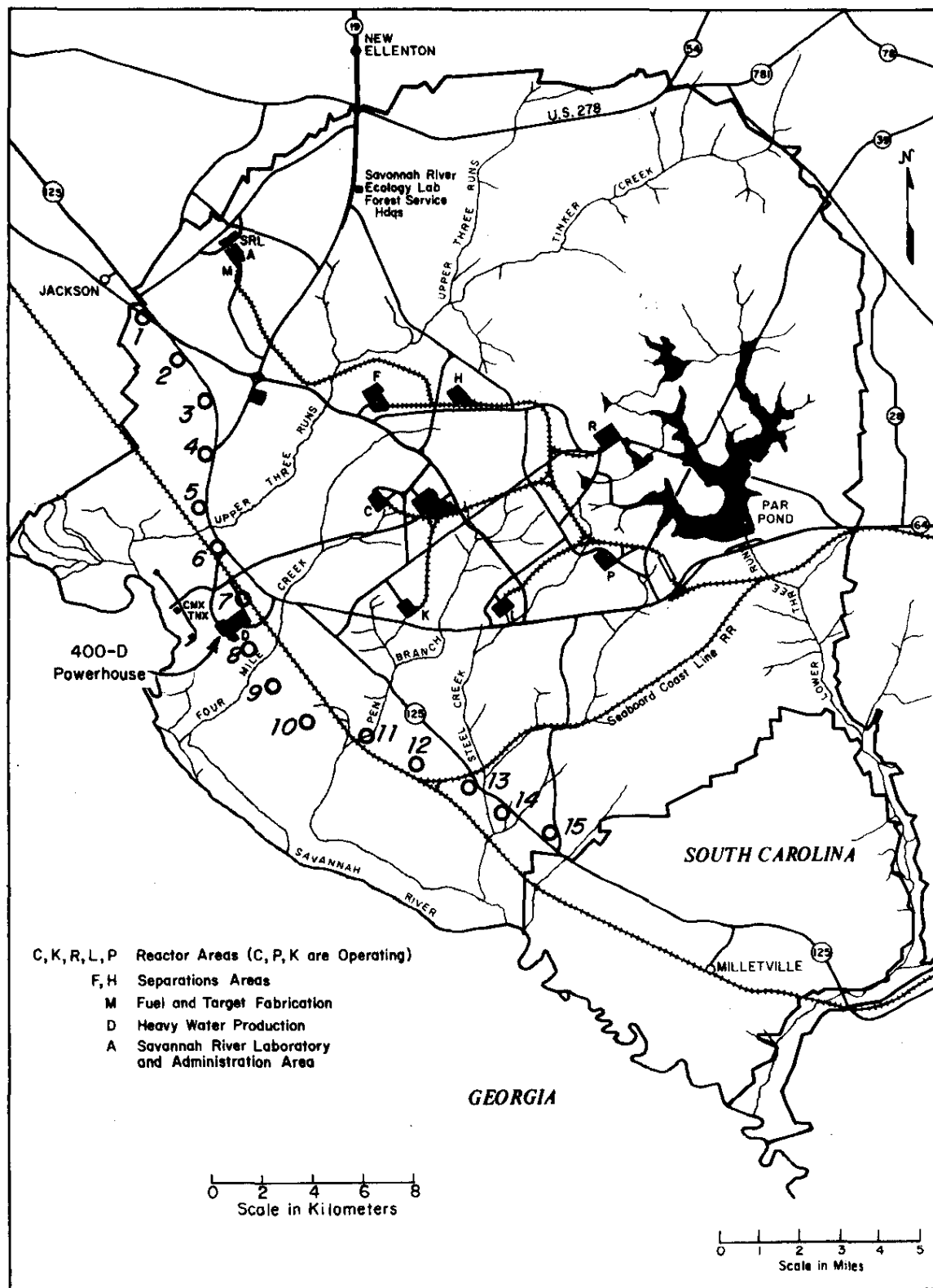


Figure 2. Environmental sampling locations

Table 3. DISTANCES FROM EACH EMISSION LOCATION
TO EACH SAMPLING LOCATION, km

| <i>Sampling location</i> | <i>D^a</i> | <i>F</i> | <i>H</i> | <i>C</i> | <i>K</i> | <i>P</i> | <i>A</i> |
|------------------------------|----------------------|----------|----------|----------|----------|----------|----------|
| 1 | 12.3 | 8.5 | 11.7 | 10.6 | 14.7 | 19.3 | 3.2 |
| 2 | 10.2 | 7.3 | 10.8 | 9.0 | 12.8 | 17.8 | 4.7 |
| 3 | 8.3 | 6.2 | 9.7 | 7.2 | 10.9 | 16.3 | 6.1 |
| 4 | 6.3 | 6.8 | 10.0 | 6.5 | 9.7 | 15.8 | 8.1 |
| 5 | 4.4 | 8.0 | 11.0 | 6.7 | 8.7 | 15.7 | 10.0 |
| 6 | 2.2 | 8.9 | 11.2 | 6.5 | 7.4 | 14.9 | 12.0 |
| 7 | 1.5 | 9.2 | 11.3 | 6.2 | 6.0 | 13.8 | 13.5 |
| 8 | 3.5 | 11.1 | 11.8 | 6.7 | 6.1 | 13.0 | 15.1 |
| 9 | 4.1 | 12.1 | 12.5 | 8.6 | 5.9 | 13.4 | 17.4 |
| 10 | 6.0 | 13.6 | 14.5 | 9.8 | 6.1 | 13.2 | 14.2 |
| 11 | 8.0 | 14.0 | 14.2 | 10.0 | 5.9 | 11.6 | 20.1 |
| 12 | 10.0 | 15.0 | 15.0 | 11.1 | 6.6 | 11.1 | 21.6 |
| 13 | 12.0 | 16.0 | 15.5 | 12.1 | 7.7 | 10.4 | 24.0 |
| 14 | 14.0 | 17.4 | 16.7 | 13.8 | 9.4 | 10.8 | 26.0 |
| 15 | 16.0 | 19.0 | 18.1 | 15.5 | 11.2 | 11.4 | 27.9 |

^a These distances differ slightly from those in the other parts of this report. It was necessary to select sites with the three species of vegetation collected in the study.

Table 4. 1974 FLY ASH EMISSIONS

| <i>Area</i> | <i>Coal consumption, tons/yr</i> | <i>Emission fraction^a</i> |
|-------------|--------------------------------------|--------------------------------------|
| D | 385,431 | 0.8070 |
| F | 47,020 | 0.0165 |
| H | 30,946 | 0.0108 |
| C | 7,906 | 0.0068 |
| K | 40,676 | 0.0654 |
| P | 45,410 | 0.0852 |
| A | 20,403 | 0.0082 |

^a Based on emission tests for each boiler type.

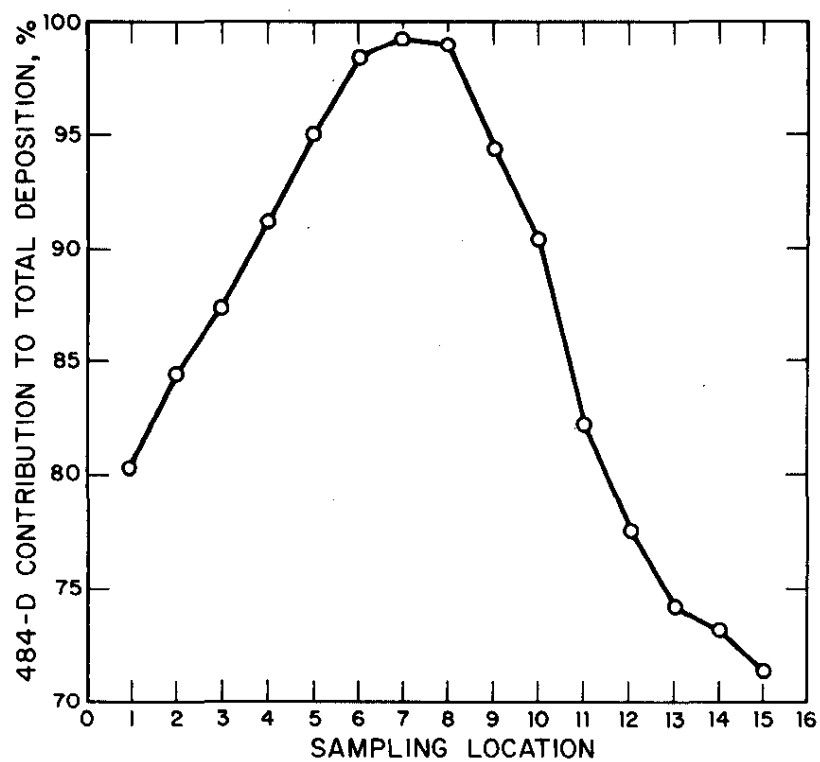


Figure 3. 484-D contribution to total fly ash deposition

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APPENDIX B

SUMMARY OF STATISTICAL REGRESSION AND CORRELATION ANALYSES

RELATION BETWEEN THE LOGARITHM OF THE CONCENTRATION OF THE TRACE ELEMENTS IN SOIL AND THE LOGARITHM OF THE DISTANCE FROM THE POWERHOUSE STACKS

| Element | <i>Correlation coefficient, r, for different soil depths</i> | | |
|---------|--|--------------------|--------------------|
| | <i>0-7.5 cm</i> | <i>7.5-15 cm</i> | <i>15-30 cm</i> |
| Ba | -0.62 ^b | -0.53 ^a | -0.56 ^a |
| Sr | -0.62 ^b | -0.74 ^b | -0.50 ^a |
| Mn | -0.55 ^a | -0.67 ^b | -0.66 ^b |
| Zn | -0.39 | -0.49 | 0.05 |
| V | -0.26 | -0.34 | -0.16 |
| Ce | -0.002 | -0.12 | 0 |
| Cr | 0.06 | -0.26 | -0.17 |
| As | -0.43 | 0.01 | -0.07 |
| Cu | -0.64 ^b | -0.48 | -0.31 |
| Ni | -0.38 | -0.20 | -0.38 |
| Ga | -0.25 | -0.36 | -0.11 |
| La | -0.09 | 0.06 | -0.02 |
| Co | -0.46 | -0.02 | -0.21 |
| Pb | 0.19 | -0.22 | 0.03 |
| Br | 0.28 | 0.30 | -0.02 |
| Sc | -0.25 | -0.28 | -0.08 |
| Th | -0.02 | 0.42 | 0.13 |
| Sb | -0.22 | 0.02 | -0.12 |
| Be | -0.43 | -0.53 ^a | -0.33 |
| Sm | -0.09 | 0.12 ^b | -0.39 |
| Se | -0.43 | -0.66 ^b | 0.36 |
| Cs | -0.16 | -0.28 | -0.06 |
| U | -0.20 | 0.26 | -0.14 |
| Eu | -0.17 | 0.08 | -0.15 |
| Yb | -0.01 | 0.20 | -0.05 |
| Tb | 0.29 | 0.33 | -0.43 |
| Hg | -0.69 ^b | 0.39 | -0.40 |

^a Significant at the 0.05 level of significance.

^b Significant at the 0.01 level of significance.

RELATION BETWEEN THE LOGARITHM OF THE CONCENTRATION OF
THE TRACE ELEMENTS IN VEGETATION AND THE LOGARITHM OF
THE DISTANCE FROM THE POWERHOUSE STACKS

| Element | Correlation coefficient, <i>r</i> , for different species of vegetation | | |
|---------|--|--------------------|--------------------|
| | <i>Dog fennel</i> | <i>Panic grass</i> | <i>Broom sedge</i> |
| Ba | -0.39 | 0.21 | -0.04 |
| Sr | -0.20 | -0.59 ^a | -0.35 |
| Mn | -0.06 | 0.06 | -0.38 |
| Zn | -0.02 | 0.15 | 0.01 |
| V | -0.78 ^b | -0.06 | -0.10 |
| Cr | 0.04 | -0.005 | 0.19 |
| Cu | -0.15 | -0.37 | 0.23 |
| Ni | -0.06 | -0.28 | -0.06 |
| Co | -0.60 ^a | -0.40 | 0.29 |
| Pb | 0.34 | 0.10 | 0.25 |
| Br | 0.65 ^b | 0.09 | -0.20 |
| Sc | -0.49 | -0.09 | 0.02 |
| Mo | -0.73 ^b | -0.27 | -0.34 |
| Be | -0.63 ^b | -0.37 | -0.19 |
| Se | -0.23 | -0.08 | 0.26 |
| Hg | 0.21 | -0.14 | 0.74 ^b |
| Cd | -0.26 | 0.20 | 0.20 |

^a Significant at the 0.05 level of significance.

^b Significant at the 0.01 level of significance.

RELATION BETWEEN THE LOGARITHM OF THE CONCENTRATION OF
THE TRACE ELEMENTS IN GROUND WATER AND THE LOGARITHM OF
THE DISTANCE FROM THE POWERHOUSE STACKS

| <i>Element</i> | <i>Correlation coefficient, r</i> |
|----------------|---------------------------------------|
| Sr | 0.26 |
| Mn | -0.54 ^a |
| Zn | -0.01 |
| Cr | -0.12 |
| Cu | 0.70 ^b |
| Co | -0.53 ^a |
| Hg | 0.19 |

^a Significant at the 0.05 level of significance.

^b Significant at the 0.01 level of significance.

RELATION BETWEEN THE LOGARITHM OF THE RATIO OF THE CONCENTRATION
OF TRACE ELEMENTS IN VEGETATION/THE CONCENTRATION OF TRACE ELEMENTS
IN SOIL AND THE LOGARITHM OF THE DISTANCE FROM THE POWERHOUSE STACKS

| <i>Element</i> | <i>Correlation coefficient, r</i> |
|----------------|---------------------------------------|
| Ba | 0.65 ^b |
| Sr | 0.67 ^b |
| Mn | 0.57 ^a |
| Zn | 0.44 |
| V | -0.07 |
| Cr | 0.11 |
| Cu | 0.43 |
| Ni | 0.35 |
| Co | 0.05 |
| Pb | 0.14 |
| Br | 0.13 |
| Sc | -0.02 |
| Mo | -0.32 |
| Be | 0.27 |
| Se | 0.16 |
| Hg | 0.49 ^a |

^a Significant at the 0.05 level of significance.

^b Significant at the 0.01 level of significance.