ELEMENTAL DEPOSITION DOWNWIND OF A COAL-FIRED POWER PLANT

LAWRENCE E. WANGEN
Health Research Division, Los Alamos Scientific Laboratory,
Los Alamos, NM 87545 U.S.A.
and
MICHAEL D. WILLIAMS
John Muir Institute, Santa Fe, NM 87501 U.S.A.

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Abstract. Dry deposition of fly ash emitted by a coal-fired power plant has been calculated using a surface depletion Gaussian plume model. The subject plant is located in the southwestern United States. Soil samples collected downwind of this power plant have been chemically analyzed for selected trace elements (As, B, F, Hg, Se, Sr, U, and V) to determine concentration vs. distance trends. Gaussian plume deposition calculations predict very little increase of trace element concentrations in soils, except for those elements highly concentrated in fly ash emissions compared to soils. Trace element soil concentrations as a function of downwind distance generally confirm these predictions, with the possible exception of Se.

1. Background

Rapid increases in the combustion of fossil fuels for production of electricity during recent decades have led to concern over the possibly hazardous effects of a concomitant release of trace elements to the environment. Andren and Klein (1975), Bertine and Goldberg (1971), and Klein and Andren (1975) suggest that environmental mobilization of certain trace elements resulting from coal combustion may be approaching or even exceeding that due to natural causes. Environmental effects of trace elements due to ground deposition of atmospheric fly ash emissions have received some attention in this regard; however the results of such studies have been ambiguous. Anderson *et al.* (1975), Connor *et al.* (1976a, b), Keith *et al.* (1974), and Klein and Russell (1973) report increased concentrations of certain elements in soils and vegetation downwind of power plants. Bradford *et al.* (1978) and Lindberg *et al.* (1975) report no elemental increases ascribable to power plant emissions.

Recent investigations by Anderson *et al.* (1975), Connor *et al.* (1976a, b), and Keith *et al.* (1974) around coal-fired power plants in the Rocky Mountain region are particularly relevant to this discussion. These studies show that the concentrations of As and Sb in surface soils, and of Co, F, Se, Sr, U, and V in certain species of vegetation, decrease with distance away from power plants, and suggest that these concentration trends may be attributable to power plant emissions.

We can calculate the amount of airborne material deposited on the ground from an elevated release of material (Slade, 1968). The most common procedure is to use a climatological-type, Gaussian plume, atmospheric transport and diffusion model to calculate average ground level pollutant concentrations. These ground level concen-

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trations are then used to estimate the amount of material deposited to the ground by means of the deposition velocity concept. Bolton *et al.* (1974) and Vaughan *et al.* (1975) made calculations for fly ash deposition from coal-fired power plants. They indicated that very little increase in total soil concentrations of chemical elements should be expected as a result of fly ash deposition to the ground.

In this paper the results of computations estimating the magnitude of fly ash deposition onto soils downwind of a southwestern power plant are reported. The pattern of these deposition estimates was used in the experimental design of a sampling scheme aimed at determining concentration vs. distance relationships for various chemical elements in soils. The objectives of this study were: (1) to compute the expected increase in soil concentration of selected elements due to dry deposition of fly ash for various deposition velocities using a source depletion Gaussian Plume model; and (2) to determine if elemental concentrations in surface soil downwind of this power plant are inversely related to distance.

2. Study Area

The subject power plant is located in northwestern New Mexico near the confluence of the San Juan and Chaco Rivers. It consists of five, coal burning, steam electric generating units – two 175-MW units, one 225-MW unit, and two 800-MW units. At full load, these five units consume about 25.4×10^6 kg of sub-bituminous coal per day. Ash content of this coal is about 22%. Particulate control of stack emissions on the first three units is accomplished by venturi scrubbers, designed to remove 99.2% by weight of the particulate matter in the flue stream. These scrubbers were installed in late 1971 and early 1972. Previously, since 1963, these three units were equipped with mechanical particulate collectors, which had a particulate collection efficiency of 77%. Particulate control on the two 800-MW units consists of 'cold-side' electrostatic precipitators, rated at 97% particulate removal. These units have been in operation since 1970.

3. Materials and Methods

3.1. FLY ASH DEPOSITION CALCULATIONS

Average annual air pollutant concentrations at ground level, $\bar{C}(X, \theta)$, for a given wind sector (represented by wind direction θ) were calculated based on the expression

$$\bar{C}(X,\theta) = \sum_{i=1}^{S} \sum_{j=1}^{N} \frac{(2\pi)^{1/2} (\sigma_z)_i C_{ij}(X, \theta, Z; H) f(\theta, i, j)}{(2\pi X/NS)} , \qquad (1)$$

where $C_{ij}(X, \theta, Z; H)$ is the wind sector average Gaussian plume concentration estimate at a distance (X) from a source located at $X = 0$, $Z = H$: The vertical despersion coefficient (σ_z) is dependent on stability. The i and j refer to stability and wind speed class, respectively, and $f(\theta, i, j)$ is the joint frequency of occurrence of wind direction, wind speed, and stability. Sixteen wind direction sectors and six wind speed classes were used. Wind speed classes were represented by speeds of 1, 3, 5, 7, 9, and 11 m s^{-1}. Six stability categories were used, corresponding to categories A, B, C, D, E, and F as reported by Turner (1969). Wind and stability data were taken from Beck (1975) and are based on pilot balloon soundings and 200-ft tower data collected at the power plant site. Stabilities were given in terms of stable, neutral, and unstable. These were apportioned into the categories A, B, C, D, E, and F using appropriate frequencies of each as determined from local airport data. These airport data stability assignments are based on the prescription given by Turner (1969). Plume rise was estimated using procedures given by Briggs (1969). Plume rise from the different stacks was treated separately and was also calculated as a function of stability, wind speed and appropriate stack parameters.

Given ground level contration, dry deposition was calculated using the expression

$$D_{ij}(X, \theta, Z;H) = V_D C_{ij}(X, \theta, Z;H) \tag{2}$$

where D is mass deposited, V_D is the deposition velocity and $C_{ij}(X, \theta, Z;H)$, X and θ are as above. V_D can be thought of as a rate of delivery of a given material, gas or particulate, to the ground. It is a function of several variables such as particle size or gas reactivity, type of surface, atmospheric stability, and wind speed, and should be given with respect to some reference height (Sehmel and Hodgson, 1976). A range of V_D from 0.1 to 10.0 cm s^{-1} was used in the original calculations in an effort to cover all likely deposition velocities.

Conservation of mass was accomplished using a source depletion concept (Slade, 1968) whereby the original source strength (Q_o) was reduced by an amount of mass equivalent to that mass deposited on the ground. That is, the source strength used to calculate the concentration of pollutant at a distance (X) from the source was calculated conceptually by an expression of the form

$$Q(X) = Q_o - \int_o^X \frac{2\pi}{16} \xi D_{ij}(\xi, \theta, Z_j H) \, d\xi . \tag{3}$$

Here ξ is the integration variable.

3.2. ENRICHMENT RATIOS

Comparison of elemental mass concentration in fly ash and soil was conveniently accomplished by calculation of enrichment ratios. These are defined as follows:

$$F_k = \frac{(C_{\text{fly ash}})_k}{(C_{\text{soil}})_k}, \tag{4}$$

where F_k is the enrichment ratio of element k in fly ash as compared to soil. A large F_k means the mass concentration of element k is much greater in fly ash than in soil.

3.3. REGRESSION MODEL

Mean elemental concentrations for surface and subsurface soil samples were analyzed for distance trends according to a method based on regression analyses as described by Anderson *et al.* (1975) and Connor *et al.* (1976a). Prediction equations were calculated using the expression

$$\log (C)_k = \hat{a} + \hat{b} \log X \, , \tag{5}$$

where $(C)_k$ is the estimate of the concentration of element k at a distance of X km from the power plant.

This model is based on the assumption that beyond the distance of maximum deposition trace element soil concentration impacts should decrease with increasing distance from the power plant if the power plant is a significant source of that element. (An area of low deposition at distances close to the plant is expected as a result of pollutant release from elevated stacks.) Surface and subsurface soil data were individually analyzed for each element. Each regression was tested for a nonzero slope using the student t statistic defined by

$$t = \frac{\hat{b}}{S_b} \tag{6}$$

where $\hat{b}$ is the slope of the regression line and S_b is the standard error of $\hat{b}$. The appropriate number of degrees of freedom is $n-2$, with n the number of samples.

3.4. SOILS SAMPLING AND ELEMENTAL ANALYSIS

Surface and subsurface samples were collected at approximate distances of 8, 12, 16, 24, 40, 70, and 120 km southeast of the power plant. The selection of sampling locations was based on a 1.5 to 2-fold decrement in mass deposition as predicted by the Gaussian plume model calculation. Collection of samples closer than 8 km was precluded due to surface disturbance of soils as a result of strip mine and construction activities. Two 3-sample composites of both surface and subsurface soils were collected from within an area of approximately 5000 m^2 at each distance. Surface soil samples were collected from the top 1 cm after scraping away plant debris. Subsurface soil was defined as that occurring at and below the zone of carbonate accumulation. These depths ranged from 25 to 150 cm dependent on rainfall and soil permeability.

All samples were dried under purified nitrogen, sieved (2 mm), and homogenized on a roller mill prior to taking aliquots for elemental analyses. The selection of trace elements was based on enrichment factors in power plant emissions relative to soils and distance trends as noted in studies by Anderson *et al.* (1975), Connor *et al.* (1976a, b), and Keith *et al.* (1974). The elements analyzed in this study, and the method of analysis, are presented in Table II.

4. Results and Discussion

4.1. DEPOSITION MODEL RESULTS

Dry deposition of fly ash has been calculated for each of 16 compass directions. A range of V_D (0.1 to 10 cm s^{-1}) was used to provide upper and lower estimates of total deposited material. The direction of maximum predicted deposition lies in a 22.5° sector centered southeast of the power plant. The estimated total mass of fly ash deposited in this sector during the 13 yr lifetime of this power plant is presented in Figure 1, for deposition velocities of 1 and 3 cm s^{-1}. A V_D of 1 cm s^{-1} probably constitutes a fairly realistic estimate for fly ash particles in this region (Sehmel and Hodgson, 1976). These values are shown graphically to illustrate the predicted deposition pattern. Note the existence of a maximum at about 4 km preceded by an area of low predicted deposition. This results because stack effluents released at altitude require a significant amount of time to reach the ground.

The dry ash deposition values in Figure 1 probably constitute upper limits because various weathering processes are not included in the model calculations. For example, wind and water erosion, and other redistribution processes, which would be expected to transport deposited material away from the deposition site, are not taken into account. The data in Figure 1 assume that fly ash is deposited on the soil and remains there while being uniformly mixed with the top 1 cm of soil mass. The distance of maximum deposition occurs about 4 km from the power plant, and, for

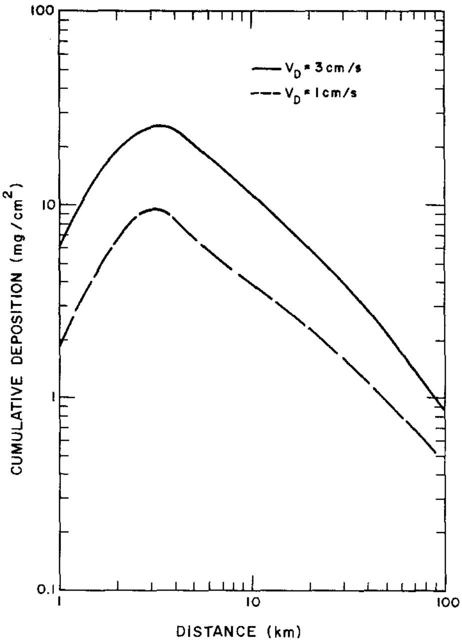

Fig. 1. Calculated 13 yr cumulative fly ash deposition southeast of power plant.

the $V_D = 3$ cm s^{-1} case, mass deposition amounts to 24 mg cm^{-2} at this location. For the $V_D = 1$ cm s^{-1} case, approximately 10% of total emissions, during times when plume flow is to the southeast sector, are deposited on the ground within 50 km of the power plant. This fraction is consistent with the value of 6%, for all directions, given by Vaughan *et al.* (1975). The corresponding percentage for the $V_D = 3$ cm s^{-1} case is 25%.

Hypothetical increases in natural soil trace element concentrations due to dry deposition of stack emissions were calculated using a range of enrichment factors (Equation (4)) and the fly ash deposition values presented in Figure 1. These calculations assume a soil density of 1.5 g cm^{-3} and uniform mixing with the top 1 cm of soil, and are tabulated in Table I for the $V_D = 3$ cm s^{-1} case only. For example, the concentration in contaminated soils of an element with an enrichment factor of 10 would be increased by 15.9% at a distance of 4 km over its concentration in natural soils not impacted by fly ash deposition. Corresponding concentration increases for $V_D = 1$ cm s^{-1} are approximately a third of those given in Table I.

If it is assumed that 50% is a reasonable estimate of the coefficient of variation for trace element concentrations in natural soils, over distance scales of 50 to 100 km, it is expected that an elemental concentration increase of about 50% would be required to detect an impact on contaminated soils. (Considerable data concerning trace element variation in soils can be found in Anderson *et al.* (1975).) These calculations (Table I) show that an elemental enrichment factor of around 30 provides concentration increases in contaminated soils of this magnitude. Using a deposition velocity of 1 cm s^{-1}, an enrichment factor of nearly 100 would be required to give an increase in concentration of 50%.

TABLE I

The percentage increase in elemental content of soils due to hypothetical enrichment ratios in deposited fly ash[a]

Distance (km)	Enrichment ratio 2	5	10	50	100
1	0.8	1.9	3.8	19	38
2	2.6	6.6	13.1	65.5	131
4	3.2	8	15.9	79.5	159
6	2.4	6.1	12.1	60.5	121
8	2	5	10	50	100
10	1.5	3.9	7.7	38.5	77
14	1.1	2.9	5.7	28.5	57
20	0.8	2.1	4.1	20.5	41
30	0.5	1.3	2.6	13	26
40	0.4	1	1.9	9.5	19
50	0.3	0.7	1.4	7	14
70	0.2	0.5	0.9	4.5	9
100	0.1	0.3	0.5	2.5	5

[a] Data are for the case of $V_D = 3$ cm s^{-1}.

TABLE II

Elemental concentrations of selected elements in surface (S) and subsurface (SS) soils southeast of the power plant[a]

Element		Distance (km)							
		8	12	16	24	40	70	120	Analytical method
% Al	S	3.5	4.6	4.2	4.3	4.2	5.0	4.2	INAA
	SS	4.0	4.7	4.2	4.5	4.2	5.2	4.6	
ppM BA	S	673	747	828	678	814	740	706	INAA
	SS	625	756	756	848	810	904	752	
% Ca	S	0.51	0.74	0.56	0.52	0.48	0.51	0.26	INAA
	SS	4.1	2.7	1.7	2.6	1.1	0.88	0.52	
% K	S	1.8	2.3	2.6	2.0	2.7	2.3	2.6	INAA
	SS	2.3	2.0	2.0	2.2	2.6	3.1	2.4	
% Mg	S	0.34	0.68	0.34	0.46	0.30	0.46	0.26	INAA
	SS	0.80	0.72	0.56	0.64	0.28	0.44	0.40	
ppM Mn	S	152	248	206	277	277	361	270	INAA
	SS	337	278	252	256	287	266	237	
% Na	S	0.84	1.04	1.12	0.96	1.04	1.14	1.00	INAA
	SS	0.88	1.02	1.14	0.98	1.02	1.15	0.96	
% Ti	S	0.12	0.18	0.13	0.19	0.16	0.33	0.28	INAA
	SS	0.18	0.18	0.17	0.20	0.15	0.20	0.22	
ppM As	S	2.1	1.8	2.6	3.2	2.5	2.8	1.6	IENAA
	SS	4.0	1.5	1.5	1.9	4.5	2.8	1.7	
ppM F[b]	S	8	9	10	13	9	10	6	Ion electrode
	SS	14	8	10	10	8	10	8	
ppB Hg	S	9	14	9	13	12	17	13	Atomic absorption spectrometry
	SS	12	10	10	12	9	8	12	
ppB Se	S	39	30	17	40	25	20	15	Neutron activation/chem. separation
	SS	21	23	19	19	23	31	–	
ppM Sr	S	124	192	176	172	156	174	116	INAA
	SS	170	203	201	212	140	149	124	
ppM V	S	20	34	24	28	22	36	27	INAA
	SS	33	39	27	35	24	30	30	
ppM U	S	1.6	2.0	1.6	2.4	1.9	2.4	2.7	Delayed neutron
	SS	1.8	1.8	1.7	2.0	1.8	2.0	2.0	
ppM B	S	20	25	18	29	25	26	22	Prompt gamma neutron activation
	SS	17	21	17	24	17	18	18	

[a] INAA = Instrumental neutron activation analysis, IENAA = Instrumental epithermal neutron activation analysis.

[b] Determinations made on fluoride extracted by 0.1 M citric acid solution.

4.2. ELEMENTAL CONCENTRATIONS AND DISTANCE TRENDS

Mean elemental concentrations for surface and subsurface soils at each distance are listed in Table II. Each of these numbers is an average of the two 3-sample composites. Linear regression results (Equation (5)) for each element are given in Table III.

Calcium and Mg concentrations were inversely correlated with distance from the power plant in both surface and subsurface soils. Most of the remaining major elements, Al, Ba, K, Na, and Ti, increased in concentration in both soil horizons with distance. These trends are attributed to natural distribution patterns. For the

TABLE III

Results of regressing surface (S) and subsurface (SS) soil concentrations on distance

		$\hat{a}$	$\hat{b}$	r^2	Enrichment factor (emission/soil)
Al	S[3]	1.25	0.060	0.22	2.7
	SS[3]	1.31	0.054	0.27	
Ba	S	6.59	0.005	0.00	2.3
	SS[2]	6.42	0.070	0.29	
Ca	S[1]	0.07	−0.234	0.50	3.9
	SS[1]	2.58	−0.656	0.68	
K	S[3]	0.59	0.08	0.19	0.3
	SS[2]	0.47	0.112	0.34	
Mg	S[4]	−0.50	−0.14	0.16	2.5
	SS[1]	0.28	−0.28	0.48	
Mn	S[1]	4.81	0.21	0.55	1.1
	SS	5.80	−0.06	0.10	
Na	S[4]	−0.14	0.05	0.16	1.2
	SS	−0.08	0.03	0.07	
Ti	S[1]	−2.75	0.32	0.70	2.5
	SS	−1.89	0.06	0.11	
As	S	0.84	−0.02	0.00	14
	SS	0.82	−0.01	0.00	
F	S[4]	2.47	−0.09	0.07	2.6–57
	SS	2.63	−0.12	0.15	
Hg	S[2]	1.94	0.17	0.36	25–590
	SS	2.45	−0.04	0.02	
Se	S[4]	4.08	−0.26	0.42	65–500
	SS[4]	2.63	0.15	0.44	
Sr	S	5.22	−0.05	0.06	2.6
	SS[1]	5.69	−0.17	0.54	
V	S[4]	3.03	0.08	0.13	5.6
	SS[4]	3.66	−0.07	0.13	
U	S[1]	0.16	0.16	0.49	5.9
	SS[3]	0.47	0.05	0.24	
B	S	1.29	0.05	0.09	25–390
	SS	1.31	−0.03	0.04	

[1] Slope different from zero at ≤ 0.01 level of significance.
[2] Slope different from zero at ≤ 0.05 level of significance.
[3] Slope different from zero at ≤ 0.10 level of significance.
[4] Slope different from zero at ≤ 0.20 level of significance.

most part these results agree with those reported by Connor *et al.* (1976b). Regression results for trace elements As, B, F, Hg, Se, Sr, U, and V showed few notable trends (Table III). No significant negative slopes ($P \leq 0.10$) were found for any of these trace elements in surface soils.

Strontium concentrations decreased in subsurface soils ($P \leq 0.05$), and are suggestive of a negative trend in surface soils. However, since Sr and Ca chemistry are similar, and Ca also showed statistically significant negative slopes, Sr trends should be attributed to natural distribution patterns.

Fluoride concentrations decreased with increasing distance in both soil horizons, suggesting a parent mineralogical control. Uranium and V concentrations increased away from the power plant, whereas those of As and B showed no discernible trends.

Mercury, which is expected to be highly enriched in power plant emissions (Crockett and Kinnison, 1977), showed a positive slope in surface soils ($P \leq 0.05$) and no trend in subsurface soils. The environmental behavior of Hg is difficult to predict because the volatility of Hg, and many of its compounds may result in considerable movement of Hg across environmental compartments. Additionally, Hg emissions from power plants are believed to be principally in the gaseous state. Thus, though Hg may be highly enriched in power plant stack emissions, this enrichment was not reflected in local soil concentrations. This lack of any trend in Hg soil concentrations is in agreement with recently published data by Crockett and Kinnison (1977).

Selenium data suggested a negative trend in surface soils and a positive one in subsurface soils ($P \leq 0.20$). These are noteworthy in spite of low significance levels because, in contrast to the surface trend, the subsurface concentrations reflect a natural or geologic trend of increasing concentrations away from the power plant. Figure 2 shows plots of Se with distance for both surface and subsurface soils. These plots evidence considerable scatter in the Se data and suggest that interpretation of distance trends using these data must be made with considerable reservation.

Based on deposition model calculations, we suggest above that contamination of soils by fly ash should not be expected except for those elements with enrichment factors in the range of 30 to 100. The last column of Table III lists estimated enrichment factors for the elements considered in this study. Note that only Hg, Se, and possibly, B and F have enrichment factors greater than 30. In view of these enrichment factors, the general lack of distance trends that can be attributed to power plant emissions is not surprising. The enrichment values listed for Hg and F are based on the assumption that vaporous forms will deposit in a manner similar to fly ash and should therefore be viewed as equivalent enrichment factors. The large range in the Se value results from uncertainty in the concentration of Se in fly ash emissions and soils.

Any attempt to interpret elemental data must consider the sources of variability (Table IV) in measured concentration. As mentioned above, two 3-sample composites

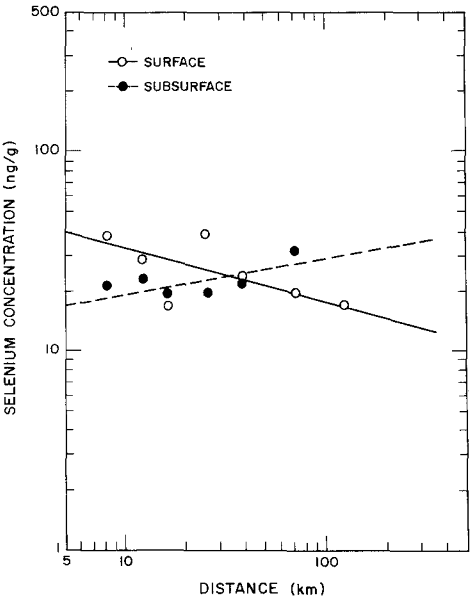

Fig. 2. Selenium concentrations in surface and subsurface soils southeast of power plant.

were collected at each of the seven distances southeast of the power plant. These seven pairs were used to estimate the coefficient of variation for trace element concentrations in soils over distance scales of 0 to 1 km (labeled as field in Table IV). Following laboratory preparation of these samples, four were split and each split was analyzed separately to obtain an estimate of coefficients of variation due to sample inhomogeneity and chemical analyses, i.e., laboratory procedures. These data (Table IV) show that there is large uncertainty associated with the concentrations of many of the trace elements. In nearly all cases coefficients of variation due to laboratory procedures are equivalent to coefficients of variation calculated from field duplicates collected within a few hundred meters of one-another. Since coefficients of variation for field duplicates include the uncertainty due to laboratory errors, we conclude that most of the variability found in field duplicates is, in fact, due to sample inhomogeneity and/or chemical analyses. It is likely that the compositing procedure during sample collection may have reduced some of the variability in field samples collected at a given distance from the power plant. The purpose of compositing was to minimize sample variation. Uncertainties in Hg and Se concentrations are of particular importance. Selenium is a very difficult element to chemically analyze with precision at the levels found in these samples, a fact aptly demonstrated by a laboratory coefficient of variation of 22%. In contrast, a coefficient of variation of 9% for Hg suggests a considerably higher degree of reliability in any conclusions drawn from Hg concentrations in soils.

TABLE IV

Uncertainty estimates from laboratory and field duplicates[a]

	Coefficient of variation (%)	
	Laboratory	Field
Al	6	6
Ba	9	9
Ca	6	16
K	5	11
Mg	13	6
Mn	8	8
Na	6	6
Ti	7	6
As	29	54
B	8	8
F	14	31
Hg	9	12
Se	22	33
Sr	8	7
U	3	14
V	10	4

[a] Field uncertainty includes that due to sample inhomogeneity and chemical analysis.

5. Conclusions

We conclude from this study that (1) estimates of fly ash deposition to soils, based on Gaussian plume model calculations, indicate that very little change in total soil concentrations will result from such deposition, even for elements highly enriched in fly ash, and (2) this method, i.e., measuring the distribution pattern of total elemental concentrations in soils around a large coal-fired power plant, may not be sensitive enough to detect a power plant impact on local soils. It is possible that analysis of the soluble fraction of soil elements or the soil clay fraction could be a more sensitive indicator of power plant impacts.

Other tentative conclusions can be made concerning studied trace elements. Since Hg concentrations in surface soils increase with distance southeast of the power plant, it appears that this power plant is not significantly altering the natural Hg budget in area soils. In contrast, these data suggest that surface soil Se concentrations near the power plant may be slightly increased due to deposition. Thus, based on these results, the possibility that the power plant may be adding to total soil Se concentrations cannot be accepted or rejected. However, statistically significant Se concentration decreases have been reported by Connor *et al.* (1976b), in a grass southeast of this power plant. Selenium concentrations in soils were not reported, apparently because of the detection limit of the analytical procedure used. For all of the other elements considered, our data indicate that the concentrations in local soils are not being measurably altered by power plant emissions.

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