

Contamination of Soil and Vegetation by Atmospheric Deposition of Trace Elements

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The term "trace elements" usually refers to a heterogeneous group of chemical elements that are present in natural systems at low concentrations. In small quantities, many trace elements are essential to biological activities. However, excessive accumulation in the biomass often results in detrimental effects. Several elements, such as cadmium, mercury, lead, arsenic, and chromium are considered biologically nonessential, and their presence in the environment always poses a potential hazard. In light of the recent discoveries of trace element induced chronic injuries to humans and the possibility of bio-accumulation through natural food chains, there is widespread concern of the gradually increasing low level, long range, and possibly continuous exposure of the general public to many trace elements introduced to the environment through man's activities. Evidences of the presence of trace elements in the atmosphere, water, and soil are numerous, but specific assessment of their impact is lacking. This presentation attempts to assess the extent of trace metal contamination in soils and vegetation caused by atmospheric deposition.

Origin and formation of atmospheric trace elements.—The atmosphere consists of primarily nitrogen and oxygen, plus small amounts of carbon dioxide, water vapor, and a few noble gases. Other constituents present in the atmosphere would be considered contaminants. The trace elements in the atmosphere may originate from several sources.

On the earth's surface, trace elements can be found in soil, water, and in vegetation. Volcanic ash also is an important source of atmospheric trace elements. When soil is weathered, water is vaporized, or organic matter is decomposed, trace elements may be fractionated and could enter the atmosphere through the agitation of the earth's surface by air turbulence. The airborne substances will then be distributed by the vertical and horizontal mixing of the atmosphere. The amount of trace elements actually entering the atmosphere through this route is difficult to estimate. Due to differences in the atmosphere's capacity to carry particles of varying sizes, trace elements are sometimes found enriched in certain areas. A review of impurities in the atmosphere of remote regions not yet affected by man's activities shows that the concentrations of naturally emitted trace elements in the atmosphere are rather low (16,25).

Besides the indigenous input, the atmosphere also receives trace elements from man-made sources, commonly termed air pollution. Today, the trace elements introduced through artificial means closely related to human activities, in many cases, far exceed the contributions from the natural sources. This shift of balance occurred with the advent of fossil-fuel utilization and the increased industrial development which accelerated the introduction of previously nonexistent impurities into the atmosphere. Activities specifically recognized as sources of anthropogenic emissions of atmospheric trace elements are the mining and production of industrial materials, combustion of fossil fuels for power and heat, municipal waste incineration, and the operation of motor vehicles.

The anthropogenic release of trace elements into the atmosphere invariably involves either a fuel-combustion process or a high-temperature conversion process of industrial materials which contain trace metals. During high-temperature combustion, many metal elements and their oxides may become volatilized. The volatilized substances would then be condensed and absorbed uniformly onto the surfaces of flue gas particulates which are

formed by the less volatile elements (15,17). Potentially toxic elements such as As, Be, Cd, Cr, Mn, Ni, Pb, Sb, Tl, V, and Zn all have been found in exhausted combustion flue gases. It also has been demonstrated that the trace metal concentration of air-borne particulates increases with decreasing particle size (14). The apparent pattern of concentrating certain trace elements in finer particles is closely related to the volatility (melting points, boiling points, and vapor pressure) of the elemental oxides. Elements of high volatility, such as As, Cd, Cr, Ni, Pb, Sb, Se, Tl, and Zn showed a pronounced concentrating effect (10). Consequently the trace element-enriched fine particulates, not readily captured by emission control devices, would be released and probably remain suspended in the atmosphere for extended periods of time.

A complete cycle of anthropogenic trace elements in the atmosphere consists of three phases: (i) release of trace elements—containing particulates of the source; (ii) transportation and diffusion of airborne trace elements; and (iii) redeposition of trace element-enriched particulates. The transport and dispersal of released particulates in the atmosphere is greatly influenced by local topographically induced mechanical air turbulence and by macro- and micro-meteorological factors. The highly variable motion of air masses is eventually responsible for the ultimate deposition of particulates. When the particle diameter is reduced from 10 μm to 1 μm (a normal range of airborne particulates), the settling velocity at the ground level could be reduced by a factor of almost 100. The redistribution of atmospheric trace elements would start the cycle of accumulation in soils and vegetation.

Trace elements in the urban environment.—Anthropogenic emission of impurities into the atmosphere has resulted in a significant enrichment of trace elements in the urban environment as exemplified by typical air quality data for urban and nonurban communities in the USA as derived from Air Quality Data for 1967 (23) (Table 1). In urbanized communities where anthropogenic activities are centered, potentially hazardous trace elements such as Cd, Cr, Ni, Pb, and Zn not only are more frequently encountered, but also are found in higher concentrations than in nonurban areas. Much of the increase of lead in the urban atmosphere is associated with the use of leaded gasoline in motor vehicles. The higher concentrations of urban atmospheric trace elements also is indicative that higher concentrations will occur in the soils and vegetative cover of the urban environment.

Using Pb as an indicator, Page and Ganje (19) found the lead content of surface soils in southern California metropolitan areas increased several-fold over a period of 30–50 yr. Since lead is used as a gasoline additive, the concentrations of this element in the surveyed areas were found closely related to the calculated motor vehicle traffic density (Table 2). In areas of low motor vehicle density, no apparent increase of Pb in the surface soil was detected. The enrichment of soils by other trace elements at a lesser magnitude in the urban atmosphere also is expected. Based on trace element fallout measurements in 77 midwestern cities during 1968 (7) estimates of the time period required to double a typical background concentration of Cd, Pb, and Zn in soil were calculated for residential, commercial, and industrial areas (Table 3). Lead appears to be the element that is accumulating most rapidly in the urban areas (Table 3). Also, calculations based on data presented by Hunt et al (7) (Table 4) show concentrations of Cd, Zn, and Pb which would occur in or on vegetation if all deposition remained with the harvested crop and the crop was exposed to atmospheric deposition for a 4-mo period. The computations overestimate the probable extent of contamination because rainfall, and wind action will remove a sizeable percentage of the

airborne particulates deposited on vegetation. They do show, however, greater accumulation of Cd and Zn in industrial than in residential or commercial regions. The largest accumulations of Pb within and upon vegetation occur in commercial regions which is a reflection of the much higher motor vehicle traffic densities in commercial than in residential or industrial regions. Although the data are computed from atmospheric concentrations, concentration of the three metals obtained from measurements in urban regions made by Page et al (20) and Lagerwerff and Specht (13) show levels within and upon vegetation of the same order of magnitude as those presented in Table 4.

Besides the general across-the-board increase of trace elements in urban soils, there are segments of the urban community that deserve special attention. Soil and plant samples taken along heavily traveled highways in the urban area usually show severe contamination of roadside soils and vegetative cover by Pb, Zn, Cd, and Ni. Lagerwerff and Specht (13), for example, have observed elevated concentrations of Ni, Pb, Cd, and Zn in surface soil and vegetation close to heavily traveled roads. Table 5 contains data which compare concentrations of certain trace elements in dusts collected from highways to their concentrations found in soil. The data show considerably higher concentrations of Pb, Cd, Cu, and Zn in highway dust than soil, and serve to explain the source of

TABLE 1. Metal concentrations ($\mu\text{g}/\text{m}^3$) of urban and non-urban air in the USA^a

Element ^b	Mean		Maximum	
	Urban ($\mu\text{g}/\text{m}^3$)	Non-urban ($\mu\text{g}/\text{m}^3$)	Urban ($\mu\text{g}/\text{m}^3$)	Non-urban ($\mu\text{g}/\text{m}^3$)
Cd	0.008	<0.001	0.018	<0.001
Cr	0.027	0.003	0.058	0.019
Co	<0.007	<0.007	0.008	<0.007
Cu	0.22	0.03	0.49	0.08
Fe	2.4	0.10	9.5	0.8
Pb	1.8	<0.1	5.5	<0.1
Mn	0.05	<0.01	0.38	0.01
Ni	0.038	0.003	0.210	0.01
V	0.03	<0.001	0.20	0.001
Zn	0.17	0.03	0.94	0.47

^aDerived from Air Quality Data for 1967 (23). Urban community is represented by data from Pasadena, CA and the non-urban community is represented by data collected in Humboldt County, CA.

^bConcentrations of the following elements were less than the limits of detectability or less than the values indicated in parentheses (in μg element/ m^3 air): Sb(0.05), Bi(0.001), Mo(0.01), Ti(0.01).

TABLE 2. Accumulations of Pb in the surface 2.5 cm of soils as related to motor vehicle (M.V.) traffic density in California^a

Location	Concn ($\mu\text{g}/\text{g}$) for soils collected:		
	Between 1919 & 1933 ^b	During 1967	Traffic density ^c (M.V./sq km)
La Verne	17	52	287
La Puente	16	52	594
Whittier	16	39	965
La Habra	17	50	575
Tustin	16	31	534
Riverside	17	38	226
San Bernardino	21	24	246
Hemet	19	21	10
Santa Paula	21	23	31
Meloland	26	25	19

^aDerived from Page and Ganje (19)

^bYear sampled: Whittier and Riverside, 1919; Santa Paula, 1926; San Bernardino, 1927; La Verne, La Puente, Tustin, 1928; Hemet, 1930; Meloland, 1931; La Habra, 1933.

^cComputed by taking total 1967 population within a 17 km radius from the site sampled and dividing by the average number of persons per motor vehicle for southern California.

contamination of these metals in roadside soils and vegetation. Most of the residual contamination is restricted to the surface soil and confined to a narrow corridor along the direction of traffic. The contamination of plant foliage results primarily from surface deposits of particulates. No significant increases of trace elements in the plant tissue due to plant absorption were found (13).

Industrial metal processing.—The air pollution aspect of metal processing long has been focused on emission and control of sulfur dioxide and particulate matter. Processing of industrial metals, however, is an important source of emission of metal elements into the atmosphere. Smelting and sintering of nonferrous metals results in enrichment of Pb, Zn, Cu, Cd, As, and Sb in the nearby environment. The major sources of emission are the ore-melting furnaces in which oxides of metals are formed and volatilized. Airborne dusts and fumes from charging the furnace, transporting scrap material, and sintering and metal-reducing furnaces also are sources of particulate fallout in and near the operation.

TABLE 3. Calculated trace metal elements enrichment of soils resulting from atmospheric deposition in urban areas of midwestern USA^a

Region	Element	Back-ground conc in surface soil ^b ($\mu\text{g}/\text{g}$)	Annual deposition ($\mu\text{g}/\text{g}$)	Estimated time required to double the concn in surface soil	
				Uncropped ^c (yr)	Cropped ^d (yr)
Residential	Cd	0.3	0.0024	125	333
	Zn	50	0.346	145	159
	Pb	15	0.315	48	49
Commercial	Cd	0.3	0.0038	99	130
	Zn	50	0.536	89	98
	Pb	15	0.767	20	20
Industrial	Cd	0.3	0.0045	67	100
	Zn	50	0.757	61	69
	Pb	15	0.591	25	25

^aBased on data by Hunt et al (7).

^bConcentration in surface 15 cm of the soil.

^cAll crop residues returned to the soil.

^dAssuming 300 kg per hectare (dry matter) of crop residues removed annually.

TABLE 4. Maximum metal contamination of vegetation arising from aerial deposition in the urban midwest USA^a

Region	Concentration within and upon vegetation ^b		
	Cd ($\mu\text{g}/\text{g}$)	Pb ($\mu\text{g}/\text{g}$)	Zn ($\mu\text{g}/\text{g}$)
Residential	0.60	79	86
Commercial	0.95	192	142
Industrial	1.13	148	189

^aData derived from Hunt, et al (7) for samples collected in 77 cities with populations between 100,000 and 1,000,000.

^bAssuming complete canopy, and that all particulates deposited remain with the crop.

TABLE 5. Comparison of trace metal concentrations in highway dusts and normal soils

Element	Concn ($\mu\text{g}/\text{g}$)	
	Highway dust	Normal soil
Pb	14,000	15
Cd	10	0.06
Cu	1,250	20
Zn	3,500	50
Ni	60	40

There are numerous published results which show elevated concentration of hazardous trace elements in soil and vegetation close to and downwind from industrial smelting and refining operations (4,6,8,12,21-24). The extent of contamination is greatest close to and downwind from the plant location. Data derived from the U.S. Environmental Protection Agency (23), presented in Table 6 for surface soils and Table 7 for vegetation, are representative of the kind and extent of contamination which occurs. The data, obtained adjacent to a lead smelter which began operations in 1888, show high levels of As, Cd, Pb, and Zn contamination near the plant site. The extent of contamination of soil and vegetation by these metals was greatest in the direction of the prevailing winds and diminished with distance from the plant. Contaminated soils and vegetation were observed at least 10 km from the plant location.

The continuing pattern of deposition over long periods of time could result in accumulation of metal elements in extensive areas. When soils around a lead smelter were sampled after more than 80 yr of continuous operation, heavy contaminations of surface soils (0-5 cm) by Pb, Zn, Cd, and Cu were detected (5). A comprehensive sampling of the area indicated that surface soil enrichment of all four elements occurred over a 5,000 km² area around the smelter. Heavy accumulations of trace elements in both soils and plants, however, were restricted to regions near the source (Fig. 1). The metal deposition patterns appeared to be influenced by the local climatic conditions at each smelter location.

Although the metal-containing particulates in the atmosphere surrounding mining and smelting operations are predominantly of submicron size and tend to remain suspended in air for extended periods of time, large size metal-carrying particles (>5 μm) at elevated concentrations contributing to the heavy accumulation of metal elements in nearby soils and vegetation can be found on days of heavy emission. The heavy contamination of the atmosphere and the soil around metal-smelting facilities always resulted in increased metal concentration in plant tissue in the affected environment (Tables 6 and 7). Vegetation can be contaminated not only by surface deposition on foliage (such as Pb) but also may be accumulated in the tissue through root uptake (such as Cd, Zn, As, etc.).

Coal-fired electric power generation.—Mineral matters are synergetically deposited with the products of fossil fuel combustion (1). Depending on the geological origin of the fossil fuel, almost all

chemical elements could be present with coal and petroleum. The major elements in fossil fuel are Si, Al, Ti, Ca, Mg, Fe, P, S, Na, and K. Probably, the most important minor elements include Zn, Cd, Hg, Cu, Pb, As, Sb, Se, Mo, and B. In oil refining processes in which crude oil is fractionated, most of the trace elements are left with the solid residue. Fuel oils used for power generation, therefore, usually do not contain trace elements at concentrations as great as those in coal. Upon combustion, trace elements in the fuel could be discharged into the atmosphere. At a fossil fuel-fired power plant, large amounts of fuel are consumed. In 1970, electrical power generation in the U.S. consumed an estimated 527 million tons of coal equivalent to 62.3% of the national consumption (3). The resultant discharge from coal-fired power plants, therefore, could constitute a significant source of trace element release to the environment.

During coal pretreatment (washing) where major amounts of mineral matter are separated, the trace element content also can be reduced. Mass balance studies of coal-fired power plants indicated the majority of trace elements (≈99%), except Se, Mo, and Hg, entering the combustion process ended in bottom ash or in captured fly ash (2). Electrostatic precipitators can be extremely effective in capturing the trace element-enriched particulates with diameters greater than 1 μm.

The fraction of trace elements which escape electrostatic precipitators are present in gaseous form or in submicron-size suspended particles. When the flue gas is dispersed by high-rise smoke stacks, only a few percent of the fine particulates would form deposits near the source of discharge. Based on published data for a simulated 3,000 megawatt coal-fired power plant operating with an electrostatic precipitator operating with an efficiency of 99%, the possible trace element fallout in the region of maximum impact adjacent to a coal-fired power plant over a 35-yr life span was calculated and compared to typical concentrations for soils (Table 8). The calculations assumed that particulates emitted from the stack were less than 3 μm in diameter. Concentrations of the various elements for particles less than 3 μm were taken from the data of Ondov et al (18) which were derived from fly ash from a similar source coal. The data show that where efficient emission control devices are employed, the amount of increase of elements in surface soil arising from coal combustion would be considerably less than the typical background concentration range of soils.

A similar analyses of the extent to which vegetation could become contaminated by emission from a 3,000-megawatt power plant is presented in Table 9. The results show that as long as coal-fired power plants are operated with continuous high efficiency (99% or greater) electrostatic precipitators, the probability that vegetation adjacent to coal-fired power plants would contain

TABLE 6. Metal concentrations of surface soil adjacent to an industrial smelting complex^a

Distance from stack (km)	Direction from source	Metal concentration			
		As (μg/g)	Cd (μg/g)	Pb (μg/g)	Zn (μg/g)
0.65	NE	50	56	1525	418
1.3	E	35	21	925	455
4.0	SW	25	6.5	190	126
7.3	W	8	2.0	85	82
Control		6	0.5	16	44

^aFrom: U.S. Environmental Protection Agency (23).

TABLE 7. Metal concentrations of vegetation adjacent to an industrial smelting complex^a

Distance from stack	Direction from source	Metal concentration			
		As (μg/g)	Cd (μg/g)	Pb (μg/g)	Zn (μg/g)
0.65	NE	8.4	7.5	48	52
1.3	E	2.5	8.6	5.4	60
4.0	SW	1.0	1.3	3.0	13
7.3	W	0.4	0.7	1.0	12
Control		0.1	0.1	3.5	6.8

^aFrom: U.S. Environmental Protection Agency (23).

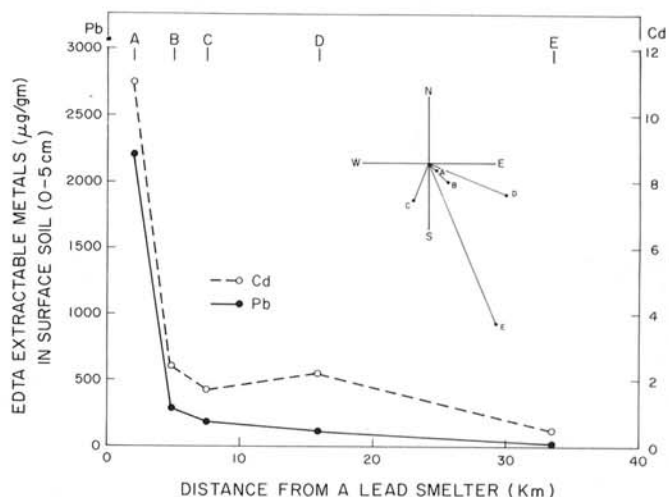


Fig. 1. EDTA-extractable Cd and Pb concentration in the upper 5 cm of soils downwind from a lead smelter (Derived from data presented by Cartright et al [5]).

concentrations of trace elements greater than those typically observed in other regions is remote. Compared to the possible massive contamination caused by metal processing and use of motor vehicles, the trace metal enrichment of soils and vegetation resulting from power generation would be less significant (our analyses and those of Klein and Russell [11]). However, the emission of the trace element enriched fine particulates by power plants could contribute significantly to the overall anthropogenic increase of trace elements in the atmosphere.

CONCLUSION

Review of sources of trace element emission to the atmosphere, reveals that certain anthropogenic activities have resulted in serious contamination of surface soils and vegetation in the vicinity of the sources of discharge. On a regional basis, the deposition of airborne particulate trace elements upon vegetation and soil in industrial,

TABLE 8. Maximum deposition of trace elements onto soil adjacent to a coal-fired power plant^a

Element	Amount deposited		Common concn	
	Annual ($\mu\text{g/g}$)	Lifetime ^b ($\mu\text{g/g}$)	Typical ($\mu\text{g/g}$)	Range ($\mu\text{g/g}$)
Arsenic	0.39	14	6	0.1-40
Cadmium	0.014	0.5	0.06	0.01-7
Lead	0.84	29	15	2-200
Molybdenum	0.15	5.2	2	0.2-5
Selenium	0.59	21	0.2	0.01-2
Uranium	0.087	3.0	1	0.1-10
Zinc	1.65	58	50	10-300
Antimony	0.06	2.1	6	2-10
Beryllium	0.03	1.0	6	0.1-40
Chromium	0.019	6.6	100	5-3,000
Cobalt	0.063	2.2	8	1-40
Copper	0.041	14	20	2-100
Gallium	0.54	19	30	0.4-300
Nickel	0.12	4.2	40	10-1,000
Thorium	0.09	3.2	5	0.1-12
Vanadium	0.98	34	100	20-500

^aDerived from data of Jurinak (9), and Ondov et al (18); coal-fired 3,000 MW power plant in western USA; electrostatic precipitator efficiency 99%.

^bAssuming 35-yr lifetime.

TABLE 9. Maximum trace element contamination of vegetation arising from emissions from a coal-fired power plant^a

Element	Concn in or on vegetation ^b ($\mu\text{g/g}$)	Typical concn ($\mu\text{g/g}$)
Arsenic	0.07	0.4
Cadmium	0.002	0.2
Chromium	0.034	1.5
Copper	0.074	10
Lead	0.15	3
Molybdenum	0.027	1
Selenium	0.107	0.2
Zinc	0.30	25
Antimony	0.011	0.06
Beryllium	0.006	0.03
Cobalt	0.011	<1.0
Gallium	0.096	1.2
Nickel	0.021	5
Thorium	0.016	0.05
Uranium	0.016	0.04
Vanadium	0.18	1

^aDerived from: Jurinak (9), and Ondov et al (18); coal-fired 3,000 MW power plant, Western USA; electrostatic precipitator efficiency 99%.

^bAssuming 100% canopy, 4 mo exposure, particulates deposited are <3 μm , and all particulates deposited remain with the harvested crop.

commercial, and residential regions has not resulted in measurable symptoms of phytotoxicity. Metal-related industrial activities and extensive use of motor vehicles are by far the largest contributors to the contamination. The concentrations of hazardous trace metals in soil and vegetation close to these discharge sources have reached, or potentially could reach levels, which with continuous exposure over prolonged periods of time, could cause serious environmental health problems. With efficient stack emission control devices, trace element contamination of soil and vegetation from coal-fired power plants can be controlled within levels now considered to be environmentally safe.

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