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Ecological Research Series

Heavy-Metal Accumulation In Soil and Vegetation From Smelter Emissions

National Environmental Research Center
Office of Research and Development
U.S. Environmental Protection Agency
Corvallis, Oregon 97330

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HEAVY-METAL ACCUMULATION IN SOIL AND VEGETATION FROM SMELTER EMISSIONS

by

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ABSTRACT

Soil and plant samples were collected along north-south and northeast-southwest transects radiating out from the Tacoma Smelter. The concentrations of lead, arsenic, cadmium and mercury in garden soil decline with increasing distance from the smelter. The concentrations of arsenic and cadmium in vegetation also decrease at increasing distance from the smelter, but lead and mercury concentrations did not appear to be related to distance from the smelter.

The heavy-metal levels in the samples demonstrate the accumulation of large amounts of metals in surface soils and the availability of metals to plants. When these values are compared to "average" heavy-metal contents a deterioration of the quality of the soil and the presence of heavy-metals at levels toxic to some plants is shown.

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Background

The Ruston copper smelter in Tacoma, Washington, has been a significant source of air pollution during much of its 84 year history. The emissions consist of SO_2, SO_3, acid mist and particulates of arsenic, lead, zinc, cadmium, copper and sulfates. In 1970, the smelter was discharging about a ton of particulate matter into the air each day, containing as much as 590 lbs. of lead and 876 lbs. of arsenic. The sulfur emissions amount to 200,000 tons per year.

There are two specific sources of emissions; (1) SO_2, and particulates at a low level around the anode furnaces and plant operations, and (2) SO_2, SO_3, acid mist and particulates from a tall stack (560 ft.). At present the smelter is functioning under intermittant controls in which plant operations are shut down when SO_2 levels are expected to exceed SO_2 air quality standards. During 1973, the plant reduced its operations 25-30 percent.

Winds in the summer are from the north and northwest and carry emissions from the smelter to residential and open areas in west Tacoma, while during much of the fall, winter, and early spring the winds are from the south and southwest and carry effluent to offshore islands (Vashon and Muary Islands) in Puget Sound.

Sampling and Site Evaluation

In response to a request from EPA, Region X for assistance in estimating the terrestrial impact of the smelter on the immediate area, visits were made to the Tacoma smelter area to collect soil and vegetation samples. A total of 68 vegetation and soil samples were collected or received from Puget Sound Air Pollution Control Agency from within six miles of the Tacoma smelter. A majority of the samples were taken from residential gardens on two transects (N-S, NE-SW) at varying intervals from the smelter. The soil samples of the top 0 - 2 inches were taken immediately adjacent to the vegetation samples. All the samples were analyzed by the Consolidated Laboratory Services, NERC-Corvallis using atomic absorption spectroscopy for lead, cadmium, zinc, copper and antimony; flameless atomic absorption for mercury and the Silver Diethyldithiocarbamate colorometric method for arsenic determination.

On August 2 and October 25th and 26th, 1973, site visits were made in the vicinity of the smelter. The area within a mile south and southwest of the smelter is striking in that only a few species of vegetation remain with a complete absence of legumes (alfalfa, clover, etc.) and Douglas fir. The species that predominate are maple, oregon grape, horsetail, laurel hedge, bracken fern, scottish broom and native grasses. At a greater distance from the smelter a larger variety of species is observed, although Douglas fir is absent 4-5 miles southwest of the smelter.

Vegetation injury reported over the years in the vicinity of the smelter has been of the SO_2 and acid mist type. At the time of observation, no SO_2 injury symptoms were observed on vegetation in the smelter vicinity. Peach leaves showed some evidence of injury by acid mist, acid particulate or arsenic. At the Busic residence (5621 N. 46th ST.) margins of peach leaves were red and showed small holes of necrotic tissue that subsequently fell out of the leaf. Small bleached spots were seen adjacent to the margins or covering the leaf. Marginal reddening of peach leaves followed by "shot hole," and then defoliation is indicative of arsenic injury. Acid mist injures leaves by burning small holes in the leaf tissue.

In general, plants reflect the geochemical environment in which they grow. However, mineral elements differ in their availability to plants and plant species differ in their ability to absorb specific elements. Many factors determine the availability of a given element to plants for example, relative abundance, form in which the element is present, soil pH, interaction of elements, physical condition of the soil, environmental factors of temperature and soil moisture and genetic variability. Absorption of toxic metals through the leaf cuticle is a significant source of contaminating in many species of plants.

Effects

A. Copper

The copper content of normal soils range from 1-200 ppm with most in the range of 25-60 ppm. Normal plants contain from 5-20 ppm copper. Copper has long been known to be toxic to plants. Levels in tissues greater than 20 ppm, in general, are indicative of copper excess. Excess copper commonly causes reduced growth and iron chlorosis symptoms in plants and is associated with stunting, reduced branching, thickening and abnormally dark coloration of rootlets of many plants. Clover, alfalfa, poppy, spinach, gladious, corn, bean and squash are known to be sensitive to copper.

The extremely high copper concentrations in grass and other leaf sample from the smelter vicinity represent a possible environmental hazard especially to sensitive plant species. These canot become established and moderately sensitive species will show reduced growth.

The absence of legumes in the smelter vicinity may be related to the levels of copper and other metals in the soil.

B. Zinc

Total zinc in normal soils varies from 10-300 ppm and a wide range of plants have concentrations of from 20-10,200 ppm. In a variety of plants normal levels of zinc range from 25-150 ppm and amounts greater than 400 ppm may indicate toxic levels of zinc.

The levels of zinc present in grass and leaves from the Tacoma smelter appear to be within the high range of normal and probably are not directly affecting plants, but may be important indirectly due to interactions and competition with other heavy metals.

C. Cadmium

Cadmium is present in many soils and is apparently taken up with ease by a great number of plant species, especially the grasses and grains; for example, wheat, corn, rice, oats and millet. Cadmium is also found in peas, beets, lettuce and radishes. The composition of an "average" plant leaf is 0.5 - 0.6 ppm for cadmium and normal soils contain .06 ppm cadmium.

Cadmium is toxic to plants at higher concentrations. In radishes grown in nutrient solutions at concentrations of 100 ppb cadmium, growth of both roots and tops was reduced. At this level, the concentration of cadmium in roots and leaves was 16.2 and 81.2 ppm respectively, but no visible injury was evident.

In Japan, near a zinc refinery, the accumulation of cadmium was extremely high in leaves of plants. Leafy vegetables such as greens, cabbages, the leaves of eggplants, green onions and the leaves of radishes and turnips, contained 3.2 - 56 ppm cadmium. Greens with 56 ppm cadmium were damaged.

Although the levels of cadmium in plant samples analyzed from the Tacoma site are of questionable toxicity to plants, cadmium is present in lettuce and cabbage in sufficient quantity to warrant toxicological evaluations. Cadmium has been associated with a number of serious human afflictions, e.g., hypertension, non-rheumatic heart disease, ostemalacia, proteinuria and emphysema.

The values that are recommended or under discussion for maximum allowable concentrations of cadmium in food is 135 micrograms per kilogram (fresh weight) or approximately 1.35 parts per million (dry weight). The cadmium levels in the cabbage family in the washed samples from Tacoma ranged form 1.2 - 8.2 ppm (dry weight), with a mean value of 3.8 (dry weight), three to seven times higher than the maximum allowable concentration.

D. Lead

Lead is present in all soils and plants. Soil contains an average of 10-15 ppm and ranges from 2-200 ppm of lead.

In general, plants respond to lead only to a limited extent. For example, the lead content of strawberries did not change when the lead content of soil was increased from 8 to 59 ppm. In radishes a 10-fold increase in soil lead content increased the lead concentration by a factor of less than two.

The samples analyzed from Tacoma follow this pattern in that lead values were essentially the same in cabbage cauliflower and brussel sprouts regardless of the lead concentrations in the soil.

Some plants showed retarded growth at 10 ppm lead in solution culture studies. Lead reduced the growth of corn in nutrient solutions and is translocated and accumulated in high concentrations in the leaves. Foliar accumulation of lead was 3-8 times greater where phosphate was deficient than were it was sufficent in the root environment. In young corn leaves, 936 ppm of lead were found in the presence of adequate phosphate while in phosphate deficient growth medium, the lead content was 6,716 ppm.

E. Arsenic

The knowledge about the toxicity of arsenic is based on the use of arsenicals over the years as insecticides, herbicides and defoliants. Arsenic accumulates in soils to levels that may be phytotoxic. In treated areas soils contain from 1.8 - 830 ppm arsenic, while untreated areas had from 0.5 - 14.0 ppm arsenic.

In soils, toxic amounts of arsenic arrest the germination of seeds, reduce the viability of seedlings and have the greatest effect at the seedling stage. In soils the rate of nitrification in the presence of arsenic is decreased and arsenic is toxic to nitrogen metabolism.

The concentration of soluble arsenic in soil necessary to cause injury varies from 1 ppm for cowpeas, 9 ppm for peas and beans, 2 ppm for barley, and 7 ppm for rice. Sodium arsenite applied to common field sand at 1, 5 and 10 ppm reduced the yield of peas, beans, and corn. Soil levels of 50 - 125 ppm of total arsenic may have a detrimental effect on the growth of beans and strawberries. In apple orchards, normal growth can be expected in soil with less than 50 ppm arsenic. Soil with 50 - 100 ppm arsenic reduces growth 50 percent and soil with over 100 ppm arsenic produces very little growth. Lead arsenate at 1-200 pounds per acre reduced the germination of string beans and lima beans and retarded the seedling growth of many vegetables. Apple seedlings grown in potted soil with 100 - 160 ppm Sodium arsenate were killed. Corn kernels rarely develop at soil concentrations of 80 - 100 ppm arsenic.

The chemical form of arsenic is more important than the total soil arsenic in phytoxic effect. The formation of arsenic compounds is affected by acidity, Fe, Al, Ca, P and humus content of the soil. Soils with high reactive aluminum levels are less phytotoxic even after heavy applications of arsenic. Six of the cabbage and lettuce samples from Tacoma exceed the arsenic tolerance levels and are a possible health hazard.

Arsenic is present in the soil and plant tissue from Tacoma at levels that can be toxic to sensitive and moderately sensitive plant species. Snap bean, lima bean, onion, peas, cucumber, alfalfa and other legumes are highly sensitive to water soluble arsenic. This may account for the fact that legumes are absent from the vicinity of the smelter.

F. Mercury

The mercury content of soils in the United States range from 10-500 ppb, and average 100 ppb. Mercury tends to be retained in the surface layers of the soil due to adsorption by organic and inorganic materials and the low solubilities of mercury salts (phosphate, carbonate, sulfide).

In most plants mercury concentrations range from 10-200 ppb (15 ppb ave.), but plants growing near mercury deposits can contain 500-3500 ppb mercury. Translocation of mercury occurs in many plant tissues, including leaves, fruit and tubers.

Toxicity of mercury to terrestrial plants apparently depends more on chemical form than on its concentration. There are but a few studies available on the toxicity of mercury to specific plants, but small amounts of volatilized mercury are known to be toxic to roses in greenhouses.

The mercury concentrations in the samples from Tacoma are well above the "normal" mercury content. The values are on the threshold of being a serious environmental hazard.

Results

Sample locations and concentrations of heavy metals in garden soil and vegetation are shown in eight Figures 1a-4b and Tables I-III. The highest concentrations of lead, arsenic, cadmium, and mercury in the soil were found close to the smelter. Generally, concentrations of lead, arsenic, cadmium and mercury decline with increasing distance from the smelter, although concentrations at points 1/4 - 1/2 mile from the smelter stack are consistently lower than those at approximately 1/2 - 1 mile due to plume rise and looping at distances from the smelter.

The arsenic concentrations of plant samples follow the same pattern as the soil samples in that the highest levels are found closest to the smelter, and the values decrease at increasing distance from the smelter. The lead and mercury present in vegetation samples did not appear to be related to distance from the stack. The highest mercury concentration in the plant sample was found 2 1/2 miles from the smelter. The lead concentrations in vegetation samples did not differ significantly regardless of location, even though the soil lead concentrations varied widely. The concentration of cadmium in vegetation samples is apparently unrelated to the distance from the smelter. The cadmium concentrations are high in plant samples relative to related soil samples, indicating that the vegetation is actively accumulating cadmium from soils acidified by sulfuric acid.

The high concentrations of sulfur in the grass samples indicate a substantial sulfur enrichment of vegetation, although injury symptoms on vegetation from sulfur were not observed. The expected sulfur content of grass is 2000 - 4000 ppm.

The analysis of vegetation and soil samples demonstrate the accumulation of high concentrations of heavy metals in surface soils and the availability of these metals to plants. The heavy metal content of these samples from the Tacoma sites are many times greater for the "average" all elements analyzed with the possible exception of zinc. All of the elements measured may be toxic to one or more plant species.

Control Measures

Methods of altering soil chemistry are available to minimize or reduce heavy metal uptake by plants from soil. The addition of lime is a common measure used to reduce metal uptake by

1. Decreasing the soil acidity to pH 6.5 may result in the precipitation of heavy metals as hydroxides, carbonates, phosphates, etc., and in immobilizing the heavy metal ions.

2. Cations will compete with the trace elements in the soil for exchange sites of the soil and root surfaces.

3. Liming may promote the capacity of plant roots to form complexes with metal ions.

Adding soil phosphates may be a means to precipitate heavy metals as compounds of limited availability to plants.

Conclusions

1. A major impact of the Tacoma smelter on the soil and vegetation in the vicinity has been observed for many years. Sulfur dioxide emissions over the years have degraded some plant species and altered the composition of plant communities. Heavy metals in soils in the smelter vicinity have undoubtedly contributed to the degradation. With increased controls on sulfur dioxide emissions, the heavy metals content in soil becomes more important to soil toxicity and limits the restoration of natural plant communities.

2. The heavy metals copper, arsenic and cadmium are present in soils in concentrations that are likely to be toxic. These have suppressed the establishment of natural and introduced plant species in contaminated areas. Cadmium and mercury also represent a possible health hazard as constituents of leafy vegetables.

3. Sulfur dioxide probably will have a lesser effect on vegetation as emission controls increase. Plant injury due to high pollution episodes should decrease and be replaced by low level chronic type injury.

General Classification of Toxicity of Elements to Plants /1

<u>Very Toxic</u>: Toxic effects may be seen at concentrations below 1 ppm in nutrient solution (included are Cu++, Hg++, Pb++)

<u>Moderately Toxic</u>: Toxic effects appear at concentrations between 1 and 100 ppm in nutrient solution (included are AsIII, AsV, Cd++ Zn++)

<u>Scarcely Toxic</u>: Toxic effects rarely appear (included are Cl-, I-, Ca++ K+)

II. The Biogeochemistry of the Elements

	(Bowen)/1	(Chapman)/2		
Arsenic	soils - 6 ppm plants - 0.2 ppm moderately toxic to plants	0.3-38 ppm	30% 50% 20%	5 ppm or less 5-10 ppm 10 ppm or more
Cadmium	soils - .06 ppm plants - 0.2 ppm moderately toxic to all organisms			
Copper	soils - 20 (2-100) ppm plants - 14 ppm very toxic to algae, fungi, seed plants	10-200 ppm 5-20 ppm		
Mercury	soils - .03-0.8 ppm plants - .015 ppm very toxic to fungi and green plants			
Lead	soils - 10 ppm plants - 100 ppm very toxic to most plants	2-200 ppm 25-150 ppm		
Zinc	soils - 50 ppm plants - 100 ppm moderately toxic to most plants	10-300 ppm 25-150 ppm		
Sulfur	plants - 3400 ppm	1300-6500 ppm		

1/ H. J. M. Bowen, Trace Elements in Biochemistry. Academic Press, London (1966)

2/ H. P. Chapman (Ed.) Diagnotic Criteria for Plants and Soils. University of California, Div. of Agricultural Sciences (1966)

TABLE I

Heavy metal content of vegetation and soil samples from the vicinity of the Tacoma Smelter. (Concentrations expressed in parts-per-million on a dry weight basis)

Sample No.	Location	Sample	Arsenic	Cadmium	Lead	Mercury	Zinc	Copper	Antimony	Sulfur
7516001	5020 Lexington St.	grass	168	5	160	2	381	608	229	8090
7516002	6102 Park Avenue	grass	56.3	4	51	2	49	350	148	9960
7516003	5309 Ruby Street	grass	396	8	450	8	208	2260	244	21900
7516004	4852 N. 50th St.	grass	797	12	470	8	288	3040	165	13600
7516005	5311 Commercial Ave.	grass	472	16	692	10	190	3150	332	8000
7532001	Court & Baltimore	grass	582	4	250	3.0	113	1400	-	-
7532002	Court & Baltimore	maple leaves	167	2	50	1.3	113	1400	-	-
7532003	5621 N. 46th St.	pear	9	2	10	0.3	49	54	-	-
7532004	"	horsebean pod	7	2	10	0.4	12	230	-	-
7532005	"	horsebean leaves	142	4	55	2.5	180	575	-	-
7532006	"	grape leaves	116	2	49	1.8	50	660	-	-
7532007	"	squash leaves	66	2	19	1.9	140	303	-	-
7532008	"	fig leaves	200	2	49	1.3	48	680	-	-

Table II

Heavy metal content of vegetation and spill samples from the vicinity of the Tacoma Smelter. (Concentrations expressed in parts-per-million on a dry weight basis)

Sample No.	Location	Sample	Arsenic	Cadmium	Lead	Mercury
7544016	5621 N. 46th St.	soil	214	2.9	238	11.0
7544034		cabbage	11.4	2.9	9	0.5
7544017	969 Altadena Dr.	soil	12.2	1.0	9	0.2
7544035		cabbage	1.5	2.7	16	0.7
7544018	1719 Naomi Pl. (Seattle)	soil	7.3	1.8	271	0.2
7544036		cabbage	1.8	1.8	12	0.3
7544037	5141 N. Ruby St.	soil	457	8.3	743	6.8
7544038		lettuce (unwashed)	110	2.3	28	1.0
7544039		lettuce (washed)	68.5	3.1	17	1.2
7545004	Manzanita Beach Muary Island	soil	36.2	1.6	305	0.6
7545001		broccolli	12.5	0.8	11	0.5
7545005	Piner Point, Muary Island	soil	36.8	1.4	70	0.2
7545002		cabbage	3.1	0.6	9	0.2
7545006	Neill Point Vashom Island	soil	39.1	1.8	68	0.3
7545003		broccolli	10.9	3.35	19	0.6
7547001	5130 N. 48th St.	cabbage (inner head)	1.6	0.9	8	0.1
7547002		brussel sprouts	30.8	0.8	7	0.1
7547003	5129 N. 47th St.	saurkraut	0.6	1.1	5	0.06
7547004	5140 N. 47th St.	cabbage (inner head)	1.2	0.8	11	0.1

Table III

Heavy metal content of vegetation and soil samples from the vicinity of the Tacoma Smelter. (Concentrations expressed in parts-per-million on a dry weight basis)

Sample No	Location	Sample	Arsenic	Cadmium	Lead	Mercury
7542001	5129 N. 47th St.	lettuce (unwashed)	657		1200	6.0
7542002		lettuce (washed)	445		700	3.2
7542003		cabbage (unwashed)	94		50	0.4
7542004		cabbage (washed)	67		45	0.1
7542005		soil	384		1100	3.9
7544001	5110 N. 40th St.	soil	44.2	1.86	115	1.37
7544019		brussel sprouts	7.8	2.3	14	0.5
7544002	5618 N. 43rd St.	soil	331	11	972	6.1
7544020		swiss chard	27.6	5.6	19	0.4
7544003	2844 N. Bristol St.	soil	24.0	1.5	44	1.5
7544021		cabbage	5.2	8.2	20	0.5
7544004	643 Skyline Dr.	soil	26.4	1.4	12	0.7
7544022		brussel sprouts	4.6	5.3	25	0.3
7544005	4845 S. 7th St.	soil	16.5	1.5	97	0.2
7544023		cabbage	1.0	3.0	14	0.3
7544006	3106 N. Huson St.	soil	55.7	2.3	120	0.6
7544024		cauliflower	5.5	2.6	23	0.7
7544007	2136 N. Mildred St.	soil	93.3	2.5	102	0.5
7544025		cabbage	8.5	7.9	11	2.4
7544008	5011 N. 25th St.	soil	24.6	0.9	30	1.0
7544026		cabbage	8.7	5.3	20	0.6
7544009	4508 N. Visscher St	soil	355	12.0	1190	3.6
7544027		cabbage	14.5	2.4	22	0.8
7544010	8002 W. 31st St.	soil	29.7	1.7	52	0.3
7544028		cauliflower	5.2	2.4	21	0.5
7544011	5130 N. 48th St.	soil	110	1.8	88	0.9
7544029		brussel sprouts	16.3	1.2	11	0.4
7544012	5140 N. 47th St.	soil	326	5.4	505	4.8
7544030		cabbage	32.9	5.0	16	0.3
7544013	5423 N. 49th St.	soil	240	8.3	291	1.9
7544031		cabbage	79.2	6.6	9	0.7
7544040		cabbage (unwashed)	128	7.8	65	1.2
7544014	5447 N. 49th St.	soil	247	7.0	819	3.0
7544032		cabbage	3.45	1.5	14	0.3
7544015	5103 N. Winnifred St.	soil	307	7.8	1240	3.6
7544033		cauliflower	35.8	3.5	13	0.5

Fig. 1a

Arsenic concentration (ppm) in vegetation in the Tacoma area.

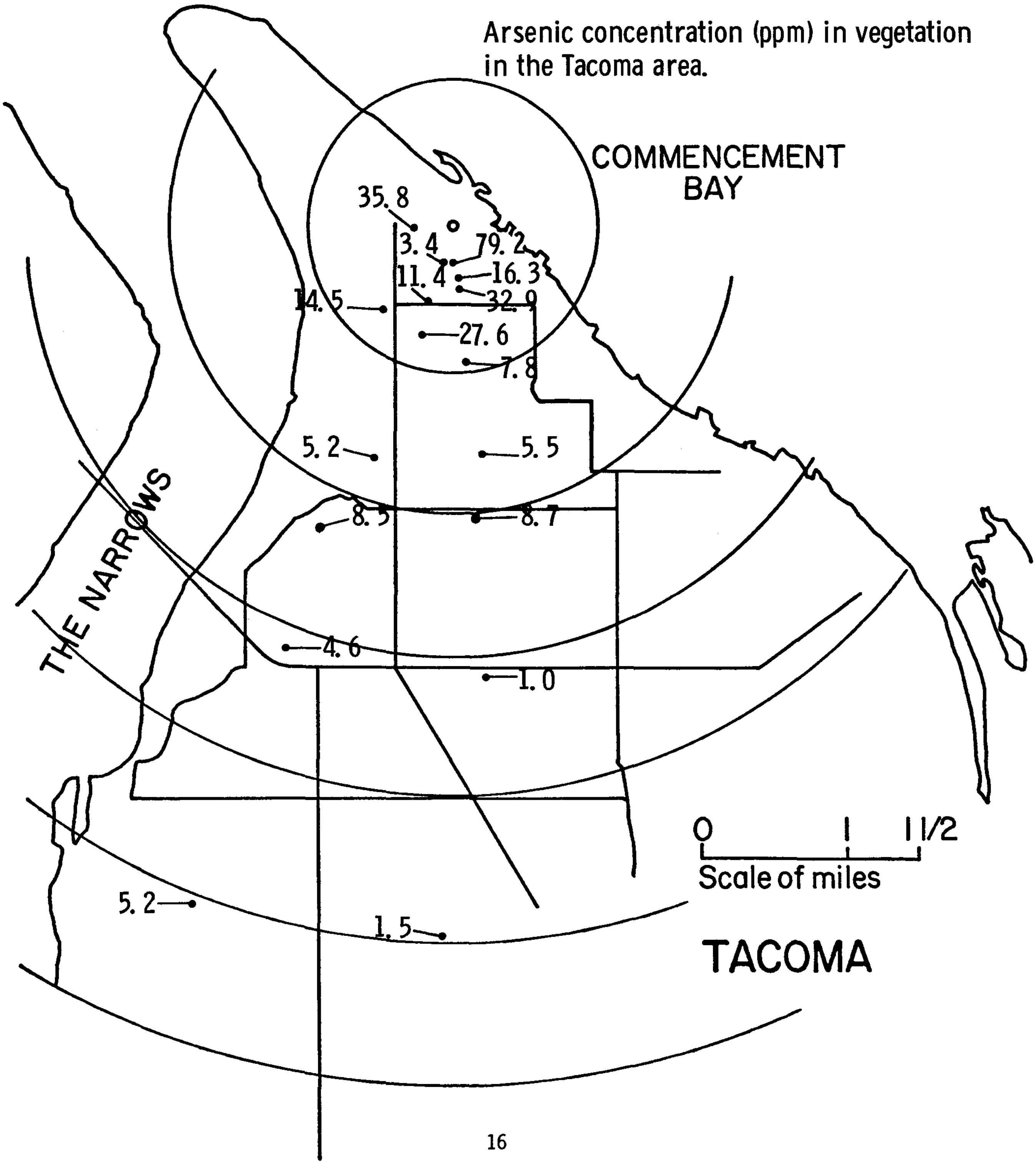


Fig. 1b

Arsenic concentration (ppm) in garden soils in the Tacoma area.

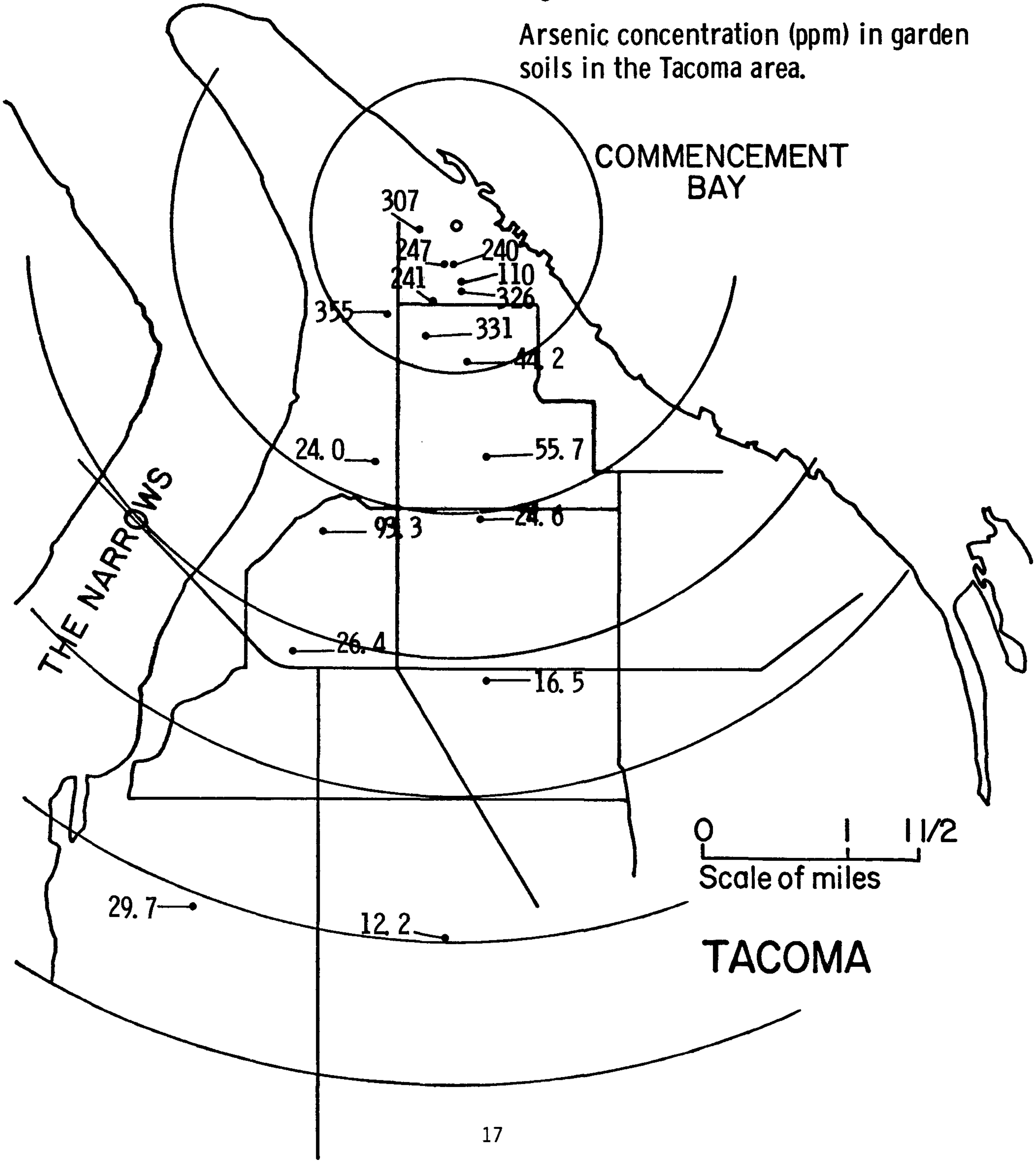


Fig. 2a

Mercury concentration (ppm) in vegetation in the Tacoma area.

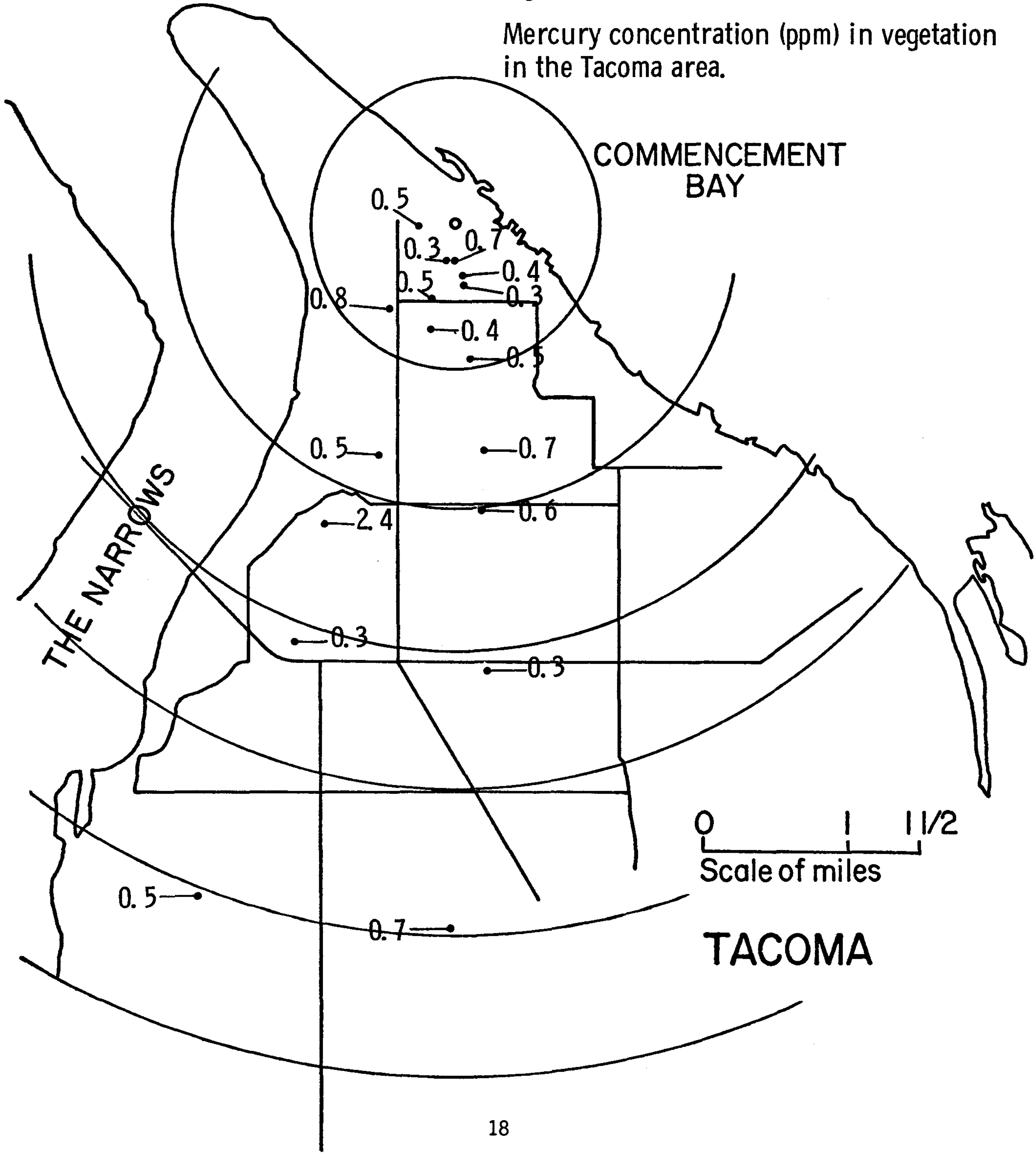


Fig. 2b

Mercury concentration (ppm) in garden soils in the Tacoma area.

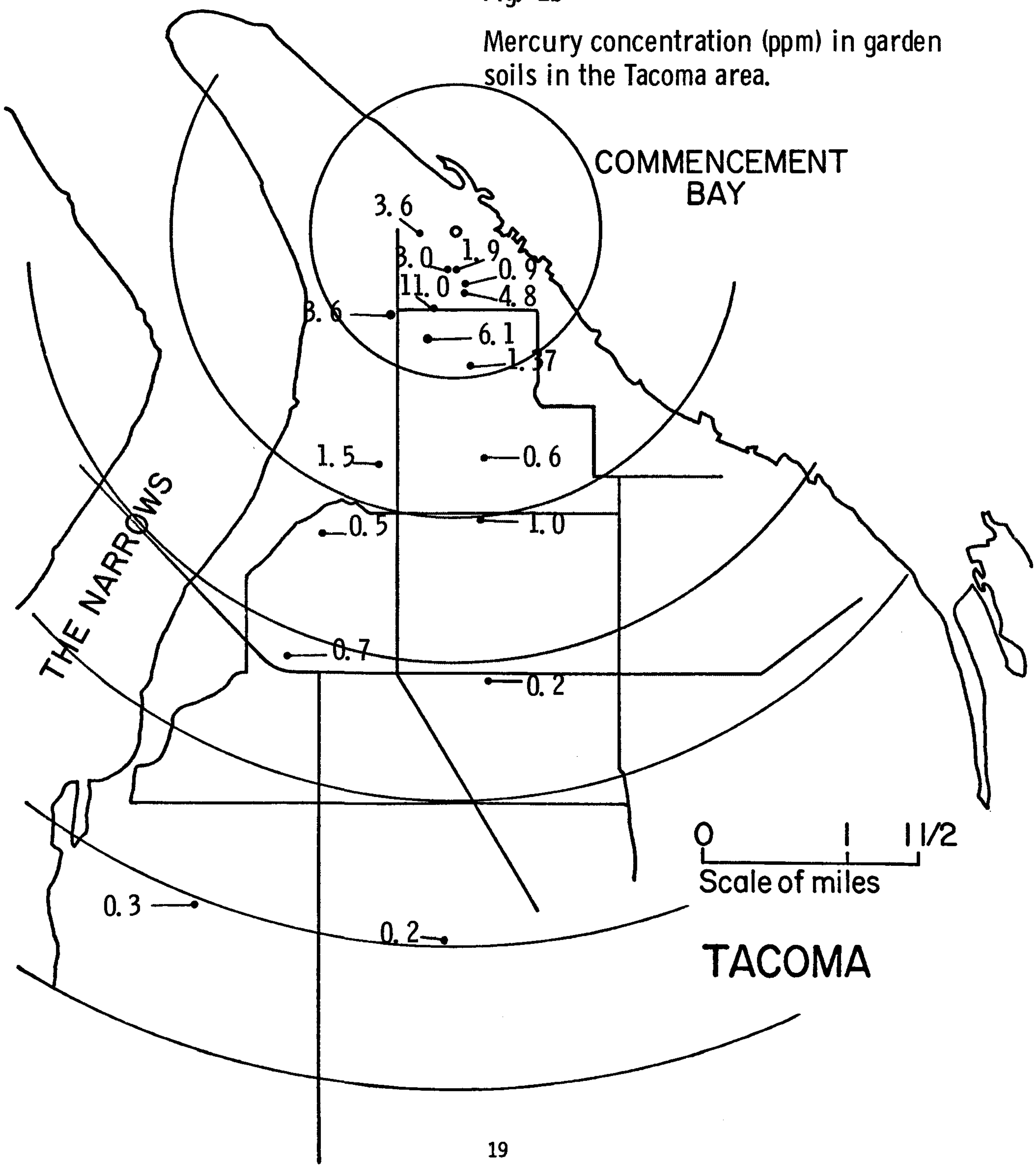


Fig. 3a

Lead concentration (ppm) in vegetation in the Tacoma area.

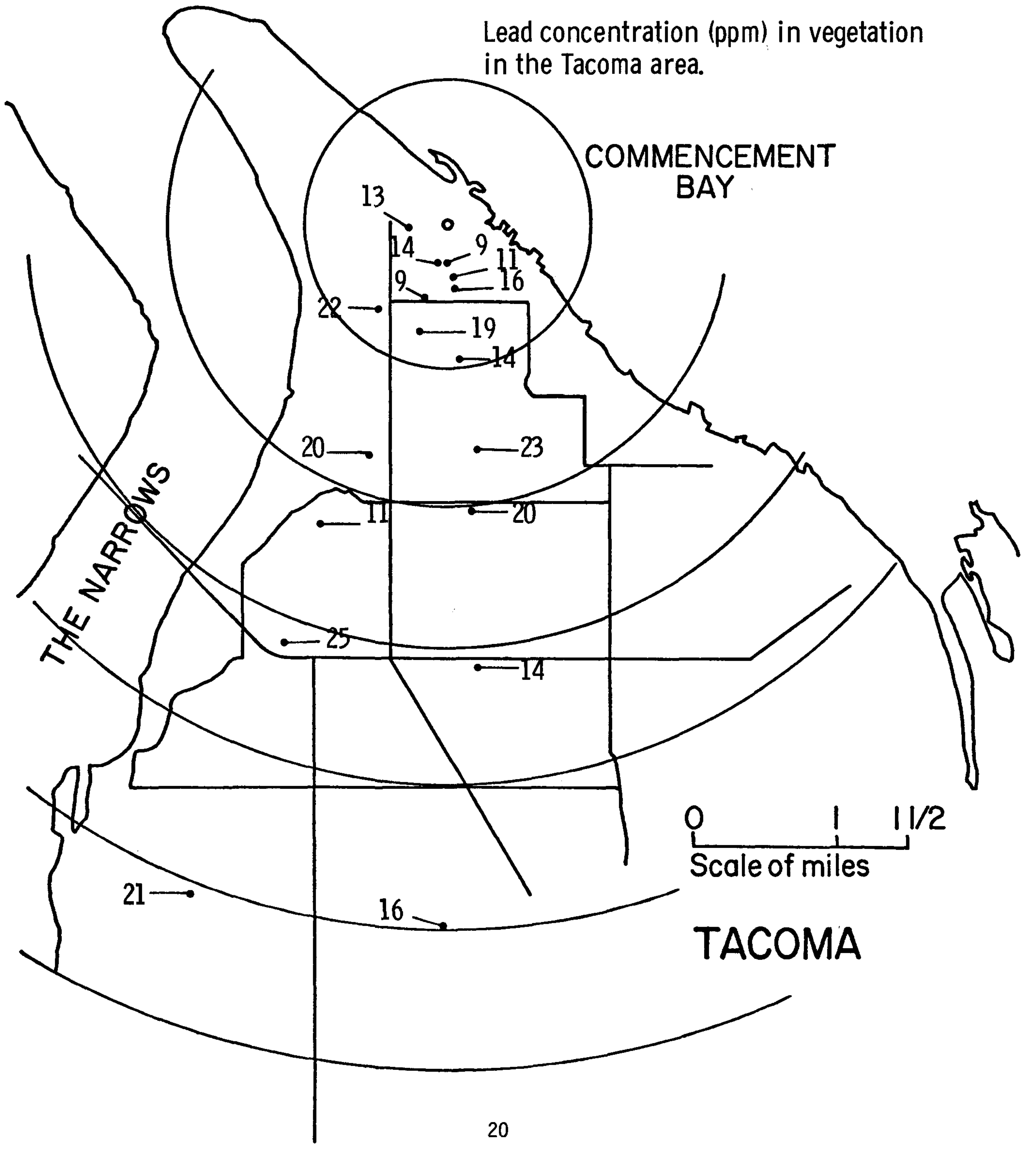


Fig. 3b

Lead concentration (ppm) in garden soils in the Tacoma area.

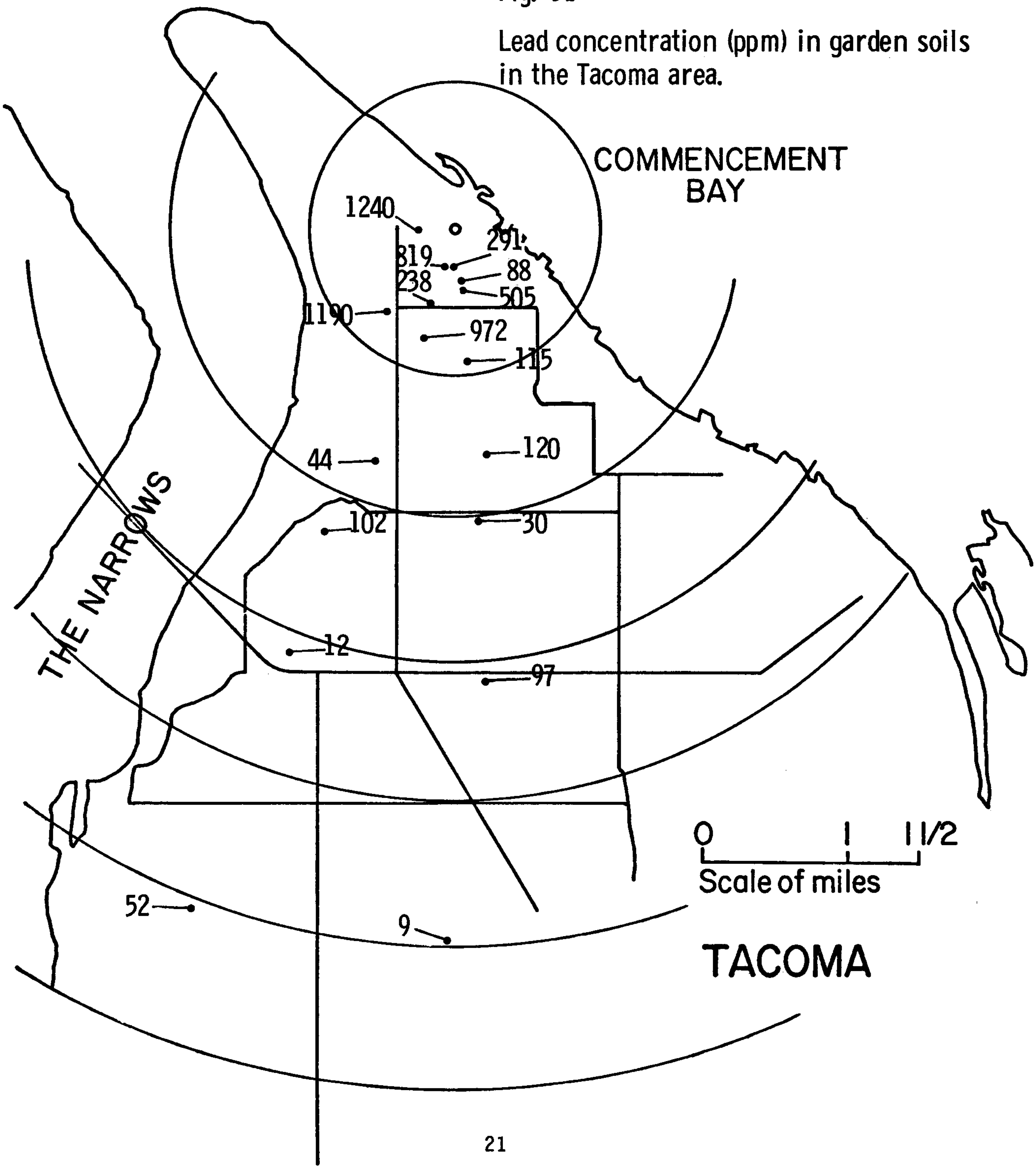


Fig. 4a

Cadmium concentration (ppm) in vegetation in the Tacoma area.

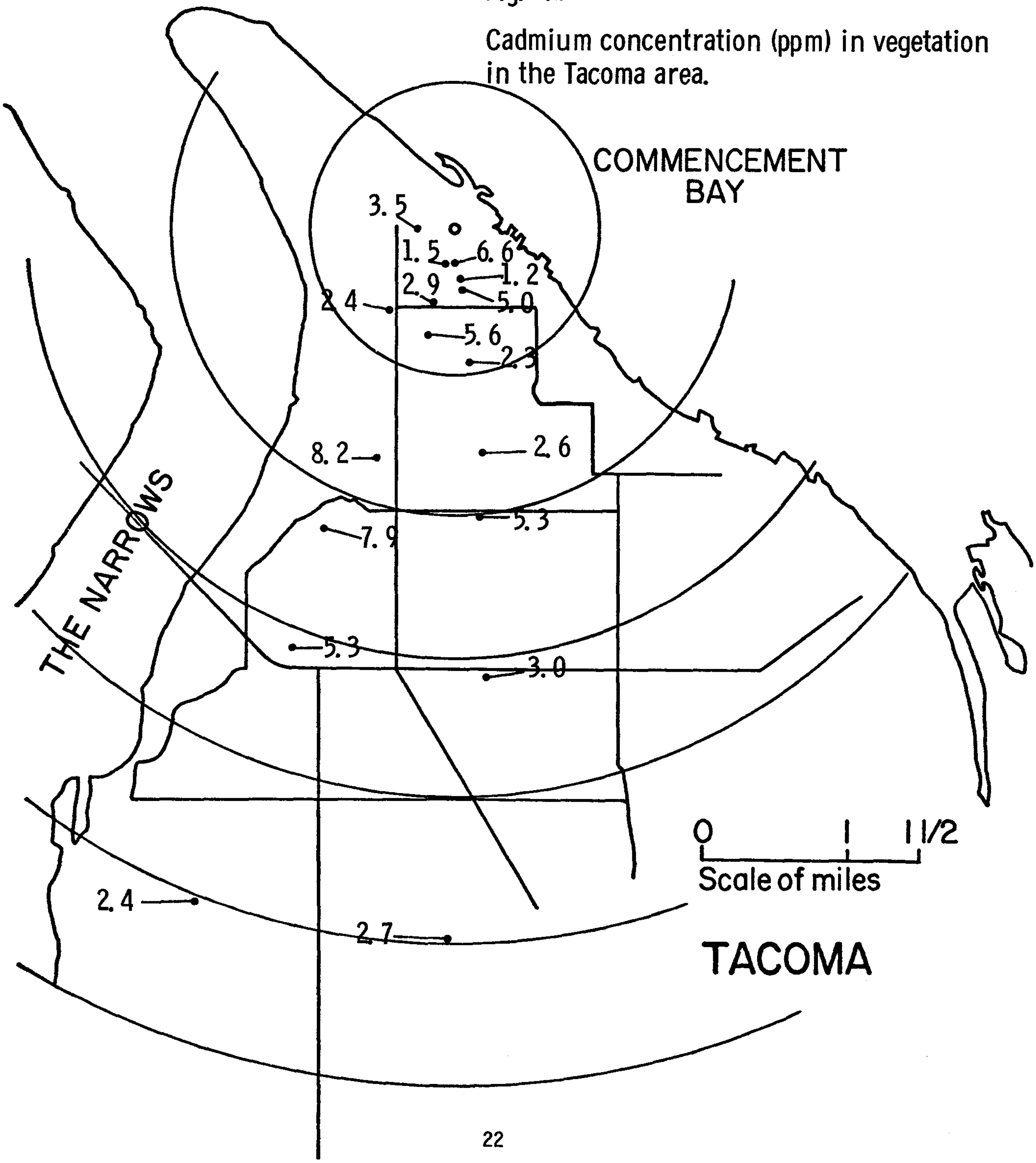
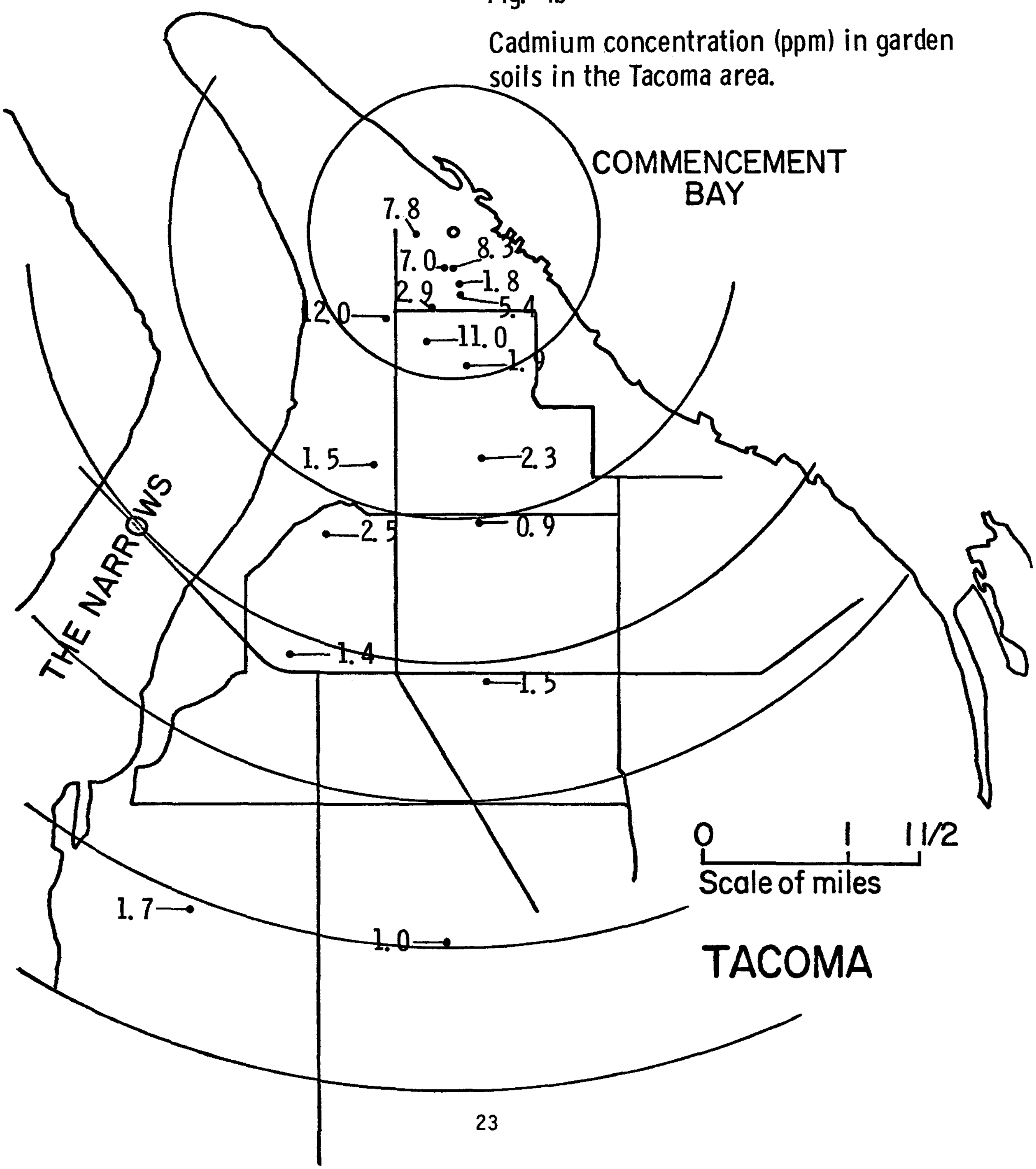


Fig. 4b

Cadmium concentration (ppm) in garden soils in the Tacoma area.



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15. Supplementary Notes

16. Abstracts

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17. Key Words and Document Analysis. 17a. Descriptors

Heavy-metal accumulation
Air pollution
Vegetation effects
soil toxicity
copper smelter emissions

17b. Identifiers/Open-Ended Terms

heavy-metal accumulation in soils
heavy-metal accumulation in vegetation
air pollution

17c. COSATI Field/Group

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