Introduction

This discussion of lead in the environment concerns natural ecosystems not under the direct influence of man and generally avoids the various subjects of human environments, agricultural systems, heavily trafficed areas, and areas directly subjected to industrial effluents of lead. The objective is to place atmospheric lead in perspective to natural ecosystems and ecological processes.

Lead is not a biologically essential element and has not been studied as extensively as some other metals, such as maganese and zinc. However, knowledge of lead in ecosystems is increasing because of concern over its release from human activities in natural environments. The greatest concern regards the release of lead into the atmosphere but this seems to pose little direct threat to natural biota. It is, however, the eventual deposition of lead by fallout or rainout into terrestrial and aquatic ecosystems that gives rise to concern over its role as an environmental pollutant.

Source of Atmospheric Lead

In the context of present-day concentrations of atmospheric lead, natural sources apparently contribute only insignificantly. Natural concentrations have been estimated to be about 0.0005 g lead/m^3 air (Patterson 1965) and result from airborne dust containing an average of 10-15 ppm lead (Chow and Patterson 1962) and gases diffusing from the earth's crust (Blanchard 1966). The latter source results in the presence of lead -210 in the atmosphere ranging from 70 x 10^{-3} dpm/kg air at ground level to about 70 x 10^{-3} dpm/kg air in the lower stratosphere (Burton and Stewart 1960). Detectable quantities of lead-210 reach the earth's surface in precipitation and on dust particles (Hill 1960, Lagerwerff 1967, Francis et al. 1970), and concentrations of 1-8 pCi/l rainwater have been reported.

Presently, the principal source of atmospheric lead is considered to result from the combustion of leaded automotive fuels (Patterson 1965). In the U.S. in 1968 alone, more than 500,000,000 pounds of lead were used as antiknock compounds, and since the advent of leaded gasolines, a cumulative amount of more than 12 billion pounds of lead have been consumed (Chow and Earl 1970a). About 75% of the lead in gasoline is introduced into the atmosphere as aerosols (Mueller et al. 1962).

By comparison with gasoline, the burning of coal has not been a serious source of atmospheric lead. In recent years, 500,000,000 tons of coal with an average lead content of 10 ppm (Chow and Earl 1970b) have been burned annually in the U.S. Contribution to atmospheric lead by coal would be only 3 percent of that derived from gasoline if lead from both were introduced into the atmosphere with equal efficiency. In modern furnaces, about 95-99% of the fly ash is removed by electrostatic precipitators and only a small portion of the lead in coal is emitted as aerosols (First 1968). Thus, the contribution by coal to atmospheric lead appears to be small.

In the past, significant amounts of lead were introduced into the atmosphere by smeltering lead ores (Murozumi et al. 1969). At the beginning of the Industrial Revolution in 1750, poor smeltering technology resulted in the loss of much lead fume. During the 19th century smeltering procedures were improved, and by the early 20th century it became economically feasible to recover lead smelter fume. This resulted in reducing the fraction of fume loss by a factor of 4, and during the past four decades the loss fraction has been further reduced by a factor of 8 (Table 1). Thus, the emission of lead aerosols into the atmosphere in the Northern Hemisphere by smeltering currently is small in comparison to the amount emitted from burning gasoline. In 1966, for example, the comparative values were 2,000 tons of lead aerosols resulting from smeltering and 100,000 tons from

leaded fuels (Murozumi et al. 1969).

A useful tool for identifying sources of lead pollution in the environment has been analysis of isotopic composition (Chow and Patterson 1962, Ault et al. 1970). Each lead ore has a characteristic composition fixed during mineral genesis, and a survey of the isotopic composition of commercially important ores has been made (Brown 1962). Additionally, Chow and Earl (1970a) have determined the isotopic composition of several lead ores, gasolines, aerosols, waters, and marine sediments (Fig. 1, in Chow and Earl 1970a). The average isotopic composition of lead in sediments from the Pacific and Atlantic Oceans are characteristic of lead from the Quaternary Period, deposited millennia ago. Contrary to this, lead in coastal surface seawater has a characteristic age of Tertiary time and is no longer the sole product of natural weathering. The isotopic composition of lead in gasoline additives and in atmospheric aerosols from cities where the fuel was obtained are similar and represent lead ore of Tertiary or older age. Chow and Earl (1970a) interpret this as implying that the aerosols could not be derived from weathering of natural surface material and instead come from automotive exhausts.

Lead in the Abiotic Environment

Although lead emissions into the atmosphere can be identified as to source and grossly quantified, it is considerably more difficult to obtain estimates of amounts actually transported to geographical regions or environmental compartments. Coarser fractions of lead settle along roadsides, within cities, or near smelteries, but in the context of natural environments it is the fine aerosols that are important, since these travel long distances and are washed out of the atmosphere in rain and snow. To gain an impression of the magnitude of this process it is instructive to assess data on the concentrations of lead in various components of the environment

as a function of time. Perhaps the most useful records in this respect are chronological layers of snow strata, available in quiescent ice sheets in perpetually frozen polar regions. Such an analysis of precipitated lead has been made in annual layers of snow from Greenland and the Antarctic continent (Murozumi et al. 1969).

Annual ice layers from the interior of northern Greenland show that lead concentrations increase from less than 0.0005 microgram/kg ice at 800 B.C. to more than 0.2 microgram/kg ice in 1965 (Fig. 2, from Murozumi et al. 1969, pg. 1285). The ice layer corresponding to 1750 A.D. represents the beginning of the Industrial Revolution and the lead concentration at that date is 25 times greater than natural levels. During the second half of the 18th century, lead concentrations tripled, and from 1935 to 1965, they rose abruplty again by a factor of three. The sharpest rise occurred after 1940, and today, lead concentrations in Greenland snows are over 400 times above natural levels.

These data dramatically document the rise of lead in certain components of the natural environment but must not be taken as representative of world-wide circumstances. For example, levels of lead in Antarctic continent ice sheets fail to show similar rises. Concentrations of lead in ice prior to 1940 were below 0.001 microgram/kg and rose to only 0.02 microgram/kg after 1940. The difference between lead concentrations in northern and southern polar snows is ascribed to barriers to north-south tropospheric mixing hindering the migration of aerosol pollutants from the northern hemisphere to the Antarctic.

Few chronological records of lead concentrations are as complete as the one described for polar regions, but some additional data do exist reflecting increasing lead in certain ecosystems. For example, it is estimated that the preindustrial lead content in marine water was about

0.02-0.04 mg/kg (Chow 1968). Today concentrations of surface waters in some areas of the Mediterranean and Pacific oceans contain as much as 0.20 and 0.35 Mg lead/kg water respectively and only deep waters, below 1,000 m, appear uncontaminated. (Fig. 3, from Chow 1968, pg. 408).

As with marine waters, the lead content of fresh waters also has increased in recent times. The available data suggest that the mean global natural lead content for lakes and rivers is from 1 to 10 micrograms/liter. (Livingston 1963). The lead content in public water supplies of the 100 largest cities in the U.S. in 1962 ranged from traces to 62 micrograms/liter (Durfor and Becker 1964), reflecting sizable additions of lead from pollution in some localities.

Rainwater in some regions apparently contains concentrations of lead greater than expected from natural sources. Lazrus, et al. (1970) reported an average of 36 micrograms lead/liter rain for a U.S. nationwide sampling network. They concluded that the lead content in rainwater at various cities is correlated with the gasoline consumption there. For example, in the Chicago area, rainfall results in an average monthly input of about 40g lead/ha compared to about 1g lead/ha in less populated regions.

In soils, a range of about 2-200 ppm lead usually occurs naturally, exclusive of areas near deposites of lead ore (Motto et al. 1970, Huff 1952, Wright et al. 1955). Natural concentration of lead in soil is primarily a function of the geological source of the parent material (Hamilton 1967), and as a rule, lead content decreases with soil depth (Table 2) (Wright 1955, Schuck and Locke 1970). Motto et al. (1970) believe that the natural content of lead in typical soils is low, usually less than 50 ppm with an average of about 15 ppm, but soils near heavily trafficed areas commonly are heavily contaminated. For example, they found 160 ppm lead in the upper 6 inches of soil within 25 ft. of a major highway. Beyond 75 ft.,

(Table 3).
however, the concentration dropped to about 80-90 ppm. Similar results have been reported by others (Schuck and Locke 1970, Chow 1970), and in general, the comparative content of lead in surface soils is a function of the volume of regional automotive traffic, proximity to roadways, and other factors such as prevailing wind direction. In remote areas, little or no increase in lead in soils is occurring (Chow, in press), suggesting that atmospheric lead aerosols deposited in precipitation are not of sufficient amounts to contaminate the soil environment. The problem is, however, severe near major roadways (Table 4).

CONCENTRATIONS OF LEAD IN BIOTA

Most information relating to lead in organisms concerns those of economic importance to humans, and far less is known of concentrations in natural biota. Fortunately, however, Swedish scientists are giving attention to this problem, and one of their studies (Ruhling & Tyler 1968) is especially valuable regarding vegetation.

In Sweden, the concentrations of lead in several mosses, herbs, shurbs, and trees were studied in relation to distances from highways. Lead contents of plants and soils within 50-100 miles of roads was high but dropped sharply and became stabilized at greater distances (Figs. 4, 5, 6, in Ruhling and Tyler 1968, pg. 324,325,327). An exception to this rule occurred with mosses which generally had substantial concentrations of lead above normal levels at distances far from roads. Additionally, mosses near highways typically had much greater contents of lead than did vascular plants.

Mosses accumulate most of their nutrients from precipitation and dust (Svensson and Linden 1965, Tamm 1953) making them suitable indicators of such air pollutants as lead (Ruhling and Tyler 1968, 1969). Pursuing this, Ruhling and Tyler (1968) demonstrated chronological increases in lead concentrations in Swedish mosses from 1860 to 1968 (Fig. 7, in Ruhling and Tyler 1968, pg. 341) corresponding to increased use of coal, 1875-1900, and 1950-1968. Mosses also were found to reflect regional gradients in atmospheric lead content (Figs. 8, 9, 10, in Ruhling and Tyler 1968, pg. 335, 336, 337).

Unfortunately, data on lead in native fauna comparable to that for the flora in Sweden are not yet available, and virtually nothing is known of the contribution of atmospheric lead to the lead burden of animals in natural ecosystems. Swedish scientists have begun studies concerning lead in natural ecosystems (Westermark 1969) but few data are yet available: only scattered

reports exist, and note of this is made by Danielson (1970). Data reflecting historical increases in lead content in humans (Jaworowski 1968, Becker et al. 1968) presumably can be considered indicative of possible increases in native fauna, but no substantive information exists.

CYCLING OF LEAD IN THE BIOSPHERE

To place atmospheric lead in proper perspective regarding natural ecosystems it is essential to understand its processes of biogeochemical cycling. Because of the lack of significance attached to lead in natural systems in past years, no comprehensive treatment of its movements within living systems has been made. There is not sufficient information on the chemical forms, amounts, and rates of transfer of lead from one component of the environment to another to permit treatment of the subject in terms of systems analysis. However, fragmentary data do exist allowing some insight into the movement of lead atoms in the biosphere, and these come primarily from studies of lead-210 in the environment.

The importance of radionuclides in tracing the pathways and determining flux rates of lead was stressed by Burton and Steward (1960). Analyses of lead-210 in rainwater has allowed calculation of its residence time in the atmosphere to be from 7-30 days, depending on environmental conditions (Ter Haar et al. 1967, Francis et al. 1970). Whether these values apply to atmospheric lead aerosols resulting from gasolines is not known, but Ter Haar et al. (1967) found that stable lead and lead-210 were well mixed in air. Should the residence time of atmospheric lead aerosols be from 1-4 weeks, the turn-over rate of these materials may be such that a world-wide steady state of lead already has been reached in the atmosphere. It should at least be possible to estimate the turn-over time and volume of lead removed from the atmosphere by natural processes.

Another interesting observation by Ter Haar et al. (1967) is that the

contribution of stable and radioactive atmospheric lead to soil is about 0.2%/year of the existing natural burdens of soil lead. This amounts to about 0.04 ppm/year and might explain the failure of most investigators to detect a significant accumulation of lead in soils removed from heavily trafficed and industrial areas.

Studies of lead-210 also have provided insight into the movemet of lead into plants. For example, Wilson & Cline (1966) concluded that soil lead largely is unavailable for uptake by plants and amounts to only 0.003-0.005% of the amount in soils. Several investigators have concluded that the primary source of lead entry into plants is from rainfall and not soil (Francis et al. 1968, Hill 1960, Mayneord et al. 1960). Contrary to this, however, Tso et al. (1966) believe that a significant amount of soil lead is available to plants, but the exclusion of soil lead by most plants (Menzel 1965) appears to rule this out.

The movement of lead into animals is a little known subject, and the relative contributions of atmospheric lead and dietary lead are poorly understood. Smith et al. (1970) found that some mammals breathing heavily contaminated air absorbed lead and concentrated it in bone tissue. Reindeer apparently assimilate lead-210 from their food and concentrate it almost solely in bone (Miettinen 1967). Accumulation of lead in bones of vertebrates may be a general rule, since this also was observed in fish, with lead-210 in bone exceeding that in muscle by a factor of over 50 (Holtzman 1967).

As in vertebrates, marine shellfish concentrate considerably more lead in hard parts than in flesh (Holtzman 1967). Accumulation in soft tissues increases with increasing concentrations of lead in water, but the process of accumulation largely is reversible whenever the source of contamination is removed (Pringle et al. 1968).

EFFECTS OF LEAD ON NATURAL BIOTA

Except in circumstances of extreme contamination, lead appears at present not to be having adverse effects on native plants or animals. There is no evidence indicating that natural concentrations of lead are toxic to vegetation, and no mention is made of damage to plants in Swedish investigations or other studies of vegetation in relation to automobile traffic. Experimental data, however, indicate that the possibility exists of lead contamination someday reaching proportions deleterious to plants (Keaton 1937, Hammett 1928, Gregory and Bradshaw 1965, Brewer 1966). For example, Koeppe & Miller (1970) recently have shown that lead chloride can be deleterious to plant respiration and that the safety margin can be narrow under certain environmental conditions.

A narrow margin of safety also exists for some estuarine mollusks (Pringle et al. 1968): onsiderable toxicity was exhibited toward animals exposed to lead at 0.1-0.2 ppm in water. Birds appear vulnerable to lead poisoning (Begley and Locke 1967) and death apparently can result with as little as 0.5-3.7 ppm lead in the liver. Most hazards posed by lead to wildlife arise from lead shot or other metallic lead substances and not from atmospheric forms.

SUMMARY & CONCLUSIONS

1. Aerosols constitute the principal class of atmospheric lead compounds reaching natural environments, and their primary source is leaded automotive fuels.

2. The primary mode of input of lead aerosols to ecosystems is precipitation.

3. Historical samples of both biotic and abiotic materials document increases of lead in certain components of the environment corresponding to the advent of the Industrial Revolution and leaded gasoline.

4. Increases in lead content are not detectable in soils removed from heavily trafficed or industrial areas.

5. Lead concentrations are increasing in marine waters and freshwaters.

6. Lead concentrations are increasing in some plants, especially mosses which may serve as indicators of atmospheric lead pollution .

7. Too few data are available to evaluate possible lead increases in most groups of animals, terrestrial or aquatic.

8. Little is known of the assimilation of lead by animals, but it appears to accumulate to some extent in the bones of vertebrates.

9. Too little attention has been given to the ecological pathways of lead and its rates of transfer from one environmental compartment to another.

10. Virtually nothing is known about the effects of chronic exposures of native flora and fauna to increasing concentrations of lead in the environment.

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Table 1. Lead aerosol production in the Northern Hemisphere. Adapted from Muzorumi et al. (1969), pg. 1289.

Date	10^5 of Pb smeltered/yr	% converted to aerosols	10^3 tons of Pb aerosols/yr from smelteries	10^5 tons of Pb burned as alkyls/yr	% converted to aerosols	10^3 tons of Pb aerosols/yr from alkyls
1753	1	2	2	-	-	-
1815	2	2	4	-	-	-
1933	16	0.5	8	0.1	40	4
1966	31	0.06	2	3	40	100

TABLE 2

Lead Content in P.P.M. (Parts per Million) of Three Different Soil Profiles as a Function of Depth

Horizon (approximate)	Depth (")	P.P.M.	Depth (")	P.P.M.	Depth (")	P.P.M.
O	0	80[1]	0	28	0	-
A	3	73	1	15	2	74
	8	52	4	9	5	67
B	14	41	8	18	7	60
	18	31	13	11	12	72
C	21	<1	-	-	16	68
	26	<1	26+	12	20+	37

[1] First profile values from Schuck and Locke (1970), other values from Wright et al. (1955)

Table 3. Lead in soil along highways in northeastern New Jersey (Motto et al. 1970).

Distance from highway (ft.)	Traffic Volume (Vehicles/24 hr. in thousands) 12.8	14.7	16.9	17.3	17.7	19.7	19.8	35.2	41.0	45.6	48.6	48.6	54.7	Av.
	0–6 inches p.p.m. Pb													
25	134	48	48	154	442	125	77	192	165	47	206	266	169	159.5
75	70	39	87	42	43	95	59	76	130	30	157	117	171	85.8
125	60	39	78	42	43	129	70	70	66	108	109	104	98	78.2
175	63	30	64	49	34	332	80	48	129	84	91	51	78	87.2
225	58	23	77	43	40	193	64	49	228	162	74	114	78	92.5
Av.	77.0	35.8	70.8	66.0	120.4	174.8	70.0	87.0	143.6	86.2	127.4	130.4	118.8	100.6
	6–12 inches p.p.m. Pb													
25	82	26	39	29	616	72	62	42	48	58	178	65	48	105.0
75	43	34	64	34	20	53	17	34	70	23	86	62	76	47.4
125	36	26	68	28	24	76	86	18	48	18	38	20	26	39.4
175	47	26	39	32	24	28	76	28	61	21	30	32	20	35.8
225	52	27	44	34	32	247	63	26	73	229	33	31	34	71.2
Av.	52.0	27.8	50.8	31.4	143.2	95.2	60.8	29.6	60.0	69.8	73.0	42.0	40.8	59.7

TABLE , LEAD CONTENT IN TOP SOILS FROM VARIOUS LOCATIONS (Chow, in press),

LOCATION			LEAD P.P.M.
Antarctica			
South Victoria Land	McKelvey Valley		6 - 8
	Victoria Valley		5 - 11
	King Valley		9
	King-David Valley		5
	Matterhorn Valley		12
Americas			
Mexico	Mexico City	Guadelupe Garden	179
Peru	Lima	Plaza Grau	223
		Cajamarquilla	72
Chile	Santiago	Natural History Museum	90
		Normal Park	53
Texas	Houston	Old Market Square	720
		Sam Houston Park	99
Florida	West Palm Beach	Currie Commons Park	75
South Carolina	Chester	Brockman High School	600
Washington, D.C.		Ellipse Park	243
Massachusetts	Boston	Public Garden	345
New York	New York	Central Park	834
		Fort Tryon Park	185
		Cloister	293
		Botanical Garden	224

New Hampshire	Meriden	Kimball Union Academy	18
California		White Mountain	8
		Laguna Mountain	6
	Borrego Springs	Palm Canyon	7
	Brawley	U.S. Dept. of Agriculture	20
	San Diego	Balboa Park	194
		Horton Plaza	164
		Newtown Park	267
	Los Angeles	MacArthur Park	422
		MacArthur Park	3,357
		Hancock Park	306
	San Francisco	Golden Gate Park	560
Hawaii	Hawaii Island	Volcano National Park	4
	Hilo	Airport Park	214
	Honolulu	Iolani Palace	224
		Foster Botanical Garden	415
		Irwin Park	1,088
Asia			
Japan	Tokyo	Hibiya Park	167
		Shiba Park	169
		Ueno Park	195
		Meteorological Research Inst	67
China	Taipei	Wan Shou Park	67
		National Palace Museum	80
Hongkong	Kowloon	Post Office	78
	Victoria	Statue Square	107

Thailand	Bangkok	Sanamluang Park	331
		Patumwam Circle	1,175
		Chulalongkorn University	143
Malaysia	Penang	Museum Park	182
		Fort Cornwallis	107
Europe			
Austria	Vienna	Prater	85
Belgium	Brussels	Gare Midi	859
Denmark	Copenhagen	City Hall	105
France	Paris	Jardin des Tuileries	220
Germany	Kiel	Forest	11
	Munchen	Englischer Garten	158
	Hannover	Masch Park	757
	Hamburg	Botanical Garden	411
Holland	Amsterdam	Museum Park	893
U.S.S.R.	Moscow	Lomonosov University	19

Fig. 1. Isotopic correlation of lead in various specimens (Chow 1970).

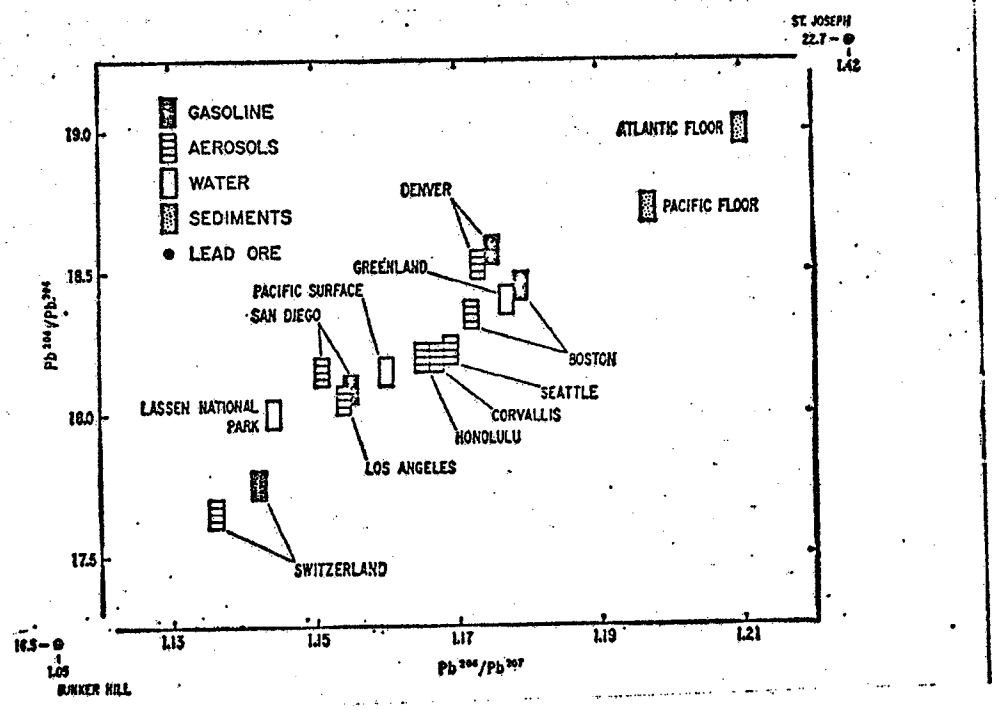

Fig. 2. Increase of lead in snow at Camp Century, Greenland since 800 B. C. Taken from Murozumi et al. (1969).

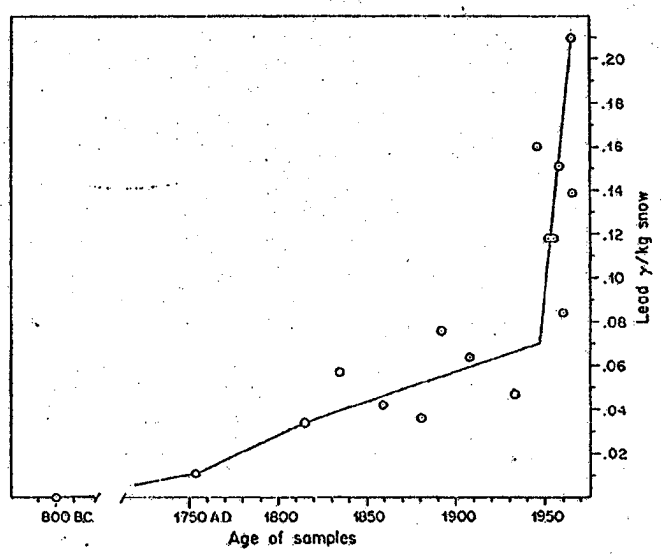

Fig. 3. Lead profiles in the major oceans (Chow 1968).

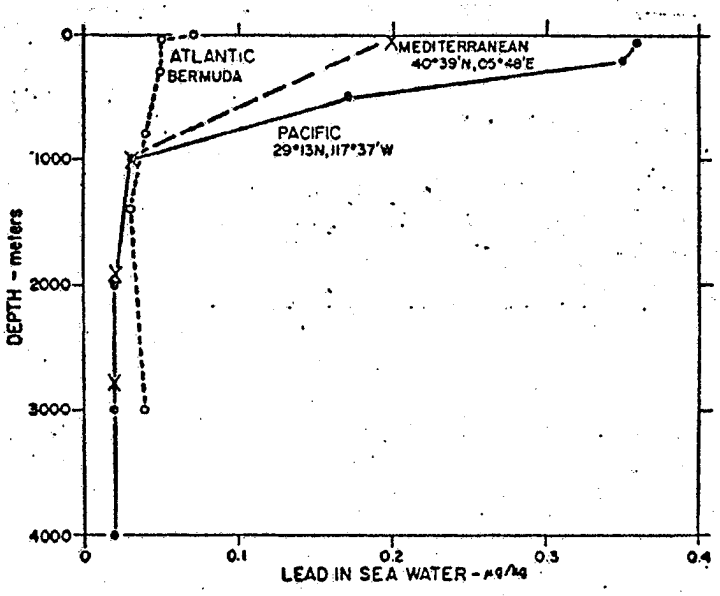

Fig. 4. Lead gradients in vegetation in a road transect at Stavsjo, Sweden. Plants sampled were Pleurozium schreberti, Lusula pilosa, Vaccinium vitis-idaea, and Picea abies. Taken from Ruhling and Tyler (1968).

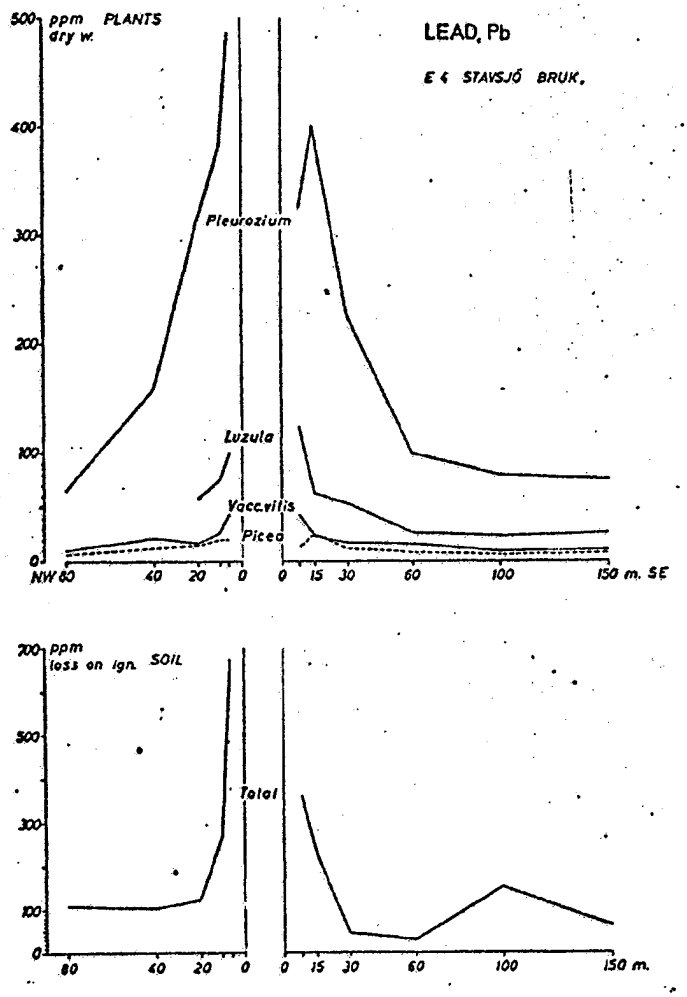

Fig. 5. Lead gradients in vegetation in a road transect at Oskarshamn, Sweden. Plants sampled were Luzula pilosa, Deschamosia flexuosa and Vaccinium vitis-idaea. Taken from Ruhling and Tyler (1968).

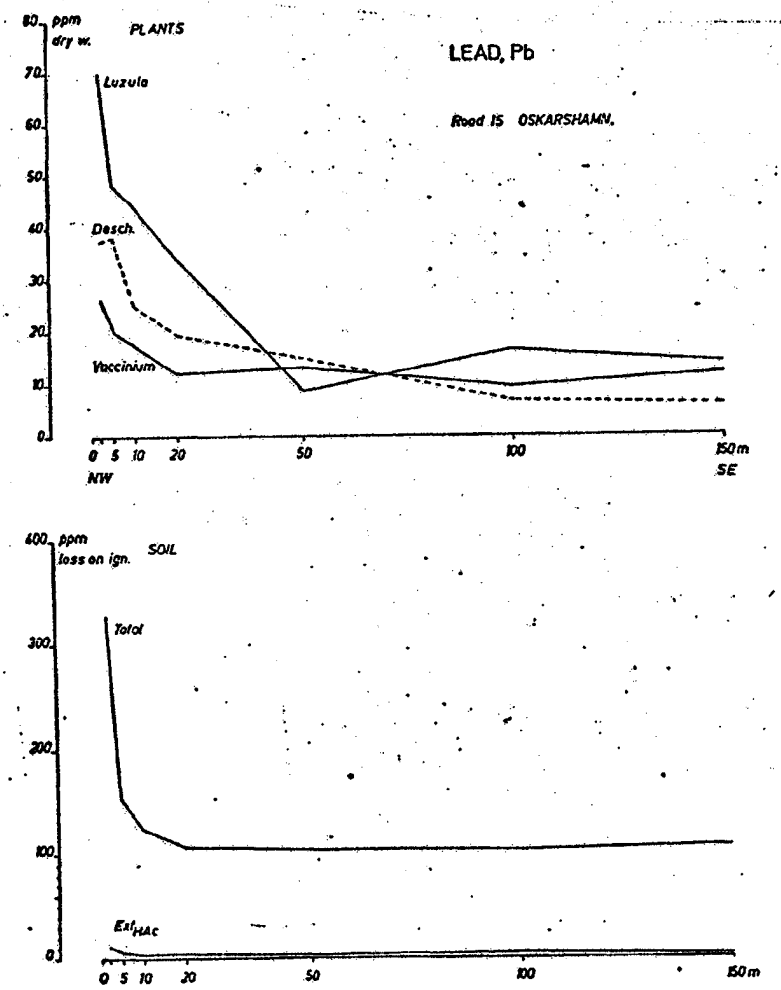

Fig. 6. Lead gradients in vegetation in a road transect at Uppakra, Sweden. Plants sampled were Brachythecium sp., Alopecurus pratensis and Anthriscus silvestris. Taken from Ruhling and Tyler (1968).

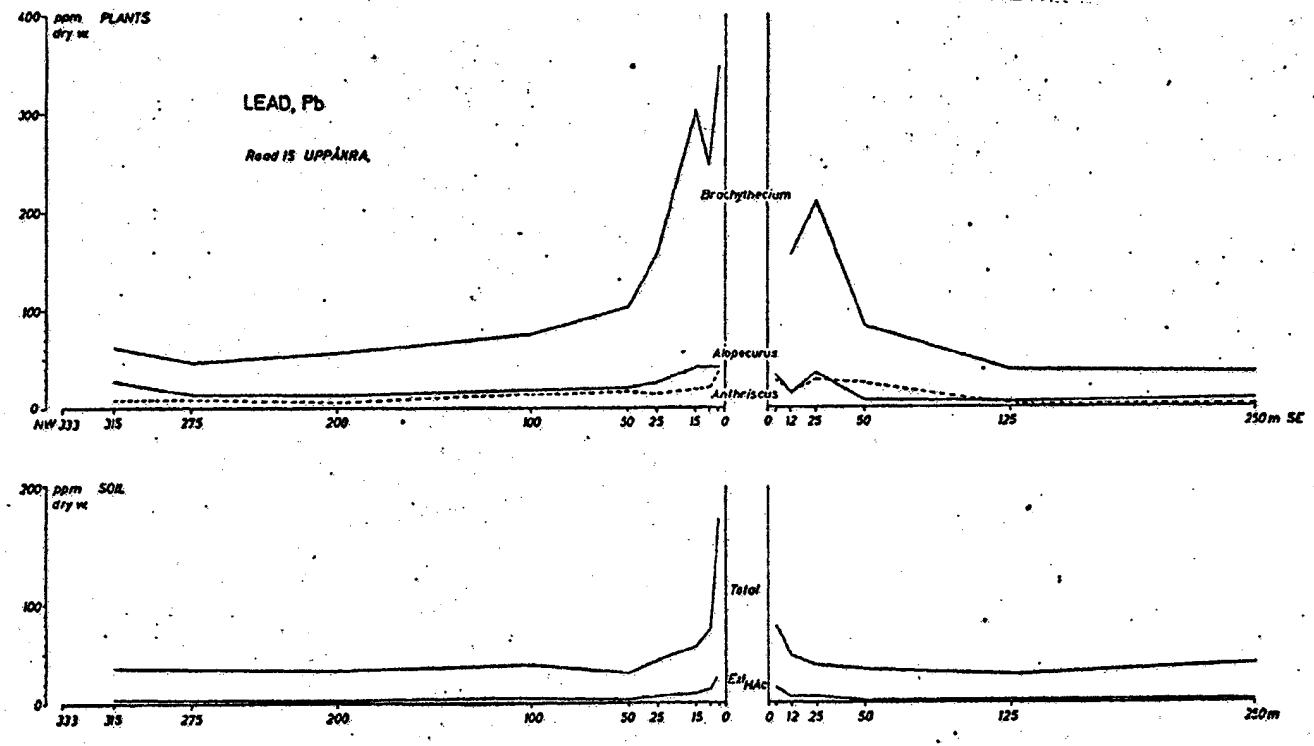

Fig. 7. Lead concentrations in samples of Hylocomium splendens, Pleurozium schreberi and Hypnum cupressiforme collected in Skane, Sweden from 1860 to 1968.

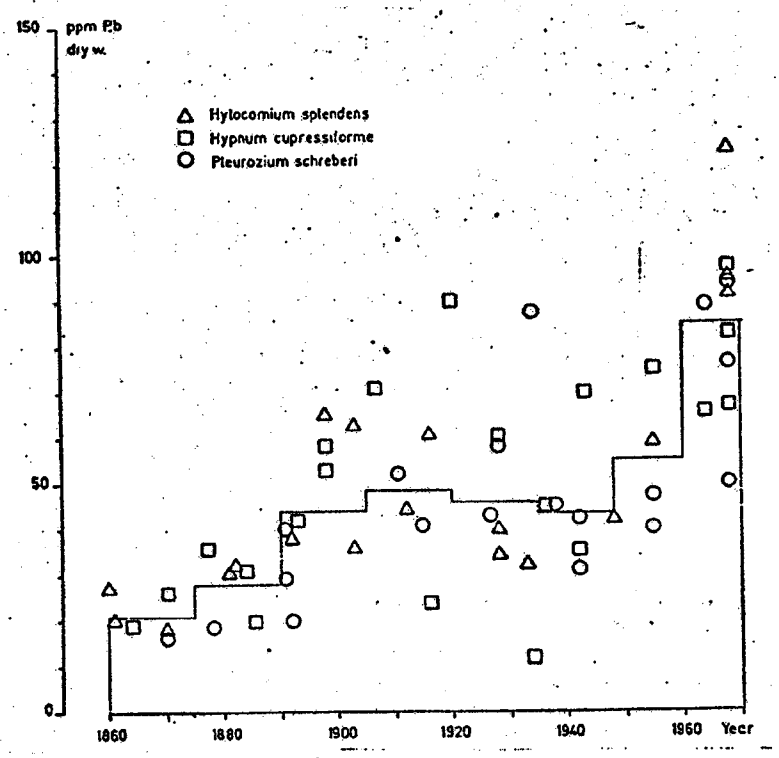

Fig. 8. Lead concentrations in Hylocomium splendens in Sweden. Taken from Ruhling and Tyler (1968).

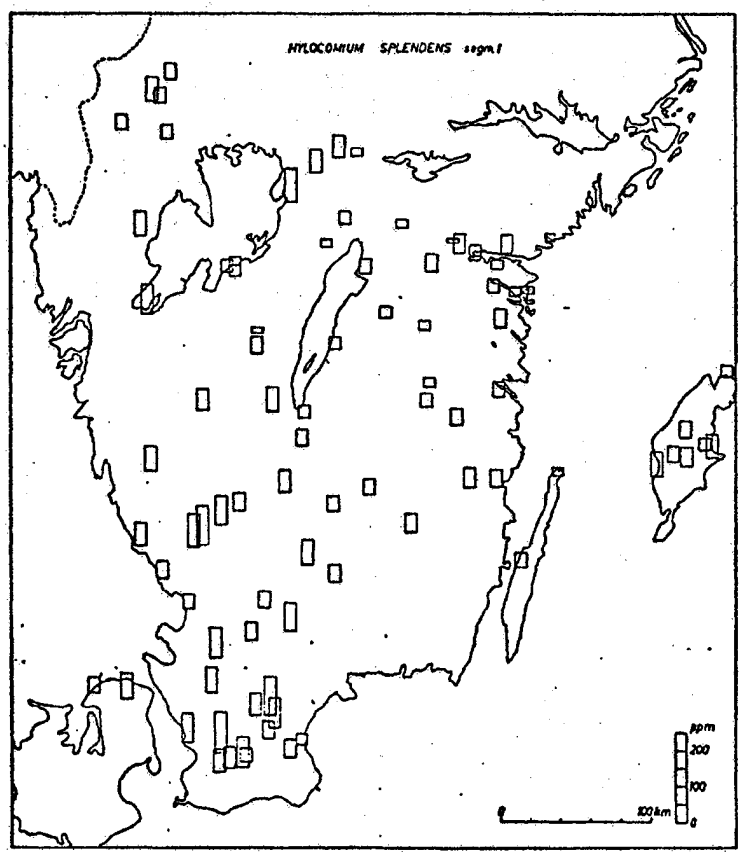

Fig. 9. Lead concentrations in Pleurozium schreberi in Sweden. Taken from Ruhling and Tyler (1968).

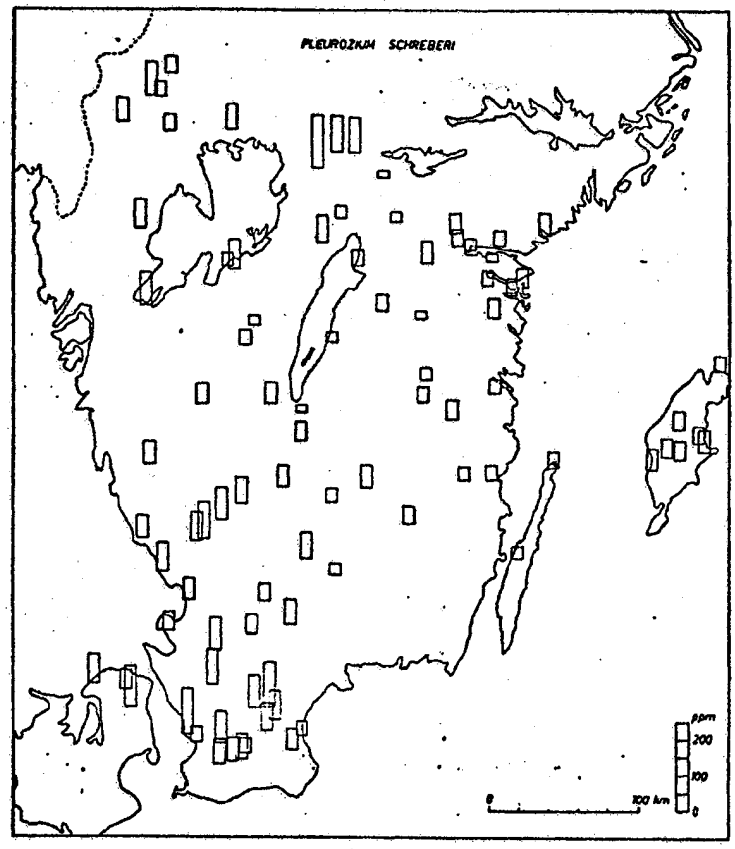

Fig. 10. Lead concentrations in Hypnum cupressiforme in Sweden. Taken from Ruhling and Tyler (1968).