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Retrospective Study of Lichen Lead Accumulation in the Northeastern United States

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Abstract. *Lichen samples collected from several locations in the northeastern United States from 1907 to 1980 were analyzed for lead content to establish accumulation trends indicative of atmospheric Pb pollution patterns. The main collection site was Plummers Island, Maryland, a Potomac River island subjected to severe atmospheric pollution stress from automobile exhaust. A nearby island, Bear Island, more remote from traffic, was also studied for comparison. In addition, lichen samples were collected in Shenandoah National Park in western Virginia, and Aton Forest and other sites in Litchfield County, Connecticut. Present-day Pb concentrations in all species sampled from Plummers Island were found to be significantly higher than concentrations in the same species from Bear Island, highest values being obtained for* Pseudoparmelia baltimorensis *and* Xanthoparmelia conspersa. *These values are the highest yet found for foliose and fruticose lichen species subjected to atmospheric automobile exhaust pollution, although higher values have been reported for lichens from Pb mining areas. Significantly increased Pb concentration values were observed for lichens collected from Plummers Island between 1907 and 1958. During this interval, Plummers Island was remote from any specific Pb pollution source; therefore, the average "background" Pb pollution level appears to have also increased during this time.* Cladina subtenuis *was found to contain the lowest Pb concentrations in all sites, suggesting that lichen morphology influences particle trapping capacity and thus regulates the rate of Pb absorption by lichen tissues.*

Increased concern over the effects of lead pollution that are caused by the use of leaded gasoline in automobiles has stimulated a number of studies of Pb accumulation by plants, particularly cryptogams, in roadside environments (Rühling & Tyler 1968, 1971; Goodman

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& Roberts 1971; Smith 1976; Burkitt et al. 1972; Rao et al. 1977). These studies have demonstrated that atmospheric Pb burden is concentrated near heavily traveled roads and that significant pollution effects are not seen at distances greater than 100 m from roads. These results are consistent with the model of Pb cycling proposed by Smith (1976) for roadside systems.

Despite the highly concentrated deposition pattern observed for atmospheric lead, however, cryptogam samples collected in "unpolluted" areas often exhibit elevated Pb concentrations (Rühling & Tyler 1968). Lead deposition rates have been found to be increasing not only in urban areas (Chow & Earl 1970), but also in areas far removed from pollution sources in the U.S.A. (Schlesinger et al. 1974). These patterns demonstrate a need to document changes in Pb burden with time.

A number of retrospective heavy metal studies have been done with cryptogams since Rühling and Tyler (1968) first published on the elevated lead content of a number of species during an interval from 1860 to 1968 in Sweden. They found two distinct periods of increased Pb concentration. One occurred during the end of the nineteenth century; the other occurred after the second World War. Although they were unable to explain the first major increase in Pb concentration, they attributed the second increase to the use of leaded automobile fuel after 1940. Rasmussen (1977) and Johnsen and Rasmussen (1977) determined the concentration of several heavy metals, including Pb, for epiphytic bryophytes from rural areas in Denmark. Even in these rural areas, Pb concentration was found to have increased approximately one order of magnitude during the last quarter century. Rao et al. (1977) reported results of an unpublished study by Robitaille and LeBlanc on heavy metal accumulation by five moss species from Mount Royal, Montreal, from 1905 to 1971. Lead was found to have exhibited the greatest increases in *Heterophyllium haldanianum* (Grev.) Kindb. from 20 μg/g in 1905 to 110 μg/g in 1971. These results indicated a steadily increasing heavy metal pollution in the area through urbanization and industrialization.

There have been relatively few retrospective studies of lichen heavy metal accumulation over time, although the few that have been published demonstrate that lichens are as effective as bryophytes as long-term integrators of atmospheric pollution burden. In central Sweden, Persson et al. (1974) studied the concentration of radiolead (Pb-210) and stable Pb in field-collected *Cladonia alpestris* (L.) Rabenh. (= *Cladina stellaris* (Opiz) Brodo) and unidentified *Cladonia* species from herbarium collections dating to 1882. They found a decreased ratio of Pb-210 to stable Pb after 1940, indicating an increased deposition of stable Pb at that time. In an examination of herbarium specimens dating to 1893 and field-collected material, Schutte (1977) found elevated Cr concentrations in the most recently collected thalli of *Parmelia caperata* (L.) Ach. (= *Pseudoparmelia caperata* (L.) Hale) and *P. rudecta* Ach. from several locations in Ohio and West Virginia. She attributed the increased Cr concentrations to the effects of air pollution in the region.

Retrospective studies of lead concentrations in lichens are limited only by the period over which samples have been collected in a given area of interest; they should provide a long-term pollution monitoring tool. We report herein Pb concentration values for three common species collected in various locations and times. The retrospective concentration values obtained for these lichens indicate a significant rise in background Pb levels as well as greatly increased levels near heavily travelled roads.

METHODS

Samples of the saxicolous lichens *Pseudoparmelia baltimorensis* (Gyel. & For.) Hale and *Xanthoparmelia conspersa* (Ach.) Hale and the terricolous *Cladina subtenuis* (Abb.) Hale & Culb. were collected from several locations. The main collection site was Plummers Island, Maryland, an island in the Potomac River. Plummers Island is located below the Cabin John Bridge, which supports the Capital Beltway (Interstate 495) highway between Maryland and Virginia. Since the bridge was built in 1965, automobile traffic has increased from a mean of approximately 40,000 vehicles per day to approximately 100,000 vehicles per day at the present time.

In order to compare the lead pollution burden resulting from fallout from the Cabin John Bridge with a relatively fallout-free habitat remote from the highway, at least five replicates of 0.5 g (oven dry weight) lichen samples were collected from both Plummers Island and another Potomac River island, Bear Island. Bear Island is located approximately 6 km upstream of the Cabin John Bridge, and supports the same species found on Plummers Island as well as numerous presumably pollution-intolerant species not found on Plummers Island.

Lichen samples were also collected from a site located near the summit of Stony Man Mountain in Shenandoah National Park, western Virginia. These samples were collected to obtain lichen lead values from a site located some distance from a major metropolitan area. Another site was located in Aton Forest in Litchfield County, Connecticut, and nearby sites. Samples obtained from both roadside and nonroadside sites were collected for comparison.

Historical lichen material was obtained from collections made in the various study areas by B. Fink in the early part of this century, E. Leonard in the 1930's and W. R. Maxon and M. Hale in the 1950's. All material used for Pb analysis was taken from especially large samples housed in the U.S. National Herbarium. Less than half of the material in each packet was used for analysis.

Most samples, including the herbarium samples, were not washed before sampling. One set of recent samples from Plummers and Bear Islands was washed four times in distilled water for 15 minutes to compare washed and unwashed samples from sites exhibiting different levels of Pb fallout pollution. Samples were oven dried at 70°C for 72 hours, ground in a Wiley mill to pass a 20-mesh screen and analyzed for Pb content by the Ohio Agricultural Research and Development Center, a state laboratory in Wooster, Ohio. The samples were digested in nitric-perchloric acid and a diluted solution was run through an Inductively Coupled Plasma Spectrograph.

Differences in mean lichen lead concentration values from various locations and sampling times were determined statistically using *t*-tests.

Results

Table 1 shows lead concentration values for lichen samples collected from the study areas at various times. Present-day Pb concentrations in all species sampled from Plummers Island were significantly higher ($p < 0.01$) than concentrations in the same species from Bear Island, suggesting a relationship between elevated Pb burden from automobile exhaust pollution and the proximity of Plummers Island to the Cabin John Bridge. This elevated Pb burden was more obvious than that observed for the Connecticut roadside study area, which has a traffic flow of less than 1000 cars per day. The values obtained for roadside lichens were far less than those obtained for Plummers Island lichens. Also, only one species (*X. conspersa*) exhibited significantly higher Pb content in Connecticut roadside habitats than in nonroadside habitats.

The prominent increase in lead concentration on Plummers Island is a relatively recent phenomenon, having taken place since the Cabin John Bridge was built in 1965. However, it is evident from Table 1 that the atmospheric Pb burden at Plummers Island had been increasing prior to the building of the bridge. Lead values for both *P. baltimorensis* and *X. conspersa* were approximately 80 μg/g in 1907, compared to approximately 250 μg/g, the "background" (i.e. isolated from specific pollution sources) Pb levels of 1979 observed for *P. baltimorensis* at Bear Island and Skyline Drive. The difference between 1907 and 1979 Pb levels is not as great for *X. conspersa,* since present-day background levels are only approximately 100 μg/g for this species.

The differences in lead concentration exhibited by the three lichen species sampled in this study suggest that growth form plays a role in Pb accumulation. In each study area sampled, *P. baltimorensis* consistently exhibited a higher Pb concentration than *X. conspersa,* and *X. conspersa* exhibited a higher concentration than *C. subtenuis*. *Pseudoparmelia baltimorensis* and *X. conspersa* are both prostrate foliose lichens and would thus

TABLE 1. Lead concentration values observed for thalli of *Pseudoparmelia*, *Xanthoparmelia* and *Cladina* species collected from the study areas on various dates.

Species	Location	Collection date (sample size in parenthesis)	Pb(μg/g)
P. baltimorensis	Plummers Is., MD	1907 (2)	82.3 ± 8.2[1]
	Plummers Is., MD	1938 (2)	127.8 ± 14.8
	Plummers Is., MD	1958 (2)	342.9 ± 12.6
	Plummers Is., MD	1978 (6)	1893.5 ± 345.2
	Bear Is., MD	1979 (5)	273.0 ± 50.6
	Skyline Dr., VA	1979 (5)	218.5 ± 100.9
	Aton Forest, CT	1958 (1)	242.0
	Aton Forest, CT	1979 (2)	307.0 ± 17.0
	Litchfield Co., CT (roadside)	1979 (2)	332.0 ± 19.7
X. conspersa	Plummers Is., MD	1907 (2)	82.9 ± 2.2
	Plummers Is., MD	1978 (9)	1647.5 ± 42.7
	Bear Is., MD	1979 (7)	103.8 ± 8.7
	Skyline Dr., VA	1979 (2)	92.6 ± 3.4
	Aton Forest, CT	1963 (3)	128.0 ± 9.4
	Aton Forest, CT	1979 (2)	121.0 ± 7.1
	Litchfield Co., CT (roadside)	1979 (2)	397.5 ± 29.4
C. subtenuis	Plummers Is., MD	1907 (1)	35.0
	Plummers Is., MD	1933 (1)	57.0
	Plummers Is., MD	1978 (1)	272.0
	Bear Is., MD	1980 (3)	73.0 ± 15.0
	Near Baltimore, MD	1978 (1)	90.0
	Skyline Dr., VA	1979 (5)	63.9 ± 5.6
	Aton Forest, CT	1949 (1)	73.0

[1] All Pb concentration values mean μg/g ± 95% confidence intervals. Values with no confidence intervals are single values.

present a greater amount of flat particle-trapping surface area than *C. subtenuis,* which is finely branched, fruticose and erect. Of the foliose lichens, *P. baltimorensis* has broader lobes than *X. conspersa* (3–6 mm vs. 1–2 mm) and could be expected to intercept more particulate material. Thus, differences in Pb content exhibited by each lichen species may reflect differences in particulate trapping ability due to growth form.

Table 2 shows the effects of washing with distilled water on lead content of lichens collected from Plummers Island and Bear Island, Maryland. No statistical test was possible between washed and unwashed samples of *Cladina subtenuis* from Plummers Island because only one unwashed sample was obtained for analysis. However, for all other cases, washed thalli were found to contain significantly lower ($p < 0.01$) amounts of Pb than unwashed thalli.

Despite decreases in lead content as a result of washing, Plummers Island samples continued to exhibit elevated Pb values when compared with Bear Island samples. This observation suggests that Pb accumulation by lichens involves not only particulate trapping but also incorporation of Pb in thallus tissues.

DISCUSSION

Smith (1976) listed a number of lead concentration values for soils and plants collected in roadside environments. The highest values are found in soils and plants adjacent to

TABLE 2. Comparison of Pb concentration values obtained for washed and unwashed samples of *Pseudoparmelia, Xanthoparmelia* and *Cladina* species collected from Plummers and Bear Islands, Maryland.

	Pb(μg/g)			
	Plummers Island		Bear Island	
Species	Unwashed	Washed	Unwashed	Washed
P. baltimorensis	1893.5 ± 345.2[1] (6)	914.0 ± 12.0 (2)	273.0 ± 50.6 (5)	191.5 ± 35.0 (2)
X. conspersa	1647.5 ± 42.7 (9)	1295.0 ± 106.0 (2)	103.8 ± 8.7 (7)	71.5 ± 3.1 (2)
C. subtenuis	272.0 (1)	160.0 ± 4.0 (3)	73.0 ± 18.3 (3)	45.0 ± 6.0 (3)

[1] Mean μg/g ± 95% confidence intervals. Numbers in parentheses are sample sizes.

roads with the highest traffic volumes. Lead accumulation by plants is thought to occur primarily by above-ground interception of aerosols, rather than by root absorption. Representative Pb values for plants from a hypothetical roadside ecosystem bisected by a roadway averaging 24,000 vehicles per day are 200 μg/g for trees, 200 μg/g for grasses and 50–400 μg/g for crop species (Smith 1976).

Table 3 shows lead concentration values obtained for a number of lichen species collected from habitats influenced to varying degrees by automobile exhaust pollution. Lead values reported for lichen material sampled from Pb mine spoil (e.g. Shimwell & Laurie 1972) do not compare with the values obtained for lichens in this study, and are not included in this table. Lead values reported for other plants are not included here either, although a number of Pb studies have been conducted with vascular epiphytes. For example, Martinez et al. (1971) obtained Pb concentrations up to 966 μg/g in samples of Spanish moss (*Tillandsia usneoides,* a vascular epiphyte) adjacent to a heavily travelled four-lane highway in Baton Rouge, Louisiana.

The fact that cryptogams and other "air plants," such as *Tillandsia usneoides* (Shacklette & Connor 1973) are particularly efficient element accumulators, however, actually makes them less sensitive than other plants as integrators of pollution level along a Pb deposition gradient. For example, in a southern Finland roadside study of Pb accumulation, Laaksovirta et al. (1976) found that the lichen *Hypogymnia physodes* (L.) W. Wats. was less sensitive to changes in Pb than was tree bark. The lichens tended to accumulate higher concentrations of Pb than tree bark in habitats influenced by low traffic densities or habitats far removed from heavy roadside Pb deposition. Thus, the extremely efficient accumulation capacity of lichens may make them less useful as biological monitors of short-term air pollution than other organisms in certain applications, such as measuring a gradient of fallout away from a point source.

In his review of lead pollution studies in roadside ecosystems, Smith (1976) found that, on the average, vehicles release approximately 80 mg of Pb per km to the atmosphere. Most of this Pb is in the form of inorganic Pb salts in particles from 1 to 5 μm in diameter. The substantial decrease in lichen Pb content measured following distilled water washing (Table 2) underscores the importance of Pb aerosol in automobile exhaust pollution. Lichen thalli exposed to aerosol pollution will trap Pb-containing particles on their upper surfaces. Internal incorporation of Pb into lichen tissues probably does not occur immediately but rather over an as yet undetermined period of time.

Previous studies of lichen lead accumulation in areas polluted by automobile-generated

TABLE 3. Selected lead concentration values reported in the literature for lichen species sampled from areas variously affected by automobile-generated atmospheric lead fallout. Arranged according to decreasing average lead content.

Species	Location	Samples washed	Pb (μg/g)	Source
Lecanora muralis	Leeds, Yorkshire, U.K. (inner suburbs)	–	285–3124	Seaward 1974
Lecanora muralis	Leeds, Yorkshire, U.K. (outer suburbs)	–	500–1090	Seaward 1974
Lecanora muralis	Near Leeds, Yorkshire, U.K.	–	375–440	Seaward 1974
Peltigera rufescens	Urban and suburban areas near Scunthorpe, Lincolnshire, U.K.	–	46–454	Seaward 1973
Hypogymnia physodes	20 m from road, southern Finland	–	120–270	Laaksvirta et al. 1976
Parmelia conspersa	Sendai City, Japan	+	50–200	Saeki et al. 1975
Parmelia chlorochroa	Powder River Basin, Wyoming and Montana	+	30–200	Erdman & Gough 1977
Cladonia furcata	Urban and suburban areas near Scunthorpe, Lincolnshire, U.K.	–	79–180	Seaward 1973
Caloplaca aurantia	Urban area, Israel	+	150	Garty et al. 1977
Cornicularia aculeata	Urban and suburban area near Scunthorpe, Lincolnshire, U.K.	–	134	Seaward 1973
Lecanora muralis	Near Forfar, Scotland	–	132	Seaward 1974
Lecanora muralis	Near Buchanty, Scotland	–	102	Seaward 1974
Hypogymnia physodes	100 m from road, southern Finland	–	40–100	Laaksovirta et al. 1976
Caloplaca aurantia	Rural area, Israel	+	49	Garty et al. 1977
Cladina arbuscula	23 sites in Finland and northern Norway	–	3.4–30	Pakarinen et al. 1978
Cladonia stellaris "C. alpestris"	Unpolluted areas in Finland	–	20	Kauranen & Miettinen 1974
Hypogymnia physodes	Unpolluted area in North Yorkshire, U.K.	+	15	Seaward 1974
Cladonia stellaris	Unpolluted areas in central Sweden	+	8–17	Persson et al. 1974
Cladina mitis	23 sites in Finland and northern Norway	–	3.4–11.8	Pakarinen et al. 1978

Pb aerosols have not demonstrated unequivocally the mode of Pb uptake. Garty et al. (1977) compared heavy metal content of *Caloplaca aurantia* (Pers.) Hellb. samples obtained from urban, suburban and rural areas in Israel. They found that Pb content was higher in urban areas than in suburban areas, a result that suggested the importance of automobile exhaust pollution in urban areas. They were able to obtain significantly lower

Pb values in lichen samples subjected to experimental leaching with distilled water, a result suggesting that particulate deposition was responsible for elevated Pb values. However, they were never able to detect Pb in the washing solution. Therefore, they were not able to demonstrate unequivocally the importance of Pb aerosols in lichen Pb accumulation.

In a study of lead uptake by *Tillandsia usneoides* collected near a highway in Baton Rouge, Louisiana, Martinez et al. (1971) found little difference between Pb content of washed and unwashed samples. This suggested to them that Pb is in the tissues of the plant rather than in external particulate matter.

Brown (1976) found that lead and zinc accumulated by lichens from mine spoil material were not easily removed by washing with water. Lead appears to be bound on anionic sites external to the cell membrane (Brown & Slingsby 1972; Puckett et al. 1973), and is therefore probably of little metabolic importance. To the extent that Pb can be removed from lichens by washing in water, the ratio between surface-deposited Pb and Pb incorporated into lichen tissues can be estimated. The fact that lichens subjected to atmospheric Pb fallout contain significantly higher amounts of Pb than lichens at a distance from Pb fallout, regardless of washing, suggests that breakdown and uptake of Pb aerosols is a continuous process in lichens.

Another aspect of lead accumulation is the possible use of lichens in monitoring long-term increases in background Pb levels through large-scale atmospheric transport. For example, the retrospective values on Plummers Island in 1907, long before automobile pollution had any effect, were about 80 μg/g. By 1958, a time still before the bridge was built, this had increased fourfold. Bear Island now has somewhat lower values. On the other hand, values for *Pseudoparmelia baltimorensis* and *Xanthoparmelia conspersa* from Aton Forest in Connecticut are higher than similar "background" levels in Maryland and Virginia, suggesting that the northeastern U.S.A. is subjected to a higher average atmospheric Pb burden than the Middle Atlantic region. Schlesinger et al. (1974) found that New England receives a relatively high deposition of Pb due to its continental downwind position and relatively high precipitation. Thus, Pb aerosols produced in large population and industrial centers of the Great Lakes area and the Middle Atlantic states tend to be carried northeast. Analysis of additional lichen and bryophyte samples collected along the Atlantic coast may reveal other Pb deposition patterns.

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