



## Project Summary

# Atmospheric Fluxes and Geochemistries of Stable Pb, Pb-210, and Po-210 in Crystal Lake, Wisconsin

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In a lake system where atmospheric input greatly exceeds other source inputs, the atmospheric supply, water column transport, and sediment accumulation should be coupled. This research examined the exchange of lead between these compartments. The approach was to use Pb-210 as a natural tracer to provide experimental evidence concerning the movement of lead between various compartments, thereby linking the geochemical regime of lead in an integrated lake system.

The investigation was conducted in Crystal Lake, Wisconsin, an oligotrophic, non-calcareous, seepage lake. The lake has a surface area of 31 ha and a maximum depth of 21 m. Crystal Lake is located in a semi-remote, heavily forested area of north-central Wisconsin. The atmosphere serves as the principal route for transport of material to the lake. Stable Pb concentrations in the water column are of the order of 200 ng/kg.

At least 85% of the atmospheric input of Pb and Pb-210 to Crystal Lake occurs by wet deposition. More than 80% of the Pb entering the lake appears to be derived from anthropogenic sources. This input of anthropogenic Pb has resulted in an eight-fold increase in sediment Pb concentrations from 19 to 145  $\mu\text{g/g}$  over the past 150 years.

The net removal of Pb-210 from the water column occurred primarily as short-term deposition events. Deposition of Pb-210 was enhanced by elevated biological activity during bloom periods. Accelerated deposition

of Pb-210 was observed during fall turnover. The rate of deposition during these events exceeded the annual mean rate of 1.81  $\text{dpm/cm}^2/\text{yr}$  by twofold to threefold. Over the entire year, the rate of Pb-210 deposition varied by as much as an order of magnitude.

Deposition of Pb-210 was most rapid during the productive stratified period, averaging 2.09  $\text{dpm/cm}^2/\text{yr}$ . Consequently, the residence time of Pb-210 was 0.1 yr or less. The rate of Pb-210 deposition decreased 33% to 1.4  $\text{dpm/cm}^2/\text{yr}$  during the less productive unstratified period. Over this period, the residence time of Pb-210 averaged several tenths of a year. The overall mean residence time of Pb-210 is 0.2 yr, the same as that estimated for stable Pb.

At least 90% of the standing crop of Po-210 in Crystal Lake is formed *in situ* by decay of atmospherically supplied Pb-210. The activity ratio Po-210/Pb-210 averaged 0.66 in the whole lake during the stratified period. The subequilibrium condition of Po-210 relative to Pb-210 is attributed to: (1) the preferential removal of Pb-210 by plankton, and (2) the rapid removal of Pb-210 from the water column. Over this period, Po-210 exhibited a mean residence time of 0.7 yr. The residence time of Po-210 averaged several years during the unstratified period. The overall mean residence time of Po-210 is 2.6 yr. This suggests that it is recycled approximately 13 more times than Pb-210 before being removed from the water column. As a result of extensive recycling, a

large portion of the Po-210 decays to stable Pb-206 in the water column and never reaches the sediment.

Based on the Pb-210 geochronology of four sediment cores, the mass sedimentation rate in Crystal Lake is about  $8 \text{ mg/cm}^2/\text{yr}$  ( $0.14 \text{ cm/yr}$ ). Using this estimate of the sedimentation rate and the vertical distributions of Pb and Pb-210 in the sediment, the present day (1979) fluxes are  $1.2 \text{ } \mu\text{g/cm}^2/\text{yr}$  and  $1.75 \text{ dpm/cm}^2/\text{yr}$ , respectively. Of the total Pb flux, at least  $1.0 \text{ } \mu\text{g/cm}^2/\text{yr}$  can probably be ascribed to accumulation of anthropogenic Pb and the majority of it appears to be delivered by atmospheric fallout.

The dynamics of lead transport from the atmosphere to the lake surface and through the water column have been studied so that observations of spatial and temporal variability will include seasonal effects. The non-steady-state input and removal processes necessitate this type of approach if a realistic description of geochemical regimes in lakes is to be developed.

*This Project Summary was developed by EPA's Environmental Research Laboratory, Duluth, MN, to announce key findings of the research project that is fully documented in a separate report of the same title (see Project Report ordering information at back).*

## Introduction

The earth's atmosphere has been receiving widespread contamination by anthropogenic lead (Pb) for about 4500 yr (Patterson 1971). Industrial Pb has polluted all parts of the earth's troposphere by a minimum factor of 100 (Elias et al. 1975). Evidence also suggests that considerable quantities of Pb and other trace metals reach lakes and oceans through the atmosphere (Robbins and Edgington 1975, Duce et al. 1976, Schmidt and Andren 1983). A large quantity of these anthropogenic atmospheric compounds may result from the burning of fossil fuels. For example, several studies indicate that emissions from coal-fired steam plants contain particles that are enriched (especially the sub-micron fraction) in several trace metals as well as in some organics (Kaakinen et al. 1975, Klein et al. 1976, McElroy et al. 1982). The efficiency and parameters controlling removal of atmospheric micrometer and sub-micrometer-size particles to surface waters by wet and dry fallout remains a major area of research. Similarly, the fate of atmospherically derived metals to freshwater

systems is not well known. The internal cycling of metals in lakes must be studied in more detail if we are to understand, for example, the effect of lake acidification.

This investigation examined the correspondence between atmospheric loading, internal cycling, and sediment accumulation of Pb in a small oligotrophic lake located in a semi-remote area in northern Wisconsin. The approach was to use Pb-210 as a natural tracer for atmospheric stable Pb supplied to the lake. The high precision and accuracy capabilities of Pb-210 measurements relative to comparatively large stable Pb analysis uncertainties allow some of the important interactions responsible for maintaining the Pb inventories to be ascertained and the resultant fluxes between them identified.

The investigation was conducted in Crystal Lake, Wisconsin, an oligotrophic, non-calcerous seepage lake. The lake has a surface area of 31 ha and a maximum depth of 21 m. It is located in a semi-remote, heavily forested area of north-central Wisconsin. The atmosphere serves as the principal route for the transport of several metals to the lake. Sampling and analytical details have been detailed earlier (Talbot and Andren 1982, 1983 a,b).

## Results and Discussion

The aerosol concentrations of Pb, Pb-210, and Po-210 are shown in Figure 1 for June 1979 through June 1980. The weighted mean concentrations of Pb, Pb-210, and Po-210 were  $21 \text{ } \mu\text{g/m}^3$ ,  $22 \text{ dpm/1000 m}^3$ , and  $10 \text{ dpm/1000 m}^3$ , respectively. During this time the total atmospheric suspended particulate matter averaged  $17 \text{ } \mu\text{g/m}^3$ .

The average atmospheric metal concentrations for eight sampling intervals during 1979 to 1980 are shown as a function of particle size (stage number) in Table 1. It is evident that the later stages of the impactor collected the largest amounts of Pb, Pb-210, and Po-210. This suggests that these metals are associated with aerosol particles of  $< 1 \text{ } \mu\text{m}$  in diameter. The aerodynamic cutoff diameters corresponding to 50% collection efficiency with the Sierra impactor are: stage 1 ( $8.4 \text{ } \mu\text{m}$ ), stage 2 ( $3.6 \text{ } \mu\text{m}$ ), stage 3 ( $2.0 \text{ } \mu\text{m}$ ), stage 4 ( $0.97 \text{ } \mu\text{m}$ ), stage 5 ( $0.52 \text{ } \mu\text{m}$ ), and the backup ( $0.49 \text{ } \mu\text{m}$ ). These theoretical cutoff diameters are quoted by the manufacturer for a glass fiber filter collection substrate and a mass flow of air of  $1.1 \text{ m}^3/\text{min}$ . Particle bounceoff and re-entrainment under conditions of low relative humidity are thought to cause an unknown amount of overlap in particle

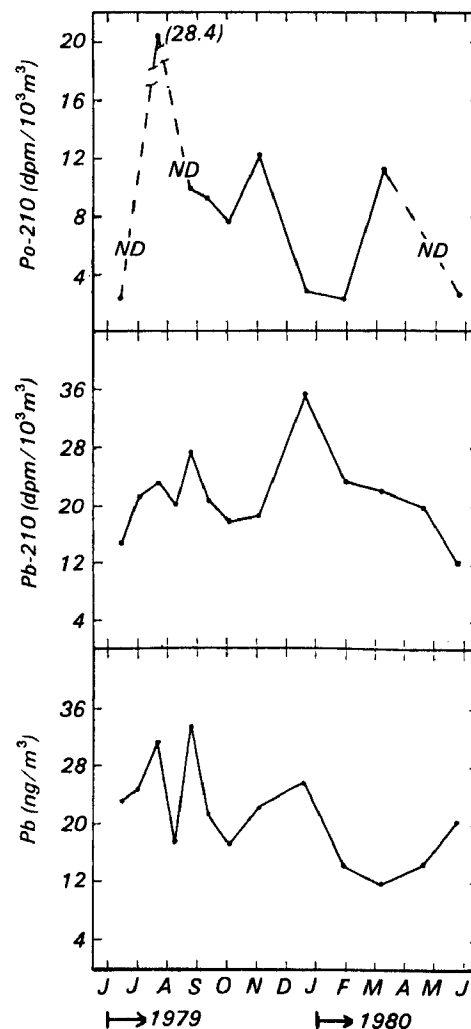


Figure 1. Atmospheric concentrations near ground level as a function of time at Vilas County, Wisconsin.

collection efficiency between successive stages of the impactor (Willeke 1975, Sievering et al. 1978).

The representative size distribution of Pb is similar to size distributions reported for Pb in urban areas of the United States (Lee et al. 1972, Gladney et al. 1974, Paciga et al. 1975, Davidson 1977) and over the Atlantic Ocean by Duce et al. (1976). In contrast to Pb, Al concentrations did not appear to vary significantly with particle size. Such a monotonous size distribution is commonly observed for Al (Paciga et al. 1975, Duce et al. 1976). The mass median equivalent diameter (MMD) was  $1.7 \text{ } \mu\text{m}$  for Al and  $0.55 \text{ } \mu\text{m}$  for Pb, consistent with the general consensus that the MMD for Al is about  $1 \text{ to } 4 \text{ } \mu\text{m}$  and is  $\leq 0.5 \text{ } \mu\text{m}$  for Pb (Lee et al. 1972, Colovos et al. 1974, Rahn 1976, Davidson 1977,

**Table 1.** Average Metal Concentrations (Mean  $\pm$  S.D.) As a Function of Particle Size ( $N = 8$ )

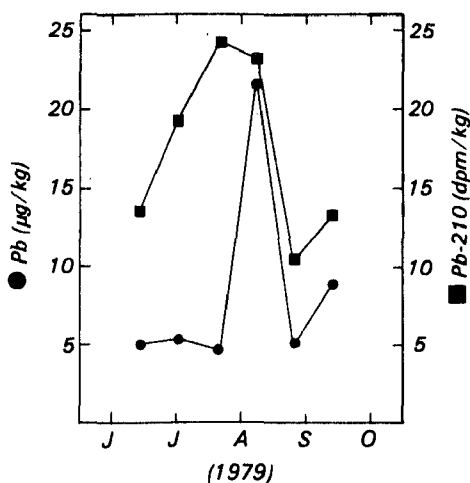
Stage	Al (ng/m <sup>3</sup> )	Pb (ng/m <sup>3</sup> )	Pb-210 (dpm/10 <sup>6</sup> m <sup>3</sup> )	Po-210 (dpm/10 <sup>6</sup> m <sup>3</sup> )
1	32 $\pm$ 24	0.5 $\pm$ 0.2	134 $\pm$ 46	127 $\pm$ 91
2	25 $\pm$ 13	0.9 $\pm$ 0.4	215 $\pm$ 130	256 $\pm$ 283
3	14 $\pm$ 16	1.2 $\pm$ 0.5	379 $\pm$ 124	315 $\pm$ 137
4	16 $\pm$ 14	1.9 $\pm$ 0.8	1,360 $\pm$ 1,313	1,372 $\pm$ 1,046
5	13 $\pm$ 12	3.6 $\pm$ 1.3	2,562 $\pm$ 1,671	2,247 $\pm$ 1,990
Backup	35 $\pm$ 18	10.3 $\pm$ 4.2	11,499 $\pm$ 4,637	3,423 $\pm$ 1,387
$\Sigma$	135	18.4	16,149	7,767
MMD <sup>a</sup>	1.7	0.55	0.40	0.65

<sup>a</sup> Mass median equivalent diameter ( $\mu$ m).

Nraigu 1978). The representative size distributions of Pb-210 and Po-210 indicate that the MMD for Pb-210 is near 0.40  $\mu$ m and for Po-210 is near 0.65  $\mu$ m. There appear to be no other estimates of MMDs reported for Pb-210 or Po-210.

The concentrations of Pb and Pb-210 in rainfall are illustrated in Figure 2. The activity of Pb-210 in rain is corrected for decay to the mid-point of the sampling interval. The weighted mean concentration of Pb was 7.9  $\mu$ g/kg and 17.4 dpm/kg for Pb-210. Lead concentrations in rain were <10  $\mu$ g/kg, except for a brief period in early August when the concentration was  $\sim$  20  $\mu$ g/kg. Mid-summer concentrations of Pb-210 were higher than those in spring or early fall. A steady increase in Pb-210 concentration was observed between June and August. Thereafter, the concentration decreased rapidly to around 13 dpm/liter, which is the same concentration observed during early June.

There is no apparent statistical relationship ( $|r| < 0.5$ ) in these data between Pb or Pb-210 concentration and the amount of rainfall. Several authors have observed a statistical relationship between

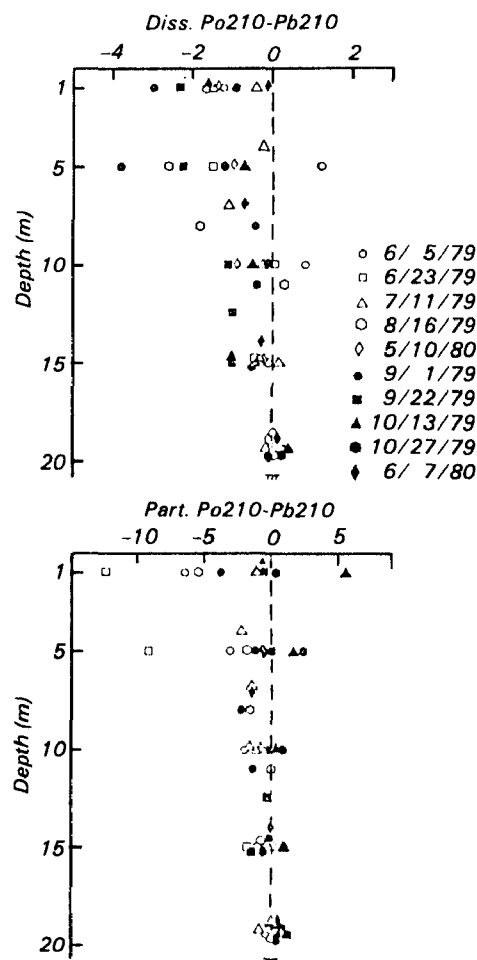


**Figure 2.** Concentrations of Pb and Pb-210 in rainfall as a function of time at Vilas County, Wisconsin.

precipitation volumes and Pb concentrations (Chow and Earl 1970, Parungo and Rhea 1970, Ter Haar et al. 1967). Stolzenburg (1979) observed an inverse relationship on one occasion between Pb concentration and the amount of rain falling on the Menomonee River Watershed, Wisconsin. Turekian et al. (1977) suggested that at some locations Pb-210 concentration is correlated positively with amount of rainfall. It is not appropriate to examine such a relationship in the samples collected for this study, since individual events were not sampled. Instead, the data represent integrated rainfall events over a two-week time interval. The data were collected in this manner since integrated samples over short time intervals are best for estimating wet deposition (Turekian et al. 1974).

Polonium-210 was detected in rain only during late spring and early fall. Concentrations of Po-210 were between 0.2 and 2 dpm/liter during these periods. Post et al. (1972) observed 0.3 to 1 dpm/liter of Po-210 in rain at Boulder, Colorado. Francis et al. (1970) found Po-210 concentrations of 0.4 to 7 dpm/liter in rain at Madison, Wisconsin. This limited set of data suggests that the average concentration of Po-210 in rain over the midwestern states may range from a few tenths to several dpm/liter. High concentrations (>1 dpm/liter) may be related to periods of high dust resuspension in the atmosphere. The 1.8 dpm/liter concentration of Po-210 observed in early June at Vilas County, Wisconsin may reflect this phenomena.

A summary of Po-210/Pb-210 disequilibrium data for the dissolved and particulate phases is shown in Figure 3. Ordinarily, more than 60% of the Pb-210 in Crystal Lake is in the particulate phase. Surface water (0 to 10 m) concentrations of Pb-210 in dissolved and the particulate phase were always higher than those in deep water (10 to 21 m). The higher surface concentrations are probably maintained by delivery of unsupported Pb-210 produced in the atmosphere by



**Figure 3.** Summary of Po-210/Pb-210 disequilibria in Crystal Lake for all sampling dates. Data are plotted as normalized concentrations (dpm/100 kg).

Rn-222 decay. The atmosphere appears to be the principal source of Pb-210 to Crystal Lake. This means the majority of Pb-210 in the lake is unsupported.

In general, particulate phase Pb-210 varied more than dissolved Pb-210. Fluctuations in total Pb-210 were, therefore, usually brought about by changes in particulate Pb-210. Much of the temporal variation in particulate Pb-210 concentrations probably is associated with changes in the lake's biomass. This is especially true in surface water during the productive spring and summer stratified periods.

Particulate Pb-210 concentrations were lowest when the lake was not stratified but when it still showed a trend of decreasing concentration going from surface to bottom. During the unstratified

period particulate phase Pb-210 decreased from an average of 4 dpm/100 kg in surface water to  $\leq 0.5$  dpm/100 kg near the sediment. About 65% of the Pb-210 was in the particulate phase and there was no discernible deviation from this distribution in the water column.

Particulate Pb-210 was highly variable in surface water during the stratified period. Concentrations varied from about 2 to 20 dpm/100 kg. Uncommonly high surface concentrations (20 dpm/100 kg) were observed during June and mid-August. More than 80% of the Pb-210 was in the particulate phase at these times. As discussed previously, the high suspended particulate matter concentrations strongly suggest that plankton blooms occurred during these periods. Freshwater plankton are known to concentrate trace metals to a high degree and concentration factors (i.e., concentration in plankton:concentration in water) of  $> 10^3$  have been observed in this study, and by Holm (1978). The high particulate Pb-210 concentrations observed in Crystal Lake may reflect plankton Pb-210 concentrations.

Deep water (10 to 21 m) particulate Pb-210 concentrations were lower and not as variable as surface water concentrations. The lowest concentrations always occurred near the bottom and usually were in the range 0.5 to 1.0 dpm/100 kg. Concentrations at depths of 10 to 15 m varied from 2.0 to 4.0 dpm/100 kg. As in surface water, deep water particulate Pb-210 concentrations were highest during suspected plankton bloom periods.

Dissolved Pb-210 concentrations in surface water varied little with time, and were ordinarily around 3.0 dpm/100 kg. Deep water concentrations ranged from 0.2 to 2.0 dpm/100 kg with the lowest concentrations occurring near the bottom sediments.

Dissolved concentrations of Pb-210 increased steadily toward the end of the stratified period (i.e., after 1 September 1979). Suspended particulate matter concentrations decreased during this time and may be partially responsible for the higher dissolved concentrations. In addition, the lower suspended particulate matter concentrations indicate that biological productivity was decreasing. This might also tend to favor higher dissolved concentrations, since dissolved deficiencies in productive ocean waters have been attributed to rapid removal by plankton (Shannon et al. 1970, Turekian et al. 1974).

Dissolved concentrations of Pb-210 varied more during the stratified period than during the unstratified period. This

is probably due to the interaction of dissolved Pb-210 with plankton during the biologically productive summer months. During the unstratified period, dissolved phase Pb-210 averaged 35% of the total Pb-210. This suggests that a steady-state condition with respect to supply and removal of Pb-210 may be maintained during the biologically less productive months.

The depth distribution of Po-210/Pb-210 disequilibria in Crystal Lake is illustrated in Figure 3. With the exception of a few spurious points, dissolved Po-210 activities always showed a deficiency relative to Pb-210. The surface water exhibited the greatest deficit, which is probably the result of rapid removal of Po-210 by plankton. Similar Po-210/Pb-210 disequilibria have been observed in the surface ocean and are attributed to rapid removal of Po-210 by biosphere activities (Shannon et al. 1970, Turekian et al. 1974, Bacon et al. 1976, Bacon 1977).

Particulate phase Po-210 activities also showed a deficit, but excess Po-210 was present on several occasions. Excess particulate Po-210 was commonly observed a few meters above the sediment. This excess Po-210 is thought to be related to activities of migrating zooplankton.

Activity ratios of Po-210:Pb-210 are summarized in Table 2. Dissolved Po-210 activities were less than those of Pb-210 by about 21%. This difference is significant at the 95% confidence level. Particulate phase activities, however, are not significantly different at the 95% confidence level. Apparently the dissolved phase deficiency is not balanced by a particulate phase excess. Total Po-210 activities are consequently less than Pb-210 activities by about 11% (significant at the 95% confidence limit). This suggests that Po-210 is removed from the water column to maintain the dissolved deficit. Interaction with living and dead particulate matter probably expedites removal. Thomson and Turekian (1976) observed a mean activity ratio Po-210:Pb-210 of 0.89 in an upwelling (productive) area of the eastern South Pacific. They attributed the depletion of Po-210 relative to Pb-210 to active scavenging of Po-210 from surface water by biological activity.

**Table 2.** Summary of Po-210:Pb-210 Activity Ratios in Crystal Lake. Confidence Limits for the Mean were Calculated at the 95% Level

	Median	Mean
Dissolved	0.63	0.79 $\pm$ 0.18
Particulate	0.89	1.04 $\pm$ 0.18
Total	0.82	0.89 $\pm$ 0.13

The data were subsequently used to interpret the metal fluxes in Crystal Lake. Details of the various models and transport calculations have recently been published (Talbot and Andren 1981, 1983 a,b). It was found that nonsteady-state processes greatly influence atmospheric input and in-lake removal of these metals. Input and removal processes in the system as a whole, however, appear to approximate steady-state conditions over a time span of about 1 yr.

The mean atmospheric fluxes of Pb and Pb-210 for 1978 to 1980 were estimated to be 1.0  $\mu\text{g}/\text{cm}^2/\text{yr}$  and 1.79 dpm/ $\text{cm}^2/\text{yr}$ , respectively. These fluxes may be considered representative of the year-to-year mean fluxes with the caveat that an uncertainty of about 20% is associated with each value. The atmospheric flux of Po-210 appears to be  $< 1.0$  dpm/ $\text{cm}^2/\text{yr}$ , or  $< 10\%$  that of Pb-210. Wet deposition accounts for at least 85% of the annual mean fallout of Pb, Pb-210, and Po-210 at this site.

Biological, chemical, and physical in-lake processes interact in a complex manner which subsequently determines the fate of Pb, Pb-210, and Po-210 in Crystal Lake. Their net removal from the geochemical regime(s) occurs primarily as short-term deposition events. Despite a scant biological population, the biota in Crystal Lake strongly influence transport dynamics of Pb-210 and Po-210. Plankton, in fact, appear to overwhelmingly mediate removal of Pb-210 and Po-210 from the water column during the productive stratified period. Fall turnover also is important in removing Pb-210.

The in-lake rate of Pb-210 deposition varied more than six-fold over the whole year. Deposition of Pb-210 during the stratified period was estimated to be  $2.09 \pm 0.57$  dpm/ $\text{cm}^2/\text{yr}$ . The flux was 33% less during the unstratified period ( $1.40 \pm 0.48$  dpm/ $\text{cm}^2/\text{yr}$ ) than it was over the stratified period. The annual mean in-lake flux of Pb-210 was estimated to be  $1.81 \pm 0.50$  dpm/ $\text{cm}^2/\text{yr}$ . The excellent agreement between estimates of in-lake deposition and atmospheric supply of Pb-210 provides strong support for the contention that the atmosphere is the principal source of Pb-210 to Crystal Lake.

The mean residence times of Pb-210 in the dissolved and the particulate phases fluctuated by more than an order of magnitude ( $\sim 0.01$  to 1 yr). The surface mixed layer (SML) is more dynamic than the deep layer (DL) in this regard, especially during the productive stratified period. The overall mean residence times

of Pb and Pb-210 in Crystal Lake essentially are equal at 0.20 yr.

Most of the Po-210 in Crystal Lake is formed *in situ* by decay of atmospherically supplied Pb-210. Extensive recycling, most likely biologically mediated, keeps Po-210 in the water column an average of 13 times longer than Pb-210. The residence time of Po-210 in the dissolved and the particulate phase is on the order of years. Thus, a large fraction of the Po-210 is lost in the water column by decay to stable Pb-206.

During the stratified period the residence time of Po-210 averaged 0.7 yr. The activity ratio Po-210:Pb-210 was 0.66 during the same period. This deficit of Po-210 relative to Pb-210 is attributed to preferential uptake and rapid deposition of Po-210 by biological pathways. When the lake was least productive, activities of Po-210 approached equilibrium levels with Pb-210.

The mass sedimentation rate in Crystal Lake appears to be about 8 mg/cm<sup>2</sup>/yr (0.14 cm/yr). Based on this estimate and the vertical distributions of Pb and Pb-210 in the sediment, the present day (1979) fluxes are 1.2 μg/cm<sup>2</sup>/yr and 1.75 dpm/cm<sup>2</sup>/yr, respectively. Of the total Pb flux, 1.0 μg/cm<sup>2</sup>/yr can probably be ascribed to accumulation of anthropogenic Pb. The input of anthropogenic Pb apparently has increased sediment Pb concentrations by eight-fold over the past 150 yr, from 19 to 145 μg/g.

This research has applicability to many lakes. Although the biological, chemical, and physical characteristics of Crystal Lake may be somewhat less complicated when compared to other lakes, metal cycling in the water column is probably controlled by analogous processes in the majority of lake systems. The removal of metals from surface waters of Lake Michigan, for example, may follow similar dynamics as observed in the whole water column of Crystal Lake.

It follows that most of the net deposition of metals in lakes is associated with biological, chemical, or physical in-lake events. It is recommended that future studies of geochemical regimes in lakes be designed to examine some of the aspects of the transport dynamics. Such studies should be coupled with an in-depth look at specific particle-metal associations. The role of plankton in metal transport also needs to be explored explicitly using *in situ* experiments. Detailed information on the specific phases carrying metals and on the corresponding transport kinetics is necessary for ultimately constructing a realistic picture of metal transport in

lakes. The results of the present research indicate that within certain limitations the natural tracer Pb-210 is a valuable tool for obtaining direct and accurate experimental data on the geochemical regimes of metals in lakes.

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*The complete report, entitled "Atmospheric Fluxes and Geochemistries of Stable Pb, Pb-210, and Po-210 in Crystal Lake, Wisconsin," (Order No. PB 84-148 360; Cost: \$13.00, subject to change) will be available only from:*

*National Technical Information Service  
5285 Port Royal Road  
Springfield, VA 22161  
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