Mercury and Lead Budgets for Lochnagar, a Scottish Mountain Lake and Its Catchment

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Lochnagar is a mountain lake located to the southeast of the Cairngorm Mountains in Scotland. The inputs and outputs of Hg and Pb and their distribution within the various ecosystem compartments were measured. Further, 17 sediment cores and 10 catchment peat cores were taken and dated using spheroidal carbonaceous particle (SCP) and ^{210}Pb techniques. Total and anthropogenic Hg and Pb inventories since the 1860s for the lake basin and the catchment peats were calculated using this multiple core strategy. Hg sediment flux profiles based on the whole lake basin show that the flux to the sediments increased from the 1880s until the 1970s. This was followed by a relatively stable period (1970s to the present), during which the flux was approximately twice that of the 1880s. Similarly, the Pb flux increased from the 1860s until the 1940s and was also followed by a relatively stable period through to the present. Hg and Pb budgets for the whole catchment for 1998 indicated that 78% of the Hg and 91% of the Pb input to the lake were transported from the catchment. Hence, the expected decline resulting from the decrease in the atmospheric deposition of Pb was obscured in the sediment record. It is estimated that 77% of the total Hg and 90% of the total Pb deposited since the 1860s, and stored in the upper layers of the catchment peat soils, are from anthropogenic sources. The increased storage of Hg and Pb in the catchment implies that this will be a major source of these metals for the lake for many years. This will delay the restoration of the lake system, despite reductions in emissions to the atmosphere and subsequent deposition.

Introduction

Lake sediments store important information about past conditions of a lake and its catchment and provide an excellent archive of historical changes in the industrial discharge of potentially toxic metals to the environment (*1−3*). Therefore, the analyses of trace metals from sediments can provide an insight into the temporal trends of metal inputs to lake ecosystems.

To study the atmospheric deposition of trace metals, the link between lake sediment records and atmospheric deposition history has been used on numerous occasions (*4−8*). Over industrial time, vast amounts of trace metals have been emitted to the atmosphere from anthropogenic sources, resulting in widespread environmental contamination (*9, 10*), while natural physical and biogeochemical cycles of these trace metals result in their redistribution (*11*). Catchment

soils contaminated by atmospheric deposition may, in turn, become a source of trace metals for a lake and change the proportion of supply of trace metals from direct atmospheric deposition to greater catchment inputs. Therefore, great care must be taken when using lake sediments to describe trends in atmospheric deposition.

The aims of this study were to calculate inventories of Hg and Pb in Lochnagar sediments, to establish decadal sedimentary flux profiles of Hg and Pb in the lake, to use mass balances to reveal the impact of Hg and Pb pollution of catchment soils to lake inputs, and to assess the possible influence of the soil inputs on the future restoration of the lake system from Hg and Pb pollution.

Environmental Setting. Lochnagar is a relatively remote, high altitude (785 m) cirque lake to the southeast of the Cairngorm Mountains in Scotland and lies below a northeast facing steep backwall, which rises to the summit of the same name. The altitude of this summit is 1155 m. This headwater lake is roughly pear-shaped and occupies an area of 9.8 ha with a precipitous catchment of 91.9 ha. Its floor slopes quite sharply, with a deep point of 26 m offset from the center of the lake toward the backwall of the cirque. Although no distinguishable inflow feeds the lake, it is not a closed system, and drains to the northeast through a series of small pools to the Lochnagar Burn which feeds the Gelder Burn, a south-bank tributary of the River Dee. The water level of the lake is usually stable, with a mean water depth of 8.4 m and a volume of 0.82×10^6 m³. Snowmelt and rainfall comprise the major inputs, and annual precipitation in 1998 was 1665 mm. The catchment bedrock is biotite granite overlain in places on the lower slopes by blanket peats. The steeper parts of the catchment consist of bare rock faces and intervening scree slopes, but despite the existence of some localized peat erosion along the relatively flat eastern shore of the lake, the lake water is clear (*12*). At over 700 m, the catchment lies above the limit of summer sheep grazing in the region, and there is no evidence of any land-use change or active land management within the catchment. The lake is known to have been contaminated by trace metals from atmospheric deposition (*12−13*), and this is the only source of anthropogenic trace metals at the site. Aquatic plants growing on the lake sediments, and fish in the lake, are rare, and bioturbation in the lake sediments is not thought to be significant.

Methods

Sampling. All sample bottles and glassware used in sample collection and analysis were rigorously cleaned. This involved soaking in a 5% Decon 90 solution for 24 h, multiple rinses with deionzed distilled water, soaking in 1 M HCl for another 24 h, followed by extensive rinsing in deionzed distilled water, and double-bagging after drying. During all of the sample processing, an extra cleaning protocol was used in order to avoid contamination (*14, 15*).

Bulk deposition collected for Hg analysis used an IVL-type bulk collector (*16, 17*) and was sampled monthly. Bulk deposition collected for Pb analysis used an RS1-type collector and was sampled fortnightly. Lake water samples for Hg and Pb were collected monthly and fortnightly, respectively, by submerging a rigorously acid-leached 250-mL Teflon bottle approximately 20 cm beneath the surface of the water near the outflow, where the lake water is well-mixed and metal concentrations are close to those in the water lost via the outflow stream. The bottles were completely filled and tightened by gloved hands under the water. Bulk deposition and lake water samples were acidified using suprapure HCl

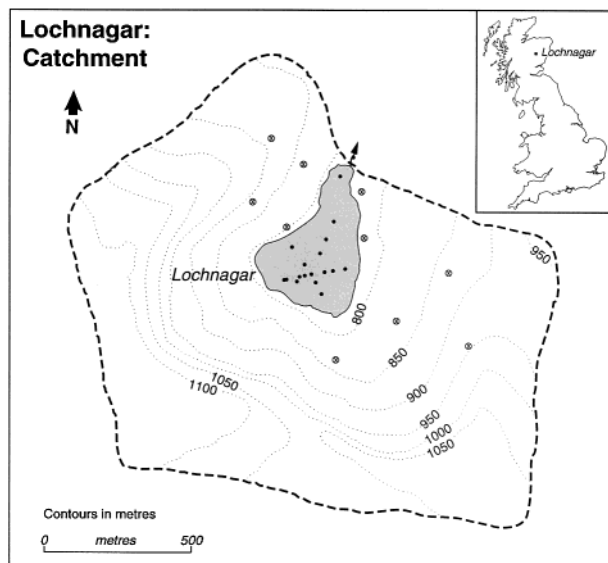* Corresponding author phone: (+44) 20 7679 5344; fax: (+44) 20 7679 7565; e-mail: hyang@geog.ucl.ac.uk.

FIGURE 1. Location of Lochnagar in the U.K. and the locations of sampling positions: (•) is a sediment coring site, (⊗) is a catchment peat coring site.

for Hg and HNO₃ for Pb analyses. All bottles were double-bagged for transport to the laboratory and stored cool (4 °C) prior to analysis.

Seventeen sediment cores were taken between June 28 and July 2, 1997, along five transects radiating from the central area of the lake using a Glew gravity corer fitted with a 7.7 cm inner diameter polycarbonate barrel (Figure 1). The sediment cores were sectioned using a stainless steel slicer at 0.5 cm intervals from the surface to 7 cm depth and then at 1 cm intervals from 7 cm to the bottom of the cores. Ten catchment peat cores representing different areas (vegetation density, catchment slope, boulder density, altitude, aspect) were taken using a 10.2 cm inner diameter PVC tube inserted by hand in July 1997 (Figure 1). The soil cores were sliced using a stainless steel knife at 1 cm intervals from the surface to 10 cm depth and then at 2 cm intervals from 10 cm to the bottom. Wet density measurements of sediment samples were conducted using a 2 cm³ container and, for soils, by using the total weight divided by the sample volume. All samples were measured for moisture content and organic matter (as loss on ignition (LOI)) and were freeze-dried.

Core Dating. Two sediment cores were dated using radiometric techniques (²¹⁰Pb, ¹³⁷Cs, ¹³⁴Cs, ²⁴¹Am) (18). Although Sarazin et al. (19) demonstrated that highly porous sediment could not be dated using ²¹⁰Pb because of sediment compaction, sedimentation rates in Lochnagar are very low, and dry sediment density values at different sediment depths are relatively stable. Therefore, sediment compaction is not high (20), and sediments were dated by ²¹⁰Pb according to the constant rate of supply model (21).

All of the sediment and soil cores were analyzed for spheroidal carbonaceous particles (SCPs) (20, 22). The onset of the SCP record in lake sediments is known to be the 1860s; hence, all dated and nondated sediment cores could be stratigraphically cross-correlated (20, 22) at this depth/date. Similarly, other SCP profile features can be used in this way (22). The soil cores were also dated using their SCP profiles (20, 23). Methods for dating Lochnagar lake sediments and catchment soils using SCPs are fully described elsewhere (22, 23).

Mercury and Pb Analyses. Bulk deposition and lake water samples were measured using cold vapor-atomic fluorescence spectrometry (CV-AFS) for Hg and inductively coupled plasma-mass spectrometry (ICP-MS) for Pb. The detection

limits for Hg and Pb using these techniques were 0.3 ng L⁻¹ and 0.002 μg L⁻¹, respectively; standard reference material 1640 was used as a reference sample for trace element measurements in natural waters, while standard solutions and blanks were measured during the measurement of deposition and lake waters.

For Hg determinations in sediment and soil samples, 0.2 g of sample was extracted using 8 mL of concentrated Aristar HNO₃ at 100 °C for 1 h in rigorously acid-leached 50-mL Teflon beakers. Mercury concentrations were measured by cold vapor-atomic absorption spectrometry (CV-AAS) following reduction with SnCl₂ (24). During digestion and measurement, standard reference material (stream sediment GBW07305) and sample blanks were analyzed every 20 samples. Measured mean concentrations of Hg in GBW07305 ($n = 12$; relative standard deviation (RSD) = 8.6 ng g⁻¹; certified value = 100 ± 10 ng g⁻¹) was 93 ng g⁻¹. Lead in sediment and soil samples was measured using isotope source X-ray fluorescence (25). Recoveries of certified reference materials (Buffalo River sediment 2704, Pond sediment no. 2) were within 10% of certified values.

Inventory Calculation. Because the distribution of sediments within a lake may be uneven, element inventories can be calculated in different ways (2, 26). In Lochnagar, there is a good relationship between both contaminated sediment thickness and trace metal concentration with water depth along individual transects, but these parameters do not have a good relationship with water depth across the lake basin as a whole (20). Therefore, in this study, the whole area of sediment accumulation was divided using depth contours, and then these were further subdivided such that each area could be represented by a nearest single sediment core. As the sediment layers equating to the 1860s had been determined for all of the sediment cores, and sediment dry densities had been measured, the dry mass accumulated since 1860 could be calculated for all cores. Using a constant dry mass sedimentation rate model (21), the 14 decadal intervals from 1860 to 1998 were determined for each core. The metal inventory for decade f in sediment area S_i represented by core i is, thus

$$T_{if} = \sum S_i R_{ij} C_{ij} D_{ij}$$

where R_{ij} is the sediment thickness for interval j within decade f in core i, C_{ij} is the metal (Hg or Pb) concentration for sediment interval j, and D_{ij} is the sediment dry density for interval j.

Summing the inventories in decade f for all individual sediment areas gives decadal inventories of Hg and Pb for the whole sediment basin. Summing all decadal inventories gives the inventories of Hg and Pb since 1860 for the whole sediment basin. Mercury and lead concentrations in the sediments formed before the 1860s are considered to be background levels.

The inventories of Hg and Pb for the whole catchment were also calculated by summing areas represented by relevant peat cores (27). In catchment peats, the relatively stable, low Hg and Pb concentrations in the lower layers of the cores were considered to be background levels. In the contaminated layers, the exceeding of the background values of Hg and Pb were termed the anthropogenic fraction. The total value was calculated from this exceeding value plus the background value existing in the sediment or peat layers formed since 1860.

Water Loss via the Outflow. In June 1996, a stageboard calibrated by dilution gauging at a range of flows was set up in the outflow stream in order to monitor the water flow. Stageboard reading was recorded fortnightly, and water loss via the outflow was estimated.

TABLE 1. Representative Core Areas, Dry-Mass Sedimentation Rates, and Dry Mass of Sediments since 1860 in the Sediment Cores

| core	NAG9	NAG10	NAG11	NAG12	NAG13	NAG14	NAG15	NAG16	NAG17	NAG18	NAG19	NAG20	NAG21	NAG22	NAG25
representative area (m^2)	1836	2223	1254	2416	4591	5654	2464	2706	1353	4108	5606	11791	5026	2513	7152
sediment depth dated at 1860 (cm)	12	6	6	30	9	7	5	4.5	4	3	24	9	15	15	2
dry mass (kg m^{-2})	22.6	9.9	14.8	38.8	13.7	9.6	25.6	11.0	6.5	16.5	18.9	11.4	23.0	20.2	2.3
dry-mass sedimentation rate (g m^{-2} y^{-1})	165	73	108	278	100	70	178	80	48	120	138	83	168	148	17
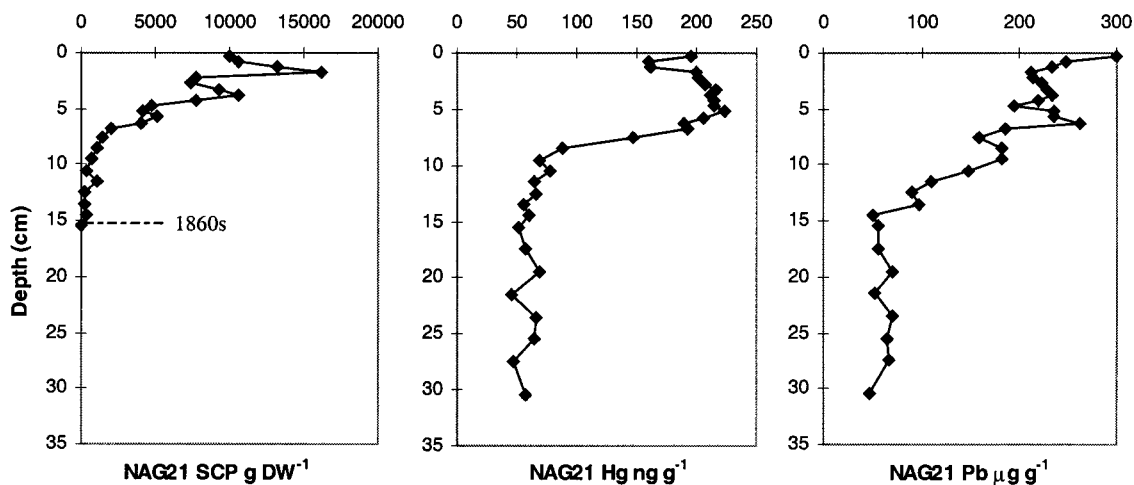
FIGURE 2. Profiles of SCPs, Hg, and Pb in sediment core NAG21 taken from Lochnagar. The start of the SCP record was dated to 1860 $\pm$ 5 using the ^{210}Pb technique.

Results and Discussion

Mercury and Pb Distribution. Mercury and Pb concentrations in bulk deposition between October 1997 and October 1998 were in the range of 12–35 ng L^{-1} with a mean of 23 ng L^{-1} ($n = 12$) and a range of 0.24–3.75 μg L^{-1} with a mean of 1.11 μg L^{-1} ($n = 26$), respectively. The mean Pb concentration in the deposition samples collected over this period is lower than the annual mean value in bulk deposition samples collected between 1987 and 1995 in Banchory, about 32 km from Lochnagar. Banchory data show that the annual mean values of Pb concentration between 1987 and 1995 reduced from about 10 to 2 μg L^{-1} (28). The calculated Hg deposition flux at Lochnagar from October 8, 1997 to October 7, 1998 (35.9 μg m^{-2} y^{-1}) is within the range of annual deposition fluxes monitored at Banchory from 1992 to 1995 (28). In 2000, Pb concentrations in Lochnagar bulk deposition samples were further reduced with a range of 0.02–1.30 μg L^{-1} and a mean of 0.35 μg L^{-1} ($n = 21$) (unpublished data). Mercury and lead concentrations in lake water between October 1997 and October 1998 were in the range of 5–21 ng L^{-1} with a mean of 14 ng L^{-1} ($n = 12$) and a range of 0.51–1.19 μg L^{-1} with a mean at 0.82 μg L^{-1} ($n = 12$), respectively. Lead concentrations in lake water samples collected during 2000 are in the range of 0.05–5.68 μg L^{-1} with mean at 0.80 μg L^{-1} ($n = 25$). Hence, lake water Pb values have remained similar, while values in deposition have been reduced. These Hg and Pb concentrations in unfiltered Lochnagar water samples are higher than "uncontaminated" lake waters in the northeastern United States sampled in 1995 and 1996 (29), and Hg concentrations are at the upper end of the range for Swedish forest lakes sampled in 1986 (16).

Mercury and lead are enriched in the upper parts of the catchment peats (27), and the peats have been severely eroded, resulting in Hg and Pb in-washed into the lake water.

On July 8, 1997, during a brief period of heavy rain at Lochnagar, many small inflows formed in the catchment and flowed into the lake. A water sample was collected from one of these inflows near the lake in order to assess the influence of such ephemeral inflows. A lake water sample was also collected. The Hg concentration of this inflow water sample was 26 ng L^{-1}, while that of the lake water sample (21 ng L^{-1}) was the highest water concentration recorded between July 1997 and October 1999. Two conclusions may be drawn from these data. First, if rainfall is very heavy, rainwater may not pass through the soil but flow over the surface. This results in enhanced Hg input to the lake, resulting in a rise in Hg concentration and, because of the rapid flushing of the epilimnion, a high Hg concentration in the water lost via the outflow. Second, rainwater flowing through the soil is depleted in metals as a result of binding to organic matter; hence, the soils store atmospherically deposited metals before they can reach the lake system. However, with erosion of these catchment soils, Hg and other stored trace metals could be carried into the lake water.

Sediment accumulation rates vary in different areas of the lake, and the sediment thickness formed since 1860 in the sediment cores varies from 2 to 30 cm (Table 1). The Hg and Pb profiles for the sediment cores generally show a slow, steady increase in Hg and Pb concentrations since the 1860s followed by a rapid increase in concentration in upper layers (e.g., NAG21 in Figure 2; 30). Preindustrial concentrations of Hg and Pb in the sediment cores are approximately 30–80 ng g^{-1} and 40–90 μg g^{-1}, respectively, with concentrations in the upper parts of the cores ranging from 150 to 250 ng g^{-1} and 150 to 300 μg g^{-1}, respectively. This shows that Lochnagar has been significantly contaminated by Hg and Pb since industrialization. However, even for the same metal, there are differences in the trends of the sediment profiles

TABLE 2. Mercury and Lead Inventories for Lochnagar[a]

	Hg(g)		Pb(kg)	
	total	anthropogenic	total	anthropogenic
lake sediments	115.7	71.4 (11.7 g/ha)	150.3	93.8 (15.5 kg/ha)
catchment peats	1060	820 (23.6 g/ha)	936	839 (24.1 kg/ha)
lake water	9.27		0.68	
catchment vegeation	2.80		0.51	

[a] Total inventories were calculated for sediments and peats formed since 1860 on the basis of the total sediment area in the lake and total peat area in the catchment.

between individual cores (*30*); therefore, a single sediment core cannot truly represent the whole lake basin.

Mercury and lead inventories accumulated within the full lake sediment and catchment soil areas since 1860 were calculated on the basis of the areas represented by individual sediment and soil cores, respectively (Table 2). The soils in the Lochnagar catchment are peats with LOI values of up to 99%, and because of this high organic content, they have a high affinity for trace metals (*20, 31*). The percentage soil cover in the catchment is 38%, and these soils have accumulated a very high amount of Hg and Pb with respect to the amount stored in the lake sediments (Table 2; *27*).

Dry-mass sedimentation rates and the dry mass of sediments accumulated since 1860 in individual sediment cores are shown in Table 1. By using decadal interval inventory data, Hg and Pb fluxes to the whole lake sediment area in Lochnagar were calculated (Figure 3), and these give more accurate pictures of trace metal contamination trends than the trace metal profiles of individual cores. The Hg sediment flux profile shows that Hg flux starts to increase from about 8 μg m^{-2} y^{-1} in the 1880s. The rate increase in Hg flux is constant at about 1.5%/y (0.13 μg m^{-2} y^{-1}) until the 1950s, when there is a decline in the rate of increase through to the 1970s. It is then relatively stable to the present Hg sediment flux of 17.6 μg m^{-2} y^{-1}. This value is slightly higher than the Hg surface flux (16 μg m^{-2} y^{-1}) of a sediment core taken from an undisturbed lake in southeastern Hudson Bay (*32*) and higher than those in recent lake sediments from West Greenland (5$-$10 μg m^{-2} y^{-1}; *33*) and from the Canadian Arctic (5$-$8 μg m^{-2} y^{-1}; *34*). Indeed, these latter contemporary fluxes are similar to that of preindustrial levels at Lochnagar. The present Lochnagar Hg flux is more than twice that of the flux in the 1880s. Swain et al. (*35*) estimated that modern Hg inputs exceed preindustrial background levels by an average annual increase of about 2%, and this is in good agreement with the measured rate of increase in Lochnagar sediment flux between the 1880s and the 1950s.

In general, global Hg burdens have not declined (*36*), although global Hg production increased from the 1910s to the 1970s and then decreased by the 1990s (*24*). However, local decreases have been seen in some areas due to the reduction of emissions from regional sources (*6, 24, 37*). Coal consumption is one of the main sources for Hg emissions, and coal consumption in the U.K. continued to increase until the late 1970s (*38*). Since this time, consumption has remained reasonably steady at about 80 million tonnes in England and Wales, while coal consumption in Scotland decreased from about 8 million tonnes in the late 1970s to about 3 million tonnes in 1985 (*22, 38*). These figures parallel the increase in the Lochnagar Hg sediment flux profile until the 1970s. From 1970 to 1998, Hg emission in the U.K. was reduced from 47.2 to 12.4 tonnes (*39*), and while Hg emissions from coal combustion to the atmosphere in Western Europe were stable from 1983 to 1992 (*40*), those from other industrial sources have decreased greatly over the past decade. Some Swedish remote lakes have recorded the decline in Hg

deposition in the surface sediments (*41*). However, the Lochnagar Hg sediment flux profile does not show this reducing signal.

The Pb sediment flux profile shows a slow, steady increase from 12.8 to 13.8 mg m^{-2} y^{-1} over the period between the 1860s and the 1880s followed by a rapid increase to about 21 mg m^{-2} y^{-1} in the 1940s. Since then, Pb sediment flux has fluctuated at about 20 mg m^{-2} y^{-1}. Global anthropogenic Pb emissions have been greatly reduced since the 1970s (*42*), and this reduction of Pb in deposition has been recorded in ice cores (*43*), ombrotrophic bog peat cores (*44, 45*), forest soils (*46*), and lake sediments (*5, 47$-$49*). However, similarly to Hg, Lochnagar sediments do not record this reduction in Pb.

Mercury and Pb Budget and Restoration. The annual bulk depositions of Hg and Pb, from October 8, 1997 to October 7, 1998, are 35.9 and 1970 μg m^{-2} y^{-1}, respectively. During this time, the precipitation in Lochnagar was 1655 mm; hence, the yearly Hg bulk deposition load is higher than those in other reported sites (*14, 50, 51*).

There are two ways for Hg to be lost from Lochnagar. One is through the outflow, and the other is via volatilization from the catchment soil and lake surface. Pb loss is only via the outflow stream. In 1998, water lost via the outflow was estimated at 1.04 $\pm$ 0.10 $\times$ 10^6 m^3, and the resulting mass of Hg and Pb lost via the outflow was estimated to be 15.2 $\pm$ 1.5 and 940 $\pm$ 94 g, respectively (Table 3). There are few reliable measurements of the volatilization rate of Hg from terrestrial and aquatic systems, as this depends on many parameters such as surface type, temperature, and geographical region (*36*). Currently, it is not possible to include evasion, emission, and re-emission of Hg from the catchment peats and lake water into the budget model.

There is no data for Hg and Pb related to groundwater, weathering of minerals, or leaching at Lochnagar. As the Hg and Pb sediment flux profiles show stability over the last 20 years, Hg and Pb inventories in the lake water are assumed to have been stable over the same period. The biomass of aquatic plants, zooplankton, and fish is low, and Hg and Pb storage in these organisms is assumed to be negligible. Therefore, the model for the annual Hg and Pb budgets for the whole catchment, including the lake for 1998, can be simplified as shown in parts A and B of Figure 4.

In the Hg budget (Figure 4A), 3.52 $\pm$ 0.35 g of Hg were deposited directly onto the lake, while 15.2 $\pm$ 1.5 g of Hg were lost via the outflow, although this loss is actually from the whole catchment. Increased storage of Hg in lake water was negligible as the Hg sedimentary flux was stable, and 1.07 g of Hg were deposited to the sediments. Therefore, according to the mass balance 12.8 $\pm$ 1.8 g of Hg must have been transported from the catchment into the lake. In the catchment, 32.96 $\pm$ 3.3 g of Hg were deposited. Assuming that increased storage of Hg in the catchment vegetation was also negligible in 1998, as Hg concentrations in the samples collected in 1997 and 1998 for the same vegetation species were similar (*20*), the storage of "new" deposited Hg in the catchment soils in 1998 was calculated to be 20.2 $\pm$ 5.1 g.

Since 1860, the anthropogenic fraction of Hg deposited at Lochnagar has increased, as has the amount of Hg transported from the catchment into the lake (*20*). The Hg budget for 1998 shows that the amount of Hg washed into the lake is 3.6 times that directly deposited onto the lake surface. This demonstrates that the catchment is currently the most important source of Hg to the lake. Mercury in deposition that falls onto bare rock may not be totally trapped by the catchment peats; hence, deposition to the catchment may also be a source of Hg for the lake. Lochnagar catchment peats have accumulated Hg (*27*) and these have now become severely eroded, resulting in Hg being transported into the

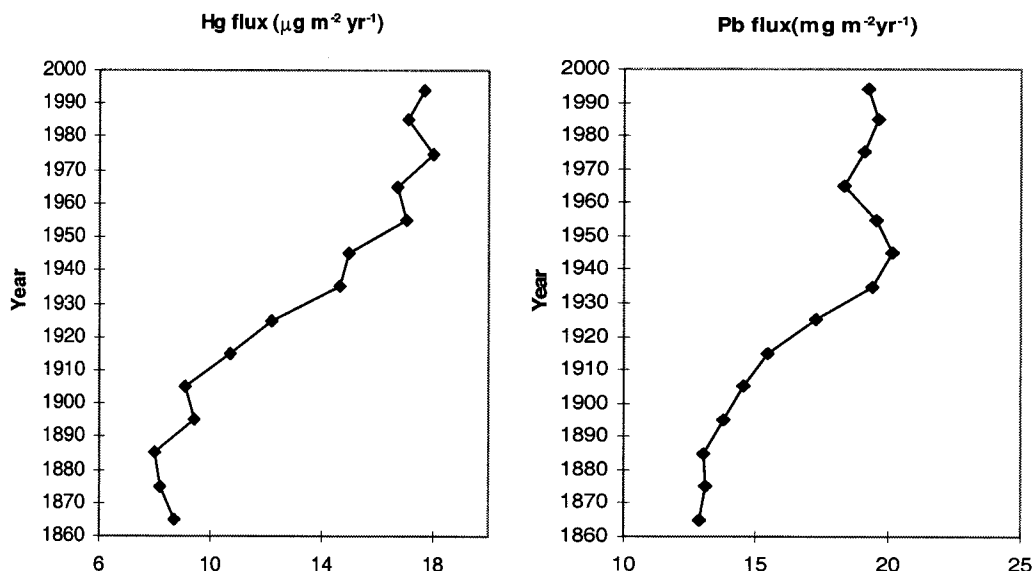**Hg flux (μg m⁻² yr⁻¹)** — **Pb flux(mg m⁻²yr⁻¹)**

FIGURE 3. Mercury and lead accumulation flux profiles for the whole sediment area based on 17 cores taken from the whole lake sediment area of Lochnagar.

TABLE 3. Amount of Water, Pb, and Hg lost via the Outflow in 1998[a]

time	Jan	Feb	March	April	May	June	July	Aug	Sept	Oct	Nov	Dec	total
water (m^3)	83000	44300	43200	135000	28200	50200	70900	51100	83900	257000	100000	90500	1040000
Pb (μg L^{-1})	0.39	0.60	1.04	0.50	0.79	1.05	0.56	0.61	1.13	1.32	0.91	1.09	
Pb mass (g)	32.4	26.5	44.9	66.8	22.3	52.7	39.7	31.2	94.4	339.2	90.5	98.6	940
Hg (ng L^{-1})	16.5	19.2	14.1	13.5	15.4	4.9	13.3	14.7	15.8	15.8	13.1	15.8	
Hg mass (g)	1.37	0.85	0.61	1.82	0.43	0.25	0.94	0.75	1.33	4.06	1.31	1.43	15.2

[a] Concentration of Pb is a mean for the period.

lake. Therefore, a slight change in atmospheric Hg deposition will not significantly change the input of Hg to the lake; hence, the lake sediment record for Hg is not sensitive to recent changes in atmospheric Hg deposition.

The Pb budget for 1998 (Figure 4B) shows that the amount of Pb transported from the catchment into the lake is larger than the amount deposited on the catchment (i.e., the net storage in the terrestrial catchment for 1998 is negative). This implies that even if all of the new Pb atmospherically deposited in the catchment in 1998 was transported into the lake, about 100 g of "old" Pb stored in the catchment (deposited before 1998) would still need to be moved into the lake to balance the budget. This also implies that the proportion of trace metals supplied from direct deposition and the catchment to the lake has changed and that even if the Pb deposition load has been reduced, the Pb stored in recent years in the sediments has not been reduced to the same level. Although errors in the estimates of Pb in the outflow may eliminate the need for loss of old Pb from catchment soils in the 1998 budget, Pb concentration data for deposition and lake water samples in 2000 indirectly confirm the importance of Pb leaching and erosion from the catchment. Lead concentrations in deposition samples in 2000 have been greatly reduced as compared with those in deposition samples in 1998 and are even lower than those in lake water taken at the same time. However, the mean Pb concentration in lake water samples taken in 2000 is still at the same level as that in the lake water samples taken in 1998. This implies that Pb from deposition to the catchment in 2000 was greatly reduced as compared to 1998 but that transport of Pb from the catchment to the lake is likely to have remained at a similar level. Therefore, more old Pb is likely to have been transported into the lake. According to the mass-balance, the catchment contributed 78% of the Hg and 91% of the Pb input to the lake in 1998.
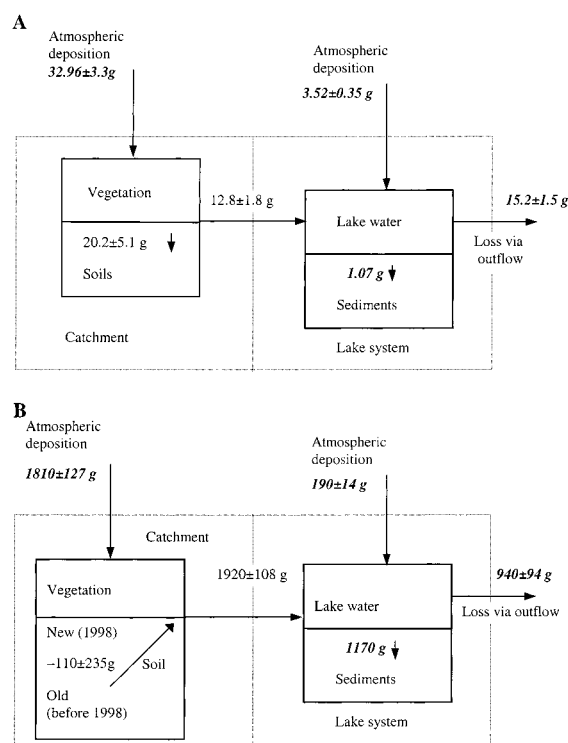

FIGURE 4. Annual total (A) Hg and (B) Pb budgets in Lochnagar for 1998. Figures in parentheses show dates. The boldface and italicized data are derived from measurements; the others are derived from the budget model.

The amount of Pb deposited in the Lochnagar catchment from anthropogenic sources in the last 140 years is more

than 839 kg (Table 2). This is more than 400 times the atmospheric deposition of Pb in 1998 and suggests that Pb deposition has been greatly reduced. Although the atmospheric deposition load of Pb has been reduced, Pb transported into the lake from the catchment has obscured any observable decline in the sediment record, and this results in errors in establishing the historical atmospheric load using Lochnagar lake sediments.

The store of past deposited Hg and Pb within the catchment has implications for continued Hg and Pb inputs to the lake as a result of continued or enhanced catchment soil erosion, despite any future reduction in atmospheric deposition. Therefore, the catchment could remain a major source of Hg and Pb to the lake for many years and will delay the restoration of the lake (52).

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