



Historical and contemporary metal budgets for a boreal shield lake



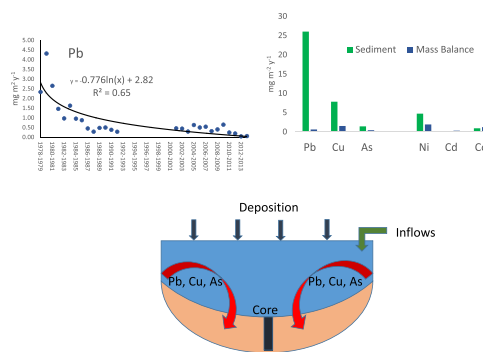
Shaun A. Watmough

Trent School of the Environment, Trent University, Peterborough, ON, Canada

HIGHLIGHTS

- 20 to 97% of metal inputs to the lake are from the terrestrial catchment.
- Surface sediment is enriched in Pb, Cu, Ni, As and Cd after 1900.
- Sediment fluxes for Ni, Cd and Co are comparable to lake mass balances.
- Sediment fluxes of Pb, As and Cu are up to 40 times greater than lake mass balances.
- Discrepancy likely due to sediment focusing

GRAPHICAL ABSTRACT



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ABSTRACT

Metal concentrations in sediment cores are widely used to reconstruct metal deposition histories, but rarely have metal budgets based on measured inputs (atmospheric deposition and inflows) and lake outflows been compared with metal fluxes estimated from lake sediment cores. In this study, budgets for six metals (As, Cd, Co, Cu, Ni and Pb) were estimated by measuring inputs in bulk deposition, inputs in the major inflow and export in the lake outflow for one hydrologic year (2002–2003) at Plastic Lake, Ontario, Canada. Inputs in bulk deposition were between $0.03 \text{ mg m}^{-2} \text{ y}^{-1}$ (Co) and $0.69 \text{ mg m}^{-2} \text{ y}^{-1}$ (Cu), which represented between 2.5 and 80.7% of total metal inputs to the lake. The estimated budgets for Co, Cd and Ni, which exhibit similar geochemical behavior in the major inflow, were comparable to budgets estimated from the upper section of a sediment core taken in 2002, taking into account previously published correction factors for sediment focusing. For example, mass budgets for Co, Cd and Ni were 1.24 , 0.28 and $1.89 \text{ mg m}^{-2} \text{ y}^{-1}$, compared with sediment budgets estimated to be 0.90 , 0.19 and $4.72 \text{ mg m}^{-2} \text{ y}^{-1}$, respectively. In contrast, measured budgets for As, Cu and Pb, which also behave similarly in inflows (and different to Co, Cd and Ni), were between 3 (As) and 40 times lower than estimates from the upper sediment core. A possible explanation for the discrepancy is that sediment focusing transfers sediment from shallow to deep areas, which for metals like Pb, which have strong affinities for organic matter and where atmospheric deposition has decreased 15 fold since 1978 ($4.31 \text{ mg m}^{-2} \text{ y}^{-1}$ in 1978 to $0.28 \text{ mg m}^{-2} \text{ y}^{-1}$ in 2013), leads to the transfer and accumulation of pollution metals to deeper parts of the lake long after a decrease in atmospheric deposition.

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1. Introduction

Metal concentrations have been measured in lake sediment cores to reconstruct changes in atmospheric metal deposition (Miller et al.,

E-mail address: swatmough@trentu.ca.

2014; Renberg et al., 2002; Graney et al., 1995). Studies have been conducted around the globe and range from lakes situated close to point source emissions (Schindler and Kamber, 2013) to lakes in remote locations that are impacted solely by long-range atmospheric deposition (Bilali et al., 2002; Miller et al., 2014; Yang et al., 2002). Most published reports have documented increases in atmospheric metal deposition associated with the onset of the industrial revolution or a specific industrial activity (Miller et al., 2014; Skierszkan et al., 2013; Renberg et al., 2002; Yang et al., 2002), although some studies have also documented increases in metal deposition occurring at various intervals over the past several thousand years ago associated with historical events such as metal smelting in the medieval times or with the expanse of the Roman Empire (Brännvall et al., 1999, 2001; Renberg et al., 2002). There have been fewer studies that have commented on decreases in lake sediment metal concentrations in response to recent atmospheric emission reductions, especially with respect to lead (Pb), where atmospheric deposition in many parts of the world has decreased dramatically (Galloway et al., 1982). In several cases decreases in metal levels in upper sediment have been noted (e.g. Graney et al., 1995; Kober et al., 1999), but the magnitude of the decrease is not usually compared with the magnitude of change in atmospheric metal deposition.

Most sediment studies report on changes in metal concentration in a single (or a few) cores and assume that these changes are primarily occurring in response to changes in atmospheric deposition and that deposition histories are preserved in sediment (i.e. watershed inputs are low/negligible and metals are relatively immobile in sediments) (Gallon et al., 2004). Some authors however, have constructed metal budgets by sampling many cores, which can take into account differences in sedimentation occurring at different locations within a lake (Blais and Kalff, 1995; Rippey et al., 2008). Likens and Davis (1975) first coined the term “sediment focusing” to describe process by which fine shallow sediments are gradually transferred to deeper parts of the lake leading to greater sediment accumulation in deeper parts of the lake (Crusius and Anderson, 1995). A number of studies in central Ontario have constructed lake metal budgets from sediment cores by sampling sediment in softwater Precambrian Shield lakes (Blais and Kalff, 1995; Dillon and Evans, 1982a, 1982b; Evans et al., 1983). These studies, which focused mostly on Pb and were based on 34–92 sites per lake showed that Pb fluxes were greatest in deeper parts of the lake and that metal accumulation rates increased with lake depth. This predictable relationship allowed for the development of correction factors that take into account sediment focusing for lakes in the region (Dillon and Evans, 1982a, 1982b).

Despite the wide use of sediment cores for reconstructing metal deposition histories, there have been few, if any, studies that have compared lake metal budgets based on measured inputs (atmospheric and stream inflows) and outputs (lake outflow) to metal budgets estimated from lake sediment cores. In this study, data from a detailed metal biogeochemistry study that was conducted in the early 2000s at Plastic Lake Ontario (Landre et al., 2009, 2010), along with long-term monitoring of atmospheric Pb, Cu and Ni bulk deposition since the late 1970s and data from a sediment core collected in 2002 were used to estimate lake budgets for Pb, Cu, Ni, Cd, Co and As. Specifically, the objectives were to 1) determine the relative contribution of atmospheric deposition compared with the terrestrial catchment for the 6 metals; and 2) compare metal budgets based on measured inputs and outputs with metal sediment fluxes estimated from a sediment core at Plastic Lake.

2. Methods

2.1. Study area

The Plastic Lake catchment is located in Haliburton County, Ontario (45°11'N, 78°50'W), about 20 km south of Dorset, Ontario, Canada. The site is accessed by an unpaved road and is approximately 200 km south-east of Sudbury, a major Cu-Ni mining and smelting region and

about 150 km north of the Greater Toronto Area and so receives pollutant inputs primarily from long-range transport as opposed to local sources. Hydrological and chemical monitoring has been ongoing for almost four decades at this site. The mean annual temperature for the Dorset area is 4.8 °C; mean monthly temperatures range from a high of 18.7 °C in July to a low of –11.1 °C in January (Environment Canada 2002; 30-year average 1971–2000). The mean annual precipitation is 1100 mm, of which approximately 75% falls as rain. The 32 ha headwater lake is fed by one major stream Plastic Lake-1 (PC1) and 6 ephemeral streams, which together drain an area of approximately 90 ha. Plastic Lake has a shoreline length of 2.92 km, a flushing time of approximately 3 years, mean depth of 8.0 m, with a maximum depth of 16 m that accounts for 2.5 ha of the lake area (Fig. 1: Dillon and Evans, 1982a). PC1 is the largest sub-catchment (23.3 ha) in the watershed, and is characterized by thin (average depth 0.37 m to bedrock) orthic humo-ferric and ferro-humic podzols (Soil Classification Working Group, 1998), formed from thin, sandy basal till (Fig. 1). The forest at Plastic Lake is dominated by white pine (*Pinus strobus*), eastern hemlock (*Tsuga canadensis*), red oak (*Quercus rubra*), and red maple (*Acer rubrum*) in the upland areas, and by white cedar (*Thuja occidentalis*) and black spruce (*Picea mariana*) in the swamp (Watmough and Dillon, 2004). A 2.2 ha conifer-*Sphagnum* swamp is located approximately 50 m above the catchment outflow in PC1 and >85% of the runoff from the Plastic Lake catchment drains through the swamp before discharging to the lake (Fig. 1).

2.2. Field sampling (2002–2003)

Field sampling during 2002–2003 has been described in detail elsewhere (Landre et al., 2009, 2010; Watmough and Dillon, 2004). Briefly, bulk deposition was collected in a small clearing located adjacent to the PC1 sub-catchment using an open container fitted with a Teflon-lined funnel (diameter 25 cm) covered with 80 µm Nitex mesh to prevent contamination from debris. Samples were collected for chemical analysis every 1 to 2 weeks. Water level or stage at PC1 and PC0 (lake outflow) was recorded continuously at weirs installed at each outflow, and daily stream discharge (m³ day⁻¹) was computed using established stage-discharge relationships for all outflows. Water samples for chemical analyses were collected at the weir at regular intervals (at least bi-weekly) when there was flow, although sampling was more frequent (2–3 times per week) during the spring melt period (March–April). During sample collection, all water samples were filtered using a coarse filter (80 µm). All metal samples were acidified to pH 2 with ultra-pure HNO₃ (Optima, Fisher Scientific) prior to analysis. Samples were analyzed for 19 metals, but only Pb, Cu, Ni, Cd, Co and As are reported here, using a Thermo-Finnigan Element II high resolution single collector inductively coupled plasma mass spectrometer (HR-ICP-MS). An internal standard (5 ppb Rh) was used to correct for instrument drift. Recoveries were determined using the standard reference material Trace Metals in Natural Waters (NIST-1640), and each of the reported certified metals typically had a recovery within +15% of the stated concentration. Method detection limits (MDLs) were calculated using the mean plus three times the standard deviation of the blank concentrations for each metal. All 6 metals were above the MDLs for all samples. A portion of the samples was also passed through a 0.45 µm filter to determine particulate versus “dissolved” concentrations of metals and concentrations were not statistically different (Landre et al., 2009). Long-term (1978–2014) bulk deposition data was collected at the same deposition site and analyzed for Pb, Cu and Ni using methods reported by the Ontario Ministry of Environment and Climate Change (OMOEC, 1983).

2.3. Lake sediment core

Details of physical and chemical properties of the lake can be found in Dillon et al. (1987). Sediment core samples in this study were

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