Science of the Total Environment 409 (2011) 548–563

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Comparison of mercury and zinc profiles in peat and lake sediment archives with historical changes in emissions from the Flin Flon metal smelter, Manitoba, Canada

P.M. Outridge [a,*], N. Rausch [b,c], J.B. Percival [a], W. Shotyk [b], R. McNeely [a]

[a] Geological Survey of Canada, Natural Resources Canada, 601 Booth St., Ottawa K1A 0E8, Canada
[b] Institute of Earth Sciences, University of Heidelberg, Im Neuenheimer Feld 236, D-69120 Heidelberg, Germany
[c] European Commission Joint Research Center, Institute for Transuranium Elements, P.O. Box 2340, D-76125 Karlsruhe, Germany

ARTICLE INFO

Article history:
Received 18 May 2010
Received in revised form 21 October 2010
Accepted 22 October 2010
Available online 20 November 2010

Keywords:
Mercury
Zinc
Smelter
Lake sediments
Peat
Flin Flon

ABSTRACT

The copper–zinc smelter at Flin Flon, Manitoba, was historically the largest single Hg point-source in Canada, as well as a major source of Zn. Although emissions were reported by industry to have declined significantly since the late 1980s, these reductions have never been independently verified. Here, the histories of Hg and Zn deposition over the past century or more were determined at five lake sediment and three peat study sites in the surrounding region. At sites spanning the range from heavy to minor pollution, lake sediment Hg and Zn concentration and flux profiles increased significantly in the early 1930s after the smelter opened. Two of the three peat archives were wholly or partially compromised by either physical disturbances or biogeochemical transitions which reduced their effectiveness as atmospheric metal deposition recorders. But the remaining peat records, including a detailed recent 20 yr record at a moderately polluted site, appeared to show that substantive reductions in metal levels had occurred after the late 1980s, coincident with the reported emission reductions. However, the lake sediment results, taken at face value, contradicted the peat results in that no major declines in metal concentrations or fluxes occurred over recent decades. Mercury and Zn fluxes have in fact increased substantially since 1988 in most lakes. We suggest that this discrepancy may be explained by catchment soil saturation by historically deposited metals which are now mobilizing and leaching into lakes, as has been reported from other smelter polluted systems in Canada, whereas the upper sections of the peat cores reflected recent declines in atmospheric deposition. However, further research including instrumented wet and dry deposition measurements and catchment/lake mass balance studies is recommended to test this hypothesis, and to provide definitive data on current atmospheric metal deposition rates in the area.

1. Introduction

Mercury (Hg) in the environment continues to be a major focus of international scientific and policy concern because of its intrinsic toxicity and its propensity to bioaccumulate and biomagnify in the human food chain. In Canada, a site of national concern from a Hg emission perspective is the copper–zinc smelter at Flin Flon, Manitoba. It is believed that the smelter, which opened in 1931, was historically the largest single source of Hg air pollution in Canada, and remained the largest emitter in 2000 despite substantial reductions during the 1990s (CCME, 2000). A study of soil humus metal concentrations around Flin Flon found an anthropogenic contribution of 95–100% of total humus Hg within 10 km of the smelter, and 36% at 50–85 km distance; background Hg levels in soil humus were attained approximately 85 to 105 km down-wind (Henderson et al., 1998; McMartin et al., 1999; Sim et al., 1999). The amount of Hg emitted prior to the 1990s is somewhat uncertain. McMartin et al. (1999) reported that 583 t of particulate Hg was released between the smelter opening in 1931 and 1995, based on industry-supplied estimates. But more recently, extrapolation of the assumed constant rate of Hg release of 19.9 t/yr prior to 1993 (Nilsen, 2003), suggests that >1000 t of particulate Hg may have been emitted since the start of operations. The smelter is also a major source of Zn and other metals. Although not of the same concern from a policy perspective in Canada, the amount of Zn emitted (total of 120,900 t from 1931 to 1995) makes it the largest single pollutant metal at the smelter, representing 26.4% of historical dust emissions (Nilsen, 2003).

Based on data submitted by Hudson Bay Mining and Smelting Co. (HBMS) to Environment Canada, metal emissions to the atmosphere from the Flin Flon smelter declined sharply in the late 1980s and early 1990s because of technology improvements. The reductions in Hg amounted to about 90% or 18 t/yr, from 19.9 t/yr prior to 1993 to an average of 1.4 ± 0.2 t/yr in 1995–2002 (particulate Hg only; Nilsen, 2003, Table 4.2; CCME, 2000). As these emission figures were based on sampling of electrostatic precipitator dust only, gaseous elemental Hg

* Corresponding author. Tel.: +1 613 996 3958.
E-mail address: outridge@nrcan.gc.ca (P.M. Outridge).

0048-9697/$ – see front matter. Crown Copyright © 2010 Published by Elsevier B.V. All rights reserved.
doi:10.1016/j.scitotenv.2010.10.041

(GEM) releases to the atmosphere were not monitored (Nilsen, 2003). The reduced emissions still constituted half of the 2.8 t/yr emitted in 2000 by the entire Canadian base metal smelting sector, and ~12% of the 12 t/yr emitted into the air by all human activities in Canada (CCME, 2000). The reported decrease at Flin Flon significantly influenced the national Hg emission inventory. Environment Canada reported that "between 1990 and 1995, Canadian anthropogenic Hg emissions dropped from approximately 35 to 11 tonnes [per year] primarily as a result of process improvements in the base metal mining and smelting industry." (Environment Canada, 2004). It can be deduced, therefore, that the ~18 t/yr decrease reported from the Flin Flon smelter accounted for most of the 24 t/yr national reduction between 1990 and 1995. Zinc emissions from Flin Flon also declined substantially in the late 1980s, amounting to an ~80% reduction from 1575 t/yr in 1989 to between 60 and 470 t/yr in 1995–2002 (Nilsen, 2003).

Surprisingly, however, these reported reductions have never been independently verified, and the historical and recent rates of atmospheric metal flux in this area have never been quantified. Metals in precipitation were measured over one year in the 1970s, but did not include Hg (Franzin et al., 1979). Interpretation of an early lake sediment core study (Harrison et al., 1989; Harrison and Klaverkamp, 1990) was hampered by the absence of sediment chronologies and sedimentation rates with which to calculate metal fluxes. A reliable reconstruction of the history of Hg and Zn deposition around the smelter is needed, first, to reveal the impact of the smelter on deposition rates in the surrounding environment, and second, as an independent verification of the effectiveness of emission control measures that were implemented beginning in the late 1980s.

This study addressed these aims by using lake sediment and peat cores from the surrounding area to reconstruct changes in regional atmospheric Hg and Zn deposition from the mid- to late 19th century to the early 2000s, and by comparing the changes over recent decades against the emissions reductions reported by industry. We determined the down-core trends of Hg and Zn concentrations and fluxes in sediment cores from five sites in four lakes, and three peat cores from peat bogs within two of the lake catchments. This combined approach using spatially clustered sediment and peat archives is uncommon (although see Lamborg et al., 2002), but it was used here because each type of archive presents advantages and disadvantages with respect to reconstructing metal deposition histories (Shotyk, 1996; Biester et al., 2007).

2. Materials and methods

2.1. Area description

The area of this study covers about a 0.5° latitude by 1.5° longitude area around Flin Flon, Manitoba (~54.5°N, 101.8°W; Fig. 1) which was established in 1930 as a service town (currently ~7500 people) for the local mines and smelter. The annual average temperature is −0.2 °C, ranging from a mean of 17.8 °C in July to −21.4 °C in January (Canadian Climate Normals, 1971–2000; Flin Flon "A" station). Annual precipitation is about 470 mm, with 30% falling as snow. Winds are predominantly from the northwest from September to February, but tend to be most often southerly from March to August. The surrounding landscape is boreal forest and lakes. Occurring as it does on the Precambrian Shield margin, the area is geologically complex, being underlain by sandy-clay tills derived from Precambrian greenstone-granites and gneisses in the northern part as well as Mesozoic sedimentary and Paleozoic carbonate rocks in the south (McMartin et al., 1996; Henderson et al., 1998). The area was glaciated by ice flowing from the north and northeast, and after ice retreat was entirely inundated by proglacial Lake Agassiz.

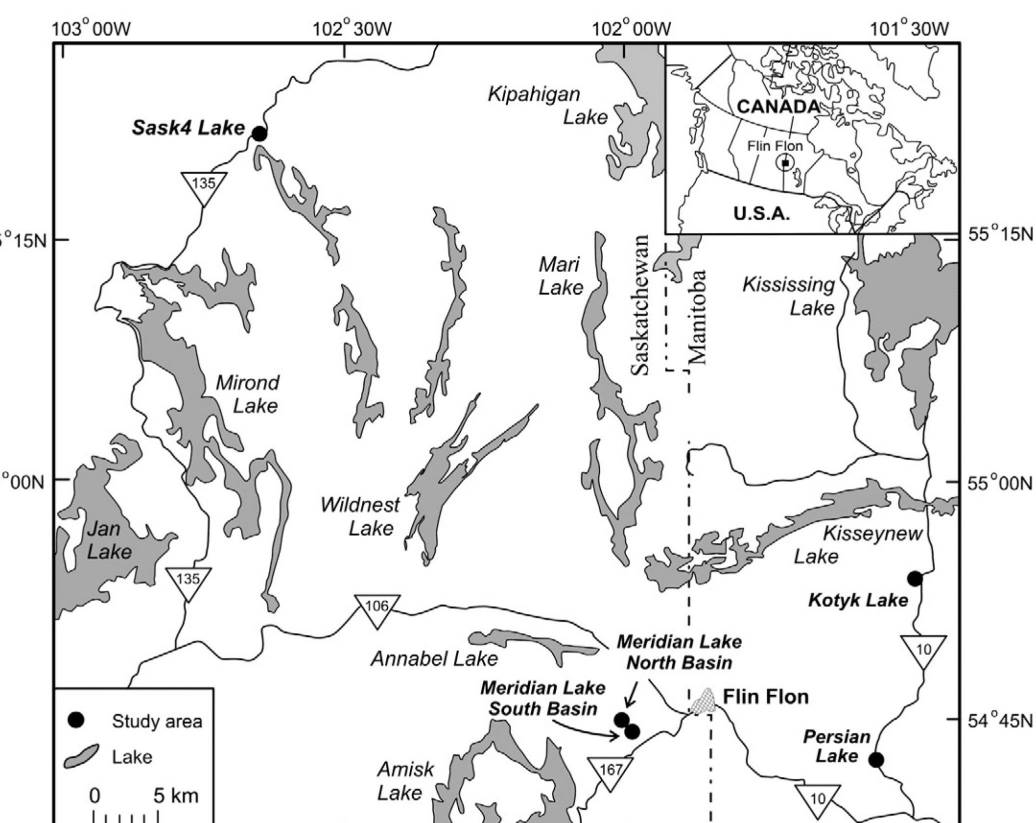

Fig. 1. Location map of the study sites near Flin Flon, Manitoba. (NAD83, UTM Zone 14 projection).