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An inventory of historical mercury emissions in Maritime Canada: implications for present and future contamination

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Abstract

Mercury is a longstanding concern in Maritime Canada due to high levels of contamination in a number of fish and bird species. The recycled component of past releases of anthropogenic mercury may be a significant source of ongoing pollution in many areas. Historical information on mercury releases can be used to quantify past and present anthropogenic contamination. We present an inventory of historical mercury emissions from anthropogenic sources in Maritime Canada for the years 1800–1995. Long-term trends in mercury emissions and the significance of the cumulative burden of mercury released from local sources are discussed. Emissions are calculated using both historical monitoring data and the application of emission factors. The nature of current anthropogenic sources of mercury is quite different than it was several decades ago when many of the existing policies governing mercury pollution were created. Our inventory illustrates that many of the most significant sources in the past such as the chlor-alkali industry, paint containing mercury additives, and pharmaceuticals, have been largely phased out with fossil fuel combustion and waste disposal remaining as the most significant modern sources. Atmospheric emissions in Maritime Canada peaked in 1945 (> 1750 kg year^{-1}), and again between 1965 and 1970 (> 2600 kg year^{-1}). Cumulative releases of mercury from anthropogenic sources for the years 1800–1995 were between 115 and 259 t to the atmosphere alone, and 327–448 t when discharges to wastewater and effluents were included. Assuming that only 0.2% (Nriagu, 1994.) of these releases become part of the recycled fraction of current fluxes, we estimate that between 570 and 900 kg Hg year^{-1} is deposited in Maritime Canada from past anthropogenic sources. Modern sources within Maritime Canada contribute at least 405 kg year^{-1} to the total annual deposition of 1.71 t over the

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provinces of New Brunswick, Nova Scotia and Prince Edward Island, leaving ~ 735 kg year^{-1} from natural sources and long-range contamination. Further study is needed to verify these estimates and clarify the significance of natural and long-range sources of mercury in Maritime Canada.

Keywords: Anthropogenic pollution; Cumulative releases; Modern fluxes; Fossil fuels; Population-based sources; Chlor-alkali; Industry; Gold mining

1. Introduction

Mercury contamination is a well known problem in most industrialized countries. In Sweden, for example, an estimated 10 300 lakes contain fish with mercury concentrations above the 'blacklisting' threshold of 1 mg kg^{-1} (Hakanson et al., 1990). In the United States, between 1 and 3% of women of child-bearing age are exposed to potentially harmful levels of mercury through consumption of fish (USEPA, 1997a). In 1998, 39 States and five Canadian Provinces, including all of the North-eastern United States and the three Maritime Canadian Provinces, put in place consumption advisories due to high levels of mercury in fish (NESCAUM et al., 1998).

Mercury has been a longstanding concern in Maritime Canada since the 1970s when concentrations > 1 ppm were noted in the muscle tissues of harbor porpoises and seals in the Bay of Fundy (Gaskin et al., 1973, 1979). Maritime Canada consists of the Eastern Canadian Provinces of New Brunswick, Nova Scotia, and Prince Edward Island. This area has a predominantly rural population base of ~ 1.8 million residents, and is directly adjacent to the north-eastern United States. Currently, high levels of mercury in fish, exceeding the human health consumption limit, are negatively impacting the viability of regionally important recreational and commercial fisheries (Peterson et al., 1989; d'Entremont et al., 1998; NESCAUM et al., 1998). In addition, mercury levels in the common loon that are among the highest in North America, are raising widespread concerns that the long-term viability of this species is being threatened (Burgess et al., 1996). These issues make accurate documentation and regulation of sources of mercury in this region particularly important.

There is limited understanding of the link between emissions of mercury and accumulation in the food web. However, in the latest Mercury Study Report to Congress, the United States Environmental Protection Agency acknowledged a 'plausible link' between anthropogenic emissions of mercury and accumulation in fish (USEPA, 1997a). The relationship between anthropogenic mercury releases and resulting concentrations in the environment is confounded by the fact that mercury is a naturally occurring element and is ubiquitous in the environment (Nriagu, 1979). In addition, the propensity of this element for long-range transport and deposition can result in contamination of regions far from the point sources of pollution (Jackson, 1997; Fitzgerald et al., 1998; Hermanson, 1998). Mercury released into the atmosphere as a result of human activity becomes integrated with the natural pool of mercury in the environment and may then be continuously deposited and re-emitted, resulting in an ongoing legacy of mercury contamination (Nriagu, 1993, 1994). The re-emission of past sources of anthropogenic mercury is likely to be a significant source of ongoing pollution in many areas, accounting for up to one-third of the current reservoir of mercury in the atmosphere (Pirrone et al., 1996). Thus, both current and historical releases need to be considered when attempting to understand current contamination problems. By quantifying the cumulative releases of anthropogenic mercury throughout history we may better understand how regulating existing sources of mercury will affect the overall burden of mercury in the atmosphere and the environment.

The purpose of this paper is to present an inventory of historical mercury emissions from anthropogenic sources in Maritime Canada for the years 1800–1995. This time period was chosen to reflect the effects of human settlement and industrialization on the amount of mercury released to the environment. In this paper we present the long-term trends in mercury emissions, and estimate the cumulative burden of mercury released in this region from historic and current anthropogenic sources. This information provides insight into sources of mercury that may be potential contamination problems in the future as well as those that require further study. In addition, using our estimates of the cumulative releases of mercury in Maritime Canada, we discuss the component of current fluxes of mercury that may be associated with the recycled component of past anthropogenic sources in this region.

One distinct aspect of our inventory is the combination of an historical perspective and a relatively small geographic scale. Such an approach allows insight into long-term trends in mercury contamination on a regional basis. There are few other historical inventories and those that do exist have mostly been compiled for larger regions (Nriagu, 1996; Pirrone et al., 1998). We feel that the geographic scale of our historical inventory complements continental and global scale studies by providing higher resolution information on areas of interest that can aid in interpretation of historical deposition profiles. For example, Pirrone et al. (1998) compiled historical inventories of anthropogenic mercury emissions in North America and the Great Lakes region, and compared historical emissions to deposition profiles in sedimentary records. The authors could not account for the observed sedimentary data using only the large-scale emission information and concluded that local discharges of mercury were most likely important determinants of the historical deposition profiles analyzed. Thus, local and regional scale information provided in studies such as ours, can be combined with larger continental and global scale inventories to help distinguish local, regional, and global contaminant signals in sediment core records. While a number of annual inventories have been compiled on comparable geographic scales to our study, they provide information on only a single year of interest and are not available on a consistent basis. In addition, there may be problems extrapolating and comparing data compiled in different time periods due to a lack of consistency in assumptions and methods between inventories, as discussed in a previous paper (Sunderland and Chmura, in press). The application of consistent and explicit methods over time is another advantage of the approach used in this study. We attempt to clearly illustrate our calculation methods and list all assumptions made in order to allow estimates to be refined as further information becomes available and to prevent misinterpretation of our results.

2. Methods used to calculate emissions

In this study, we divide all anthropogenic sources of mercury into three major categories: (1) energy; (2) population-based sources; and (3) industry. Where possible, we provide high and low estimates to represent the range in moderate emissions scenarios. The high emissions scenario represents the upper end of moderate emissions and includes those sources for which information was highly uncertain and little data was available. The low estimate is more conservative and is derived from the low end of emission factors reported in the literature. Because it was not possible to quantify the uncertainty in our estimates, we use this approach to represent the probable range of emissions that have occurred in the past and to avoid underestimation of actual emissions. However, there are several sources for which only one estimate was available due to limited data on past emissions and releases associated with a particular activity.

2.1. Energy

The category ‘energy’ describes mercury emissions resulting from combustion of fuels to produce energy and includes wood, coal and refined petroleum products. Emission factors were combined with historical statistics on quantities of fuels consumed in Maritime Canada to estimate mercury releases to the environment. Long-term trends in emissions from this category, calculation methods, and justification for emissions factors selected are reported in detail elsewhere (Sunderland and Chmura, in press). A summary of these methods is presented in the following section.

2.1.1. Wood

We assumed that the majority of wood burned for anthropogenic applications throughout history in Maritime Canada was for residential use. High and low estimates of emissions were based on emission factors of 50 and 13 g Hg/KT wood combusted, respectively [Sheffield, 1983; FIRE (V.51b), 1993].

2.1.2. Coal

Coal combustion was divided into electric power generation and residential use. High (0.19 ppm) and low (0.10 ppm) estimates of the mercury content of coal were selected to represent the range of values reported in the literature. We assumed that all mercury in coal is released to the atmosphere upon combustion if no emissions control devices are in place (Lindberg, 1987). By assuming residential coal combustion in Maritime Canada has taken place without the use of emissions control devices, we estimated high and low releases of 190 and 100 g Hg/KT coal combusted, respectively. Because most electric power-generating facilities have emission control devices in place, and the effectiveness of these instruments varies widely (USEPA, 1993), we assumed there would be a substantial difference between controlled and uncontrolled devices resulting in an overall reduction in mercury emissions of ~50%. Thus, we applied emission factors of 95 and 50 g Hg t^{-1} of coal used by electric power-generating facilities.

2.1.3. Refined petroleum products

The major categories of refined petroleum products are heavy and light fuel oils, diesel oil, and gasoline. The low estimate of emissions includes mercury released from the combustion of heavy fuel oil (HFO), light fuel oil (LFO), and diesel oil. Gasoline was not included in the low estimate because of the high degree of uncertainty regarding the magnitude of mercury released from motor vehicles (USEPA, 1997b). Emission factors for HFO (0.025 kg Hg ‘000 m^{-3}), LFO (0.050 kg Hg ‘000 m^{-3}), and diesel oil (0.38 kg Hg ‘000 m^{-3}) obtained from the FIRE database were used to provide a conservative estimate of emissions from these sources. The high estimate was generated following the method used by Environment Canada (Jaques, 1987; Environment Canada, 1973), which assumes that there is a ‘crude equivalent’ to the total amount of refined petroleum products consumed (e.g., all mercury contained in the crude oil eventually makes its way into refined petroleum products and is released upon combustion). The average mercury concentration in crude oil was estimated to be ~30 g Hg ‘000 m^{-3} oil (Jaques, 1987).

2.2. Population-based sources

The category ‘population-based’ sources describes releases of mercury from healthcare activities and the consumption and disposal of consumer goods. All of the sources in this category are partially contingent on the total demand for goods and services by the resident population and are, therefore, related to population size. These sources include hospitals (medical waste incineration, mercury thermometers), dental uses (amalgams, dental offices, crematoria), pharmaceuticals, paint application, electrical goods (fluorescent bulbs), and municipal solid waste (MSW) disposal. In some cases historical data on mercury use for these sources were not available for Maritime Canada and consumption data from the United States were extrapolated and applied to the demographics of Maritime Canada.

2.2.1. Hospitals

Direct releases of mercury from hospitals may result from the breakage of thermometers containing mercury and medical waste incineration. We have not attempted to comprehensively account for the use of mercury in the hospital sector in products such as sphygmomanometers, monometers, lab chemicals and other devices; rather we have targeted the major sources of mercury emissions. Based on a survey conducted by Environment Canada in the early 1970s (Environment Canada, 1973), we assumed that there were on average nine thermometers broken per hospital bed annually in that region. The number of active hospital beds in the region over the duration of our inventory was obtained from Statistics Canada (Statistics Canada, 1880–1997; Leary, 1983; Urquhart and Buckley, 1965). Atmospheric emissions from thermometer breakage were estimated using an emission factor of 0.2 g Hg per thermometer broken (Environment Canada, 1974; Cooke and Beitel, 1971). We assumed that the remaining mercury in thermometers would enter the waste stream with other mercury-containing goods that are used in hospitals. Medical waste generation in Maritime Canada was estimated by dividing current data by the number of active hospital beds in the region, and multiplying that figure by the number of active hospital beds between 1900 and 1995 (Resource Integration Systems Ltd., 1996; Doiron et al., 1998; Statistics Canada, 1880–1997). We selected high and low emission factors of 14 and 20 g Hg t^{-1} of medical waste incinerated in Maritime Canada (Sang and Lourie, 1997; USEPA, 1993). There is some error associated with the resulting estimates due to the changing composition of medical waste and mercury content over time such as the more widespread use of pharmaceuticals that contained mercury in the past. In addition, the number of active hospital beds is not always a good indicator of medical waste generation because it does not account for day surgeries, outpatients, laboratories and research uses. Hence, we expect that historical emissions in this category are underestimated but do provide an approximation of mercury losses from hospitals.

2.2.2. Dental uses

Mercury released as a result of abrasion and wear of mercury-amalgams, losses that occur during application in the dental office, and crematoria were considered in this category. An emission factor of 0.15 g Hg $year^{-1}$ per adult was used to estimate the mercury loss to sanitary services as the result of abrasion and wear of dental amalgams (Hawley and Sheridan, 1978). Cooke and Beitel (1971) estimated that approximately half of the material used during restorative procedures is flushed down the sink or evaporated into office air. Based on this study, we assumed that ~ 40% of the mercury used in dental offices in Maritime Canada was lost to municipal wastewater. Atmospheric emissions from dental offices were estimated using high and low emission factors of 20 and 8.5 kg Hg t^{-1} Hg consumed (Jaques, 1987; USEPA, 1993). Historical data on the number of dentists performing restorative procedures in Maritime Canada was obtained from the Canadian Dental Association (CDA) and Statistics Canada (Statistics Canada 1880–1997; Urquhart and Buckley, 1965, B. Keeping, CDA, 1998, Personal Communications; Leary, 1983) To estimate emissions from crematoria we used high and low emission factors of 1 and 0.32 g Hg per cremation (USEPA, 1993; Hogland, 1994). The number of cremations in Maritime Canada was estimated from the annual mortality rate in the region and the fraction of the deceased cremated (Statistics Canada 1880–1997; Statistics Canada, 1966, 1974).

2.2.3. Pharmaceuticals

It is widely assumed that a portion of the mercury in all pharmaceutical products, with the exception of diuretics, evaporates after application (Davis, 1971; Environment Canada, 1973; Jaques, 1987). We estimated atmospheric emissions from this source using high and low emission factors of 0.5 and 0.2 kg Hg kg^{-1} Hg used in pharmaceutical products other than diuretics. Because no data on the consumption of mercury in pharmaceuticals in Maritime Canada were available, we extrapolated data from the USA to generate a per capita consumption figure that could be applied to the demographics of Maritime Canada (Statistics Canada 1880–1997; MRBC, 1945–1995; USEPA, 1993, 1997b). We assumed that all of the mercury contained in these products that was not lost to the atmosphere would eventually be released to municipal wastewater. We were unable to find data on the amount of mercury used for pharmaceutical applications prior to 1945. However, Nriagu (1979) notes that peak use in the USA occurred in 1943. It is, therefore, likely that emissions from this source in Maritime Canada were also significant prior to 1945. This should be kept in mind when considering total mercury releases in Maritime Canada prior to 1945.

2.2.4. Paint application

Mercury added to paint is lost from painted surfaces through volatilization to the atmosphere or as an effluent from outdoor surfaces. High and low emission factors of 0.66 and 0.25 kg kg^{-1} Hg contained in paint were used to estimate emissions of mercury to the atmosphere (Sheffield, 1983; Jaques, 1987; USEPA, 1997b). We assumed that effluent losses would be the difference between measured releases from indoor and outdoor losses, which based on the results of past studies, are between 20 and 75% of the total mercury contained in paint (Taylor and Tickle, 1969a,b; Taylor and Hunter, 1972). These figures were used to represent high and low releases in effluents. Because no data on the quantity of mercury consumed in paint products in Maritime Canada are available, annual quantities of mercury consumed in paint per household in the USA were extrapolated to estimate consumption in Maritime Canada (Statistics Canada 1880–1997; MRBC, 1945–1995; D'Itri, 1972; Nriagu, 1979).

2.2.5. Electrical goods

With the exception of electric lamps containing mercury, the majority of mercury in electrical goods enters the solid waste stream. Hence, only losses from the breakage and disposal of electric lamps are considered in this category. Mercury losses from other electrical goods are included in the releases from municipal waste disposal. Because the majority of electric lamps containing mercury are fluorescent bulbs (NEMA, 1992), mercury emissions in this category are based on mercury losses from the breakage of fluorescent tubes. We estimated atmospheric emissions from this source by assuming 75% of the new lamps manufactured each year replaced broken ones and applied high and low emission factors of 1 and 0.275 kg mercury per kg of mercury contained in broken lamps (Jaques, 1987; USEPA, 1997b).

2.2.6. Municipal waste disposal

Emissions of mercury from municipal waste incinerators and landfills were both considered in this category. Emissions of mercury from municipal waste incinerators were estimated by combining data on the amount of waste generated in each of the Maritime Provinces and the estimated fraction of waste incinerated, with emission factors for mercury released during waste combustion. Provincial waste generation data were only available between 1965–1995 (Environment Canada, 1974; Resource Integration Systems Ltd., 1996; Doiron et al., 1998). Mercury emissions from incineration prior to 1965 have not been included in our inventory due to lack of information on the amount of mercury contained in waste and quantities of waste incinerated. Emission factors ranging from 1.5 to 4.79 g Hg t^{-1} of waste combusted were chosen to reflect varying quantities of mercury in waste and changes in emissions control technology over various time periods. Emissions from landfills were estimated to be 0.1 g Hg per person per year based on a study by Lindqvist et al. (1991). Landfills were only included in the high estimate of emissions because we did not account for variations in mercury over time or regional variations in types of waste and estimates are highly uncertain.

2.3. Industrial sources of mercury

This category includes chlor-alkali facilities, agricultural applications, pulp and paper plants, base–metal smelting, and gold mining operations. Both emission factors and point source data were used to estimate emissions in Maritime Canada. There are several additional industrial sources of mercury emissions for which we were unable to generate an estimate of emissions, the significance of which will be discussed below.

2.3.1. Chlor-alkali facilities

Chlor-alkali facilities produce chlorine, caustic soda, and hydrochloric acid and are often associated with pulp and paper operations. In the past, most of these facilities employed a ‘mercury cell process’ that resulted in the use and subsequent release of large quantities of mercury to wastewater, solid wastes, and the atmosphere. Two chlor-alkali facilities in Maritime Canada were included in this inventory. The first opened in Dalhousie, New Brunswick in 1963, and is the last remaining chlor-alkali facility in Canada that uses the mercury cell process. The second facility operated in Point Abercrombie, Nova Scotia, between 1970 and 1992.

Mercury releases from these two operations were obtained from plant records (Johnson, ICI Forest Products, personal communication, 1997; Curry, Canso Chemicals Inc., personal communication, 1997) and Environment Canada monitoring data (Buffa, 1973; Environment Canada, 1973; Marsh and Sullivan, 1976; Pascoe, 1976; Paine, 1994). It should be noted that our estimates of total releases from the facilities in Maritime Canada include mercury lost in solid wastes. In some other inventories that include these facilities (Sheffield, 1983; Jaques, 1987; Doiron et al., 1998) mercury in solid waste was not a component of losses considered and their reported releases may, therefore, appear smaller in magnitude.

2.3.2. Agricultural applications

Mercurial fungicide sprays and seed treatments were used extensively in Canada between 1940 and 1970 to control a variety of fungal and bacterial growths (Pest Management Information Service, 1998). Mercury was also used in foliar applications of fungicides to a variety of fruit and vegetable crops, and in turf control products for golf courses, lawns, shrubs and trees (Fimerite, 1970). All mercury in agricultural products was released directly into the environment and made its way into soil, groundwater, or was volatilized to the atmosphere (Gilmour and Miller, 1973). High levels of mercury in game birds and fish prompted the Canadian Department of Agriculture to ban the use of mercurial seed treatments in 1971 (Gurba, 1971). The last three pest control products that contained mercury as an active ingredient were discontinued on 31 December, 1995 (Chagnon, personal communication; Pest Management Information Service, 1998).

We were unable to find suitable historical data on the use of pest control products containing mercury in Maritime Canada and instead relied on comparable national and regional studies to estimate historical usage levels. For the period between 1948 and 1978 we used national statistics on pest control product sales in Canada to approximate the quantities of agricultural chemicals containing mercury consumed (Statistics Canada, 1948–1978). These data were unavailable in Canada for the years prior to 1945 and post-1978, due to industry confidentiality. For these years we used comparable USA consumption data (data from: MRBC, 1945–1995; D’Itri, 1972; Nriagu, 1979). We were unable to find any statistical information on use of these products before 1945. However, between 1945 and 1950 there was rapid growth in the use of mercury in these products in the USA. Based on the trajectory of consumption after 1945, we can postulate that releases of mercury from the application of agricultural chemicals in Maritime Canada were not significant before this time.

Emissions of mercury resulting from the application of agricultural chemicals were estimated using the average concentrations of mercury in seed treatments (1.5%) and fungicide sprays (50%) reported by Environment Canada (1973). Since the use of agricultural chemicals containing mercury implies a direct release of mercury into the environment, we assumed all mercury in these products was lost upon application. We chose to compile only one estimate of emissions from this source due to the limited availability of data on the past use of mercury in these products.

2.3.3. Pulp and paper plants

Slimicides containing mercury were used in the Canadian pulp and paper industry between 1940 and 1970 (Sherbin, 1979). Mercury is lost in final products and effluents leaving the mill (Paavila, 1971). There is no published data on quantities of mercury used in slime control products by pulp and paper mills in Canada or the Maritime region (Li, Pulp and Paper Related Industrial Sectors, Environment Canada, 1998, personal communication). A survey conducted by the Pulp and Paper Technical Association of Canada (PPTAC) in 1953 (Patte, 1954) confirmed that 30 out of 46 ($\sim 65\%$) of the mills surveyed used organic mercurial slimicides. In the USA, consumption of mercury for pulp and paper applications was most significant between 1955 and 1965 (MRBC, 1945–1995) and a survey conducted by Environment Canada (1973) indicated that use of mercury for this purpose in Canada had been phased out by 1970. Therefore, we assumed that use of these products was most significant between the years 1955 and 1965.

Seven pulp and paper mills operated in Maritime Canada between 1955 and 1965. All mills were contacted directly for historical data on slimicide use. An estimate of 1 gal of slimicide per week containing 5.3% mercury by weight was provided by one of the mill representatives and the chemical company that supplied the mercury slimicide (Wellwood, Bowater Mersey Inc., 1998, personal communication; Hoekstra, Buckman Laboratories International, 1998, personal communication). Based on the survey conducted by the PPTAC, we assumed 65% of the mills in Maritime Canada used slimicides containing mercury between 1955 and 1965. As no other data were available, we then assumed that those mills using slimicides containing mercury would consume ~ 1 gal week^{-1}. All mercury contained in slimicides was assumed to be lost in effluents from the mills.

2.3.4. Base–metal smelting

During the smelting of base metals, mercury contained in feedstock material is volatilized due to the high temperatures used during this process. The only smelter in Maritime Canada is located in Belledune, New Brunswick. Emissions from this facility were monitored internally during the 1990s and reported to be undetectable due to the low concentrations of mercury in feedstock and the emissions control devices used (Deveau, Environmental Services Manager, Noranda Inc., 1998, personal communications). Emissions control equipment for this facility includes baghouse devices and acid scrubbers. These devices have been in place since the plant commenced operation in 1966 and are known to capture a substantial portion of the mercury that would otherwise be released to the atmosphere. However, monitoring data reported by the facility have not been confirmed by additional external testing. Given that emission factors for facilities with comparable control devices reported by the USA Environmental Protection Agency's FIRE database (V.51b) are not negligible, we felt it was appropriate to include a high estimate from this source of emissions in our inventory.

The low estimate of emissions is based on emissions reported by the facility. Atmospheric emissions were negligible, and 30 kg year^{-1} was lost from in-product releases for the duration of operation (Deveau, Environmental Services Manager, Noranda Inc., 1998, personal communication). The high estimate was generated by combining yearly production figures (Statistics Canada, 1880–1997, 1960–1985; MRBC, 1945–1995) with the lowest reported emission factors obtained from the FIRE database for comparable facilities with the same emissions control devices. These emission factors were: 10.57 g Hg t^{-1} Cu produced; 1.25 g Hg t^{-1} Zn produced; and 1.58 g Hg t^{-1} Pb produced.

2.3.5. Gold mining

Gold was first discovered in Nova Scotia in 1860 and mining continued until the mid-1900s. During these years large quantities of mercury were used for gold recovery and amalgamation. The amount of mercury consumed for gold amalgamation in Nova Scotia was estimated using a commonly reported emission factor of 32.2 g Hg oz^{-1} of gold produced (D'Itri, 1972) which is similar to other estimates in the literature (Doiron et al., 1998; Wong et al., 1999). This emission factor was combined with the quantity of gold produced annually by these mines obtained from Statistics Canada (Statistics Canada, 1880–1997) to estimate the total amount of mercury consumed.

There are several possible routes of mercury loss during the amalgamation process: losses due to tailing deposits, atmospheric emissions, and losses due to liquid and solid wastes. We assumed that atmospheric emissions would comprise between 10% (low estimate) and 50% (high estimate) of the total mercury consumed (Environment Canada, 1973; Jaques, 1987). We assumed that all mercury used during the amalgamation process would eventually enter the environment and based our total estimate of mercury released on estimated consumption. Mercury lost to tailings deposits may have particular significance due to their slow release mechanisms, thus maintaining the high levels of mercury in abandoned mining sites such as Goldenville, Nova Scotia (Wong et al., 1999). We have not accounted for dispersal of mercury from these tailings deposits in the local environment over a number of years. Instead we consider a total quantity released to all media during the period in which the amalgamation activity took place.

2.3.6. Additional industrial sources of mercury loss

Additional industrial sources of mercury include cement manufacture, glass manufacture, general lab use, iron and steel production, and plastic factories. There was insufficient data to estimate mercury losses from these sources. However, the results of other inventories suggest that only cement manufacture was a significant source of emissions during the time period considered in this inventory (Environment Canada, 1973, 1974; Jaques, 1987; Doiron et al., 1998). Doiron et al. (1998) estimated that releases from cement manufacture in Atlantic Canada (including the province of Newfoundland) were ~ 46 kg in 1995. We were unable to obtain historical data on cement production in Maritime Canada due to industry confidentiality.

3. Results

Cumulative releases of mercury in Maritime Canada between 1800 and 1995 are shown in Fig. 1. The total burden of anthropogenic mercury released to the atmosphere was between 115 and 259 t (low and high estimates, respectively). Coal combustion comprised the largest single component of emissions, accounting for > 25% of cumulative atmospheric emissions. Accordingly, ‘energy’ (fuel combustion) was the dominant category, and made up between 37 (high estimate) and 42% (low estimate) of the total atmospheric emissions. Total mercury releases, which includes mercury lost in wastewater, effluents, and atmospheric emissions, were between 327 and 448 t. The largest individual sources contributing to this total were chlor-alkali facilities, paint application, coal combustion, and gold mining. Each of these sources accounts for > 10% of total releases.

Annual emissions of mercury to the atmosphere in Maritime Canada peaked at approximately 1945 and again between 1965 and 1970 (Fig. 2). In 1945 emissions were > 1750 kg year^{-1}, while between 1965 and 1970 they reached an overall maximum of between 2626 (low estimate) and 4164 kg year^{-1} (high estimate). Categorically, ‘energy’ comprised a relatively constant and significant portion of atmospheric mercury emissions, while the peaks in the 1940s and 1970s were attributable to ‘population-based’ sources and industry (Fig. 2b). By 1995, emissions had fallen from less than 35% (low estimate) to 50% (high estimate) of their earlier maximum. Releases of mercury from fuel combustion and population-based sources accounted for the majority (~ 94%) of emissions in 1995, while industrial sources had fallen from ~ 50% of total emissions during the 1970s, to only 6% in 1995 (Tables 1 and 2).

Total releases of mercury also peaked at approximately 1945 (> 6500 kg year^{-1}) and reached an overall maximum in 1970 between 7896 and 9562 kg year^{-1} (low and high estimates, respectively) (Fig. 3). Due to the large quantities of mercury lost in effluents from individual industrial and population-based sources such as dental uses, gold mining and the chlor-alkali industry, the category ‘energy’ comprises a much smaller component of total releases. Total releases of mercury had fallen dramatically by 1995, to < 36% of the peak level reached in 1970. The category ‘population-based sources’ dominated total releases in 1995, accounting for between 50 and 75% of total releases.

Fig. 4 shows the long-terms trends in emissions from the major individual sources based on the low estimate of releases. Anthropogenic emis-

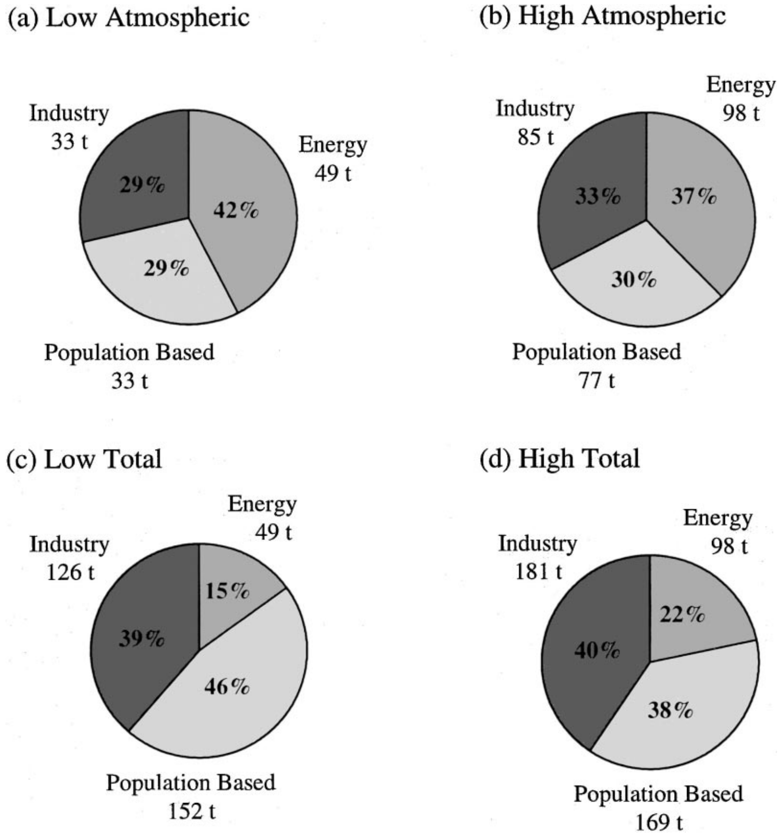

Fig. 1. Atmospheric and total cumulative mercury releases from anthropogenic sources in Maritime Canada from 1800 to 1995. Panels (a) and (b) show high and low estimates of cumulative anthropogenic emissions to the atmosphere, which are 115 and 259 t, respectively. High (448 t) and low (327 t) estimates of total cumulative releases are given in panels (c) and (d), and include mercury discharges in effluents, wastewater, and atmospheric emissions.

sions of mercury in Maritime Canada became significant at around 1850, with gold mining accounting for the majority of mercury released to the atmosphere and lost locally in effluents and tailings deposits. Coal combustion also became a significant source of mercury in this region in the late 1800s, rising in magnitude until 1940, when emissions peaked at > 740 kg year^{-1} (Table 1). The peak period of emissions in 1945 is the result of large quantities of mercury used in pharmaceuticals at that time, combined with moderate levels of emissions from coal combustion and agricultural chemicals. The overall maximum reached in 1970 is largely attributable to releases of mercury from the two chlor-alkali facilities in Maritime Canada. Atmospheric emissions of mercury from these two plants totaled > 1100 kg in 1970, and > 4700 kg when mercury lost in effluents and solid waste was included, comprising $> 55\%$ of total releases.

4. Discussion and conclusions

Although there are no historical inventories of mercury contamination compiled for Maritime Canada, we can cross-check our results against emissions estimates for single years in other studies. We do not expect to have perfect agreement with these inventories as many of the methods used in the past to estimate emissions were changed in later inventories. Thus, our study has the advantage of temporally consistent methods.

However, we do expect our results to show general agreement where comparable. Doiron et al. (1998) estimated anthropogenic emissions in Atlantic Canada (includes Maritime Canada and the province of Newfoundland) in 1995 as 958.3 kg, including a small component of mercury (94.1 kg) lost in municipal wastewater. Our low estimate of atmospheric mercury emissions in Maritime Canada was similar in magnitude at 809 kg, while our high estimate of emissions was somewhat larger at 2098 kg. However, when all losses 'in-product', as effluents, and in wastewater are added, our estimated total releases increases to 1972 and 3480 kg (high and low estimates, respectively). The use of emission factors to generate a high estimate of releases from the base–metal smelter in New Brunswick, as discussed in Section 2, is one major difference between these two inventories. In addition, we included emissions from landfills (high estimate only) and motor vehicles in our inventory, despite considerable uncertainty regarding emissions from these sources. As mentioned previously, this was done to provide an upper bound for mercury releases in Maritime Canada and to avoid consistent underestimation of emissions. Finally, our estimates of total mercury releases are somewhat higher than those provided by Doiron et al. (1998) due to the inclusion of potential mercury losses from dental offices, abrasion and wear of amalgams, and crematoria. There is considerable uncertainty regarding the quantities of mercury lost during the construction, application, and wear of dental amalgams. Our estimates were based on emission factors reported in the 1970s (Cooke and Beitel, 1971; Environment Canada, 1973; Hawley and Sheridan, 1978) and to the best of our knowledge there is no information on how the use and release of mercury from dental applications has changed over time in Maritime Canada. However, our estimates indicate that this may be a significant component of mercury lost in wastewater and effluents. Hence, we recommend further study to clarify how mercury released from dental uses has changed over time in Canada and Maritime Canada.

We can also compare our estimates with those compiled by Environment Canada (1973) for at-

Table 1
Sources of anthropogenic mercury in Maritime Canada

Source	Period of significance	Atmospheric emissions (kg Hg year^{-1})		Total releases (kg Hg year^{-1})	
		Low max.	High max.	Low max.	High max.
Industry					
Chlor-alkali facilities	1965–1995	1112	1345	4709	5432
Agricultural applications	1945[a]–1980	905	905	1899	1899
Pulp and paper slimicides	1955–1965	0	0	11	11
Base–metal smelting	1965–1995	0	597	30	597
Gold mining	1870–1960	82	660	825	825
Energy					
Coal combustion	1850–1995	743	1411	743	1411
Wood combustion	1800–1995	34	130	34	130
Petroleum products	1940–1995	237	332	237	332
Population-based					
MSW disposal	1970[a]–1995	318	497	318	497
Paint application	1945[a]–1995	481	1269	1201	1461
Hospital uses	1900–1995	35	46	35	46
Dental uses	1870–1995	22	52	1209	1240
Pharmaceuticals	1945[a]–1995	723	1808	4450	4450

[a] No data available prior to this year.

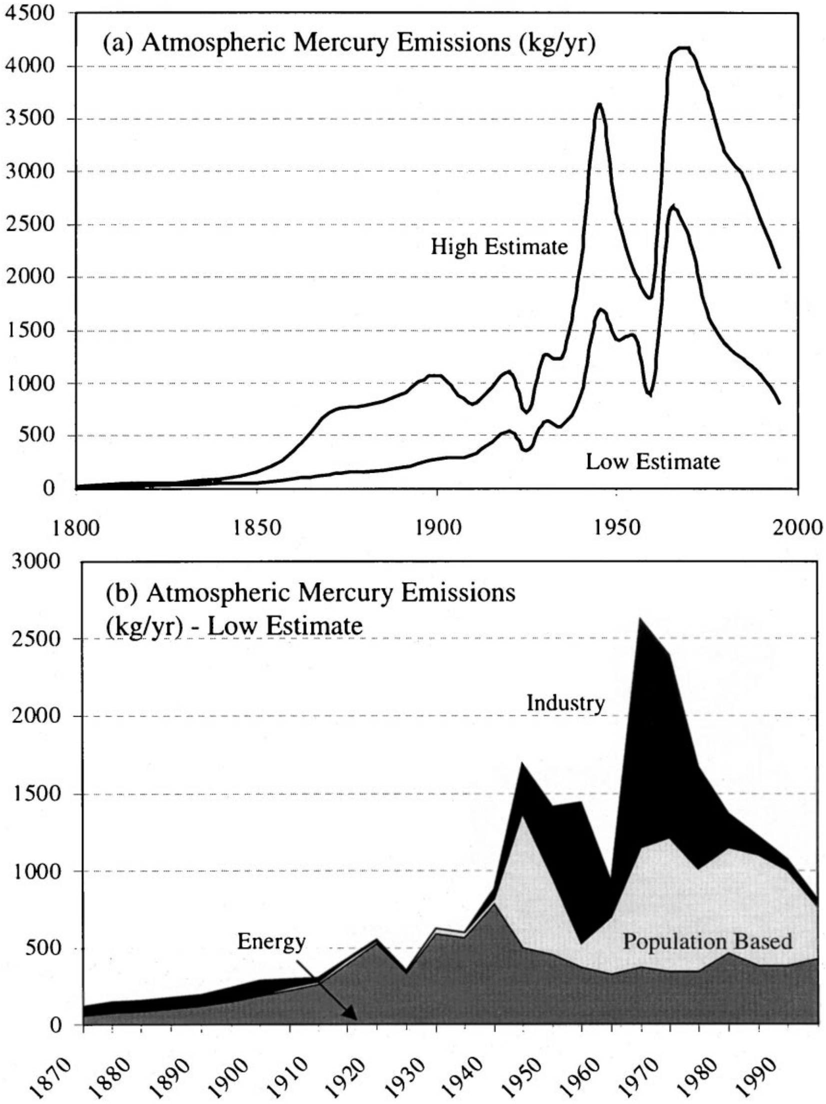

Fig. 2. Historical annual emissions of mercury to the atmosphere in Maritime Canada. Panel (a) shows high and low estimates of total atmospheric releases. Panel (b) shows emissions from each of the three major source categories: industrial sources; fuel combustion (energy); and population-based (miscellaneous sources).

mospheric emissions in 1970 (Table 3). With the exception of coal and petroleum, there is reasonable agreement between our two studies. Estimated emissions from fossil fuel combustion are much higher in the Environment Canada inventory due to higher emission factors used in that inventory. These factors have since been refined with the availability of better measurement technology and cleaner sampling techniques. Once again this illustrates the importance of temporally consistent and explicit calculation methods, as results using different calculation can easily be misconstrued as a trend in historical emissions.

The long-term trends in annual emissions of mercury can reveal much about current and future contamination scenarios. Many of the most significant individual sources throughout history that are depicted in Fig. 4 declined dramatically

Table 2
Major sources of anthropogenic mercury in Maritime Canada for the years 1995, 1970, 1945, 1900 and 1870[a]

	% Atmospheric emissions		% Total releases	
	Low	High	Low	High
1995				
Petroleum	23.2	15.5	9.5	9.3
Coal	28.2	20.6	11.6	12.5
Base–metal	< 5	28.4	< 5	17.2
Chlor-alkali	6.1	< 5	< 5	9.6
Dental uses	< 5	< 5	56.1	32.6
Incineration	28.6	11.0	11.7	6.6
Landfills	N/a	8.7	N/a	5.3
Other	13.9	15.8	11.1	6.9
1970				
Petroleum	7.7	5.2	< 5	< 5
Coal	6.4	7.0	< 5	< 5
Base–metal	< 5	7.5	< 5	< 5
Chlor-alkali	46.5	32.3	59.6	56.8
Dental uses	< 5	< 5	7.2	6.1
Paint	20.1	30.5	15.2	15.3
Incineration	12.8	7.3	5.9	< 5
Other	6.5	10.2	18.0	21.8
1945				
Coal	27.5	24.2	6.9	12.1
Agricultural	17.3	8.0	9.2	8.5
Pharmaceuticals	42.9	49.8	66.5	61.4
Dental uses	< 5	< 5	8.3	7.8
Paint	6.0	7.4	< 5	< 5
Other	6.3	10.6	9.1	10.2
1900				
Coal	55.7	27.8	11.8	19.3
Wood	8.4	8.5	< 5	5.9
Gold	29.1	61.3	61.7	53.1
Dental uses	< 5	< 5	23.6	20.8
Other	6.8	2.4	2.9	0.9
1870				
Coal	23.5	7.9	< 5	5.8
Wood	21.7	14.9	< 5	10.7
Gold	53.7	76.6	77.9	69.4
Dental uses	< 5	< 5	15.6	14.1
Other	1.1	0.6	6.5	0.0

[a] *Note.* All sources < 5% of total releases are included in the category 'other'.

or were phased out by 1995 (Table 1). Such sources included a number of direct or 'advertent' uses of mercury in chlor-alkali facilities, agricultural chemicals, pharmaceuticals, paint application and gold mining. Most of these sources were subject to some form of regulatory action after public concern regarding the toxicity of mercury compounds rose dramatically in the 1960s (MRBC, 1945–1995 Fimerite, 1970). For example, the chlor-alkali industry was the largest individual source category in both Canada and Maritime Canada in the early 1970s, accounting for 47% of total mercury consumption in Canada in 1969 (Buffa, 1973) and between 30 and 60% of atmospheric and total releases in Maritime Canada (Table 2). However, following implementation of the Chlor-alkali Liquid Effluent Regulations under the Fisheries Act of Canada in 1972, emissions from these facilities dropped dramatically. By 1995, the New Brunswick chlor-alkali operation was the last remaining facility in Canada using the mercury-cell process and produced only a fraction of the 1970 emissions levels (Fig. 4). Mercury releases from this facility are unlikely to increase in the future due to stringent regulation and emissions control technology (Paine, 1994).

The direct application of mercury in agricultural chemicals, paint and pharmaceuticals was also subject to regulation and gradually phased out. By 1995, emissions from these sources were also insignificant. The largest single component of total atmospheric releases of mercury in Maritime Canada in 1995 was municipal waste combustion, corresponding to other studies in other regions (Pacyna, 1984; Pirrone et al., 1996). However, the majority (~ 88%) of mercury in the solid waste stream is contained in batteries (USEPA, 1997b). A voluntary industry program to phase out mercury in batteries means emissions from MSW incineration will also decline dramatically in the future (Chevalier et al., 1996). In contrast, inadvertent releases of mercury from the combustion of fossil fuels and base–metal smelting have remained unchanged or grown in significance since the 1970s. Thus, the nature of current anthropogenic sources of mercury is quite different than it was several decades ago when many of the existing policies governing mercury releases to the environment were created. Given the phase-outs of most mercury containing goods,

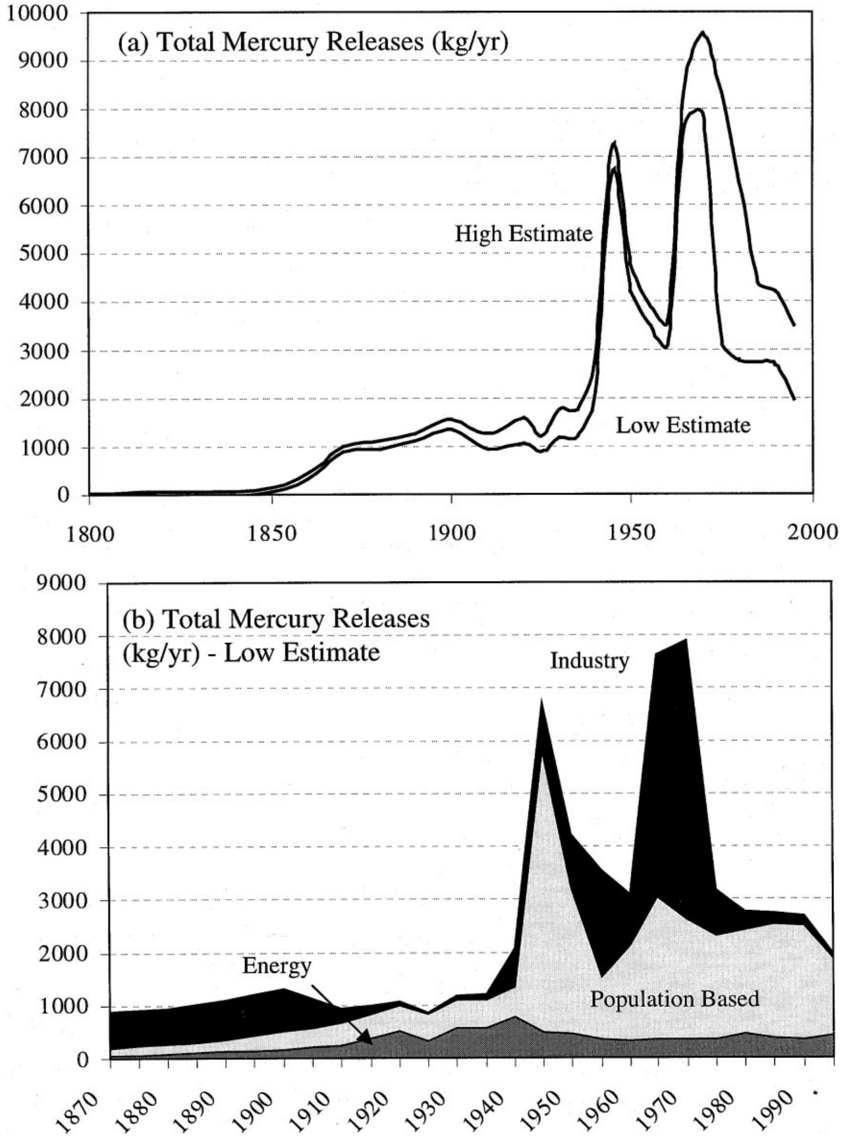

Fig. 3. Total annual releases of mercury in Maritime Canada. Panel (a) shows high and low estimates of cumulative releases from 1800 to 1995, while panel (b) gives components of total releases in the three major source categories: industry; energy; and population-based sources.

cutting back on direct applications of mercury is no longer a plausible regulatory mechanism for reducing current emissions levels. Inadvertent releases of mercury, such as fossil fuel combustion, are much more difficult to regulate as they are often the by-products of multiple activities and there are often diffuse sources. Thus, future efforts to regulate sources of mercury in Maritime Canada may be most effectively focused on technological improvements to existing control technology in order to curb emissions from power plants, mobile sources (motor vehicles) and nonferrous metal extraction operations (Brown et al., 1999).

Comparing the results of our study to the historical inventory for North America compiled by

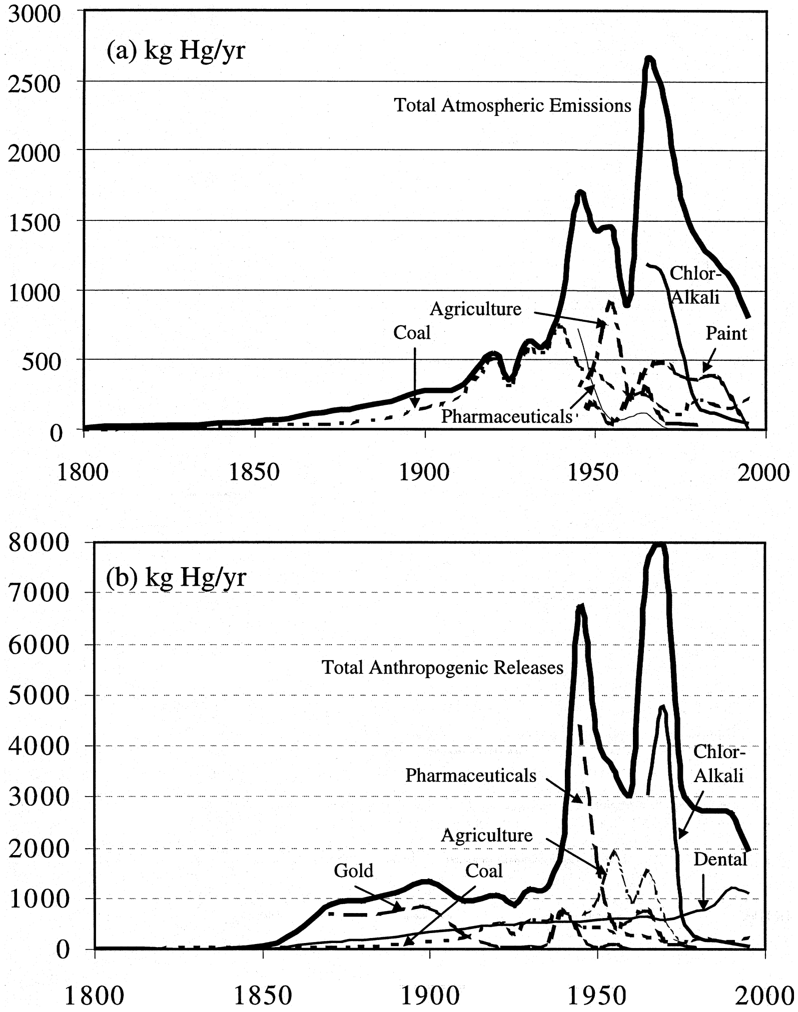

Fig. 4. Principle components of atmospheric (a) and total mercury releases (b) (low estimate shown) in Maritime Canada.

Pirrone et al. (1998) illustrates the degree of correspondence between local, regional and continental scale emissions trends. According to the authors' estimates, atmospheric emissions in North America peaked in 1879 (1708 t), 1920 (940 t), 1947 (247 t), 1970 (325 t), and 1989 (330 t). In Maritime Canada there were two prominent peaks in emissions in 1945 and between 1965 and 1970 (Figs. 2 and 3); however, we do not observe the earlier peaks in the late 1800s and early 1920s. In addition, the peaks in both emissions and wastewater/effluent discharges in Maritime Canada at these times were much more pronounced than those for North America noted by Pirrone et al. (1998). In North America, the high levels of emissions in the late 1800s and early 1900s were the result of extensive gold and silver mining. Although some gold mining did take place in Maritime Canada, it was nowhere near the magnitude of that in other areas (Nriagu, 1993, 1994). We also note a major decline in emissions in Maritime Canada from 1970 to 1995 (as discussed above) in contrast to the further emissions maximum in North America in 1989. This is the result of regulation of industrial sources of mercury and a decline in use of many mercury-containing

Table 3
A comparison of estimates compiled by Environment Canada and this study for atmospheric mercury emissions in Maritime Canada in 1970 (kg Hg)

Category	Environment Canada	This study (low estimate)	This study (high estimate)
Chlor-alkali industry	1795	1112	1345
Dental amalgams (dental uses this study)	9.1	9	21
Instruments	1.8	n/a	n/a
Agricultural chemicals	72.3	69	69
Pharmaceutical use	40.8	25	62
Paint application	344.7	481	1269
Coal combustion	662.1	153	290
Petrol	1932	183	218
Wood	54.4	3	11
Refuse and misc. wastes (landfills this study)	299 + 36	306	306 + 154
Metal recovery	172	0	313

products in Maritime Canada, while Pirrone et al. (1998) noted an overall increase in industrial sources on the continental scale. In summary, although there is some correspondence between peaks in emissions in North America and Maritime Canada in the mid-1900s, the overall patterns of emissions in these two regions are distinct. Further study should be used to analyze whether these differences are distinguishable in sedimentary records of historical deposition, thus helping to clarify the relative significance of local and long-range mercury sources in Maritime Canada.

Quantifying the cumulative releases of mercury in a region provides information on the extent of past pollution in the region. This information can be used to estimate the relative flux of mercury associated with the recycled component of past anthropogenic sources. High and low estimates of cumulative releases of mercury in Maritime Canada from anthropogenic sources reported in this study are 327 and 448 t, respectively. This is $< 0.55\%$ of the 82 000 t released in North America for the same period (Pirrone et al., 1998). However, the significance of this amount should not be discounted. Following the logic of Nriagu (1994), a reemission of only 0.2% of the cumulative quantity of mercury lost from anthropogenic sources in Maritime Canada would be in the range of 650–900 kg year^{-1}. Such releases would be on the same order of magnitude as atmospheric releases from anthropogenic sources in the 1990s (Fig. 2).

It is difficult to determine the absolute contribution of past anthropogenic sources in Maritime Canada to current deposition. However, Pirrone et al. (1996) estimated that approximately one-third of the total burden of Hg in the atmosphere can be attributed to recycled anthropogenic pollution. Estimated wet deposition of mercury in the Atlantic region is ~ 8.5 μg m^{-2} year^{-1} (Beauchamp, 1998). Dry deposition is difficult to measure using existing analytical techniques; however, it is often assumed to comprise approximately 50% of wet deposition (Iverfeldt, 1991; Lindqvist et al., 1991; Fitzgerald et al., 1994). In addition, simulations using the RELMAP atmospheric mercury deposition model in the North-eastern USA indicated that dry deposition makes up 46% of total deposition from anthropogenic sources (NESCAUM et al., 1998). Given that the surface area of the three Maritime Provinces is 134 332 km^2 (Stanford, 1977), and total deposition is approximately 12.8 μg m^{-2} year^{-1} (wet + dry), this corresponds to a total of 1.71 t year^{-1} of atmospheric mercury deposited in the region. If we assume that one-third of atmospheric deposition is from the recycled component of past anthropogenic sources, we can estimate that a flux of ~ 570 kg year^{-1} is associated with past anthropogenic sources. This estimate is similar to that calculated using 0.2% of cumulative Hg releases in Maritime Canada.

It is commonly assumed that ~50% of mercury is deposited locally/regionally (100–2000 km), and ~50% circulates in the atmosphere and is deposited far from its original source (Expert Panel on Mercury Atmospheric Processes, 1994). Using our low estimate of atmospheric releases for 1995, this would result in ~405 kg year^{-1} deposited terrestrially from local and regional sources within Maritime Canada. This estimate is a simplification of reality because it does not take into account potential transport to and contamination from the north-eastern USA. However, the significance of past anthropogenic sources of mercury is clear, even in a region where annual emissions of mercury are low relative to other areas.

Using the above calculations, fluxes of mercury associated with natural and long-range sources of contamination in Maritime Canada should be in the range of 735 kg year^{-1}. If we use estimates of pre-industrial fluxes of mercury derived from sediment core studies from other regions in North America of between 2.0 and 3.7 $\mu\text{g m}^{-2}\ \text{year}^{-1}$ (Swain et al., 1992; Hermanson, 1993, 1998; Lockhart et al., 1998), then natural fluxes can be roughly estimated as being between 270 and 500 kg year^{-1}, leaving 235–465 kg year^{-1} as contamination from sources outside of the Maritime region. Obviously these calculations are somewhat speculative, however, they do provide a preliminary indication of the relative importance of different types of contamination in Maritime Canada. Further study is needed to estimate dry depositional fluxes of mercury in Maritime Canada, fluxes prior to human influences, and fluxes transported from long-range and transboundary sources of pollution.

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