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The history of mercury emissions from fuel combustion in Maritime Canada

E.M. Sunderland[a,*], G.L. Chmura[b]

[a]School of Resource and Environmental Management, Simon Fraser University, Burnaby, BC, Canada V5A 1S6
[b]Department of Geography and Centre for Climate and Global Change Research, McGill University, 805 Sherbrooke St. West, Montreal, QC, Canada H3A 2K6

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"Capsule": *An inventory of historical emissions of mercury from combustion in maritime Canada is presented.*

Abstract

In this study, we present an inventory of historical emissions of mercury resulting from combustion of wood, coal and refined petroleum products in Maritime Canada. The pattern of emissions illustrates the strong influences of population growth, industrial development and prevailing fuel preferences in the region. According to our calculations, anthropogenic mercury releases from fuel combustion in Maritime Canada have cumulatively totaled more than 50 tonnes since 1800. We have compiled both high and low estimates of annual mercury releases in this region. Mercury emissions from fuel combustion in Maritime Canada reached a maximum level in the 1940s. At this time, emissions were between 778 (low) and 1494 (high) kg per year, coinciding with the period of most intensive coal use in Maritime Canada. In 1995, emissions were ~54% of the level reached in 1940, at 427 (low)–800 (high) kg per year. In presenting this emissions inventory, we hope to refine past estimates with current information on the mercury content of different fuel types, and create a comprehensive database on how mercury emissions from various sources have changed over time.

Keywords: Historical records; Fossil fuels; Coal; Wood; Inventory

1. Introduction

Mercury is a known neurotoxin and can cause reproductive and developmental problems in exposed organisms (Clarkson, 1997). Unlike many other naturally occurring elements, the toxic threshold of mercury lies very close to levels that occur naturally in the environment (reviewed by Jackson, 1998). Thus, small amounts of anthropogenic mercury can result in major contamination problems. This is illustrated by the high levels of mercury in fish in remote areas that are distant from point sources, implicating small amounts of mercury scavenged from the atmosphere as the principle source of contamination (Grieb et al., 1990; Hakanson et al., 1990; Rolfhus and Fitzgerald, 1995).

Mercury is continuously recycled between different components of the environment. Human activity can alter the magnitude and distribution of various sources and sinks of mercury, disrupting natural biogeochemical cycles (Mason et al., 1994). Thus, today's population bears the burden of not only the amount of mercury they have produced, but also a cumulative burden of the contaminant released throughout much of human history. One example is the postulated rise in the global reservoir of mercury in the atmosphere over the last 100 years from ~0.3 ng m^{-3} (Mason et al., 1994) to an estimated global average of 1.5 ng m^{-3} (Pirrone et al., 1996). This phenomenon provides one explanation for the observed two- to five-fold increase in atmospheric deposition rates relative to pre-industrial times (Swain et al., 1992; Hermanson, 1993; Lockhart et al., 1998). In order to fully understand and manage environmental problems associated with mercury contamination, it is important to document both past and present sources of anthropogenic mercury. Information on the cumulative amount of mercury released as the

* Corresponding author. Tel.: +1-604-291-5776; fax: +1-604-291-4968.

E-mail addresses: ems@sfu.ca (E.M. Sunderland), chmura@felix.geog.mcgill.ca (G.L. Chmura).

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result of anthropogenic activities will help clarify the expected impact of emissions reductions programs on mercury levels in the ambient environment, and the relative significance of natural and anthropogenic sources of mercury (Rasmussen, 1996).

In a country such as Canada there is great variability between the physical and demographic characteristics of different regions, as well as the level of industrial development. Although there are national-scale inventories of mercury sources in both Canada (e.g. Environment Canada, 1974) and the USA (e.g. USEPA, 1993b), these inventories may ignore sources that are important on the local and regional scale, and do not take a historical perspective on mercury releases. Information on the historical patterns of mercury released on a local scale may be used to interpret geochemically archived records as discussed by Nriagu (1996), through a comparison of local emissions with historical emissions and deposition on larger spatial scales (Pirrone et al., 1998). This information may also help to resolve the ongoing debate regarding the relative importance of natural, local and long-range sources of atmospheric mercury contamination (Rasmussen, 1994; Fitzgerald et al., 1998).

The overall significance of various sources of anthropogenic mercury has changed throughout history. In the past, the most important sources of mercury pollution on a national scale were point sources such as chlor-alkali facilities (Buffa, 1973). Large quantities of mercury were also used in products such as pharmaceuticals and agricultural pesticides (Environment Canada, 1973). In recent years, the total magnitude of mercury consumed and released from these sources has declined dramatically (Chevalier et al., 1996). Accordingly, inadvertent releases of mercury, such as those resulting from combustion of fossil fuels have grown in significance.

In this study we quantify the magnitude of anthropogenic mercury releases resulting from the combustion of fuels in Maritime Canada. We discuss the trends in contaminant releases throughout history and the various factors affecting the amount of mercury released from this sector. In this review, historical emissions are calculated using consistent and explicit methods that may be refined without misinterpretation if further data becomes available. The work presented in this paper is part of a larger study that documents all sources and releases of anthropogenic mercury throughout history in Maritime Canada. The overarching objectives of this study are to refine past estimates of mercury emissions in Maritime Canada with current information on the magnitude of mercury releases and create a comprehensive database on how mercury emissions from various sources have changed over time.

Maritime Canada includes the Eastern Canadian provinces of Nova Scotia, New Brunswick and Prince Edward Island. Maritime Canada can be distinguished from other regions in Canada by its proximity to the highly industrialized northeast USA, predominantly rural population base and relatively low levels of industrial development. Despite regulation of existing sources, high levels of mercury in freshwater fish, common loons, porpoises and seals have been observed throughout this region (Gaskin et al., 1973, 1979; Elliott et al., 1992; Evers et al., 1998; Tatsutani, 1998). Hence, mercury contamination is a longstanding regulatory concern in this area.

Our inventory extends from the year 1800 to 1995. We chose this period to represent the onset of the industrial revolution in the 19th century. Prior to 1800 anthropogenic emissions of mercury in Maritime Canada are assumed to be negligible. This assumption is supported by sediment core studies of historical mercury pollution in the region, which show that contaminant concentrations were not significant until approximately 1880 (Buckley et al., 1995).

2. Materials and methods

In this section we outline the methods used to estimate historical emissions of mercury from fuel combustion in Maritime Canada. The methodology used should be generally applicable to any region for which an estimate of mercury emissions from energy generation is desired.

The three principal fuels used in Maritime Canada are wood, coal and refined petroleum products. Trace quantities of mercury contained within these fuels are emitted to the atmosphere upon combustion. The term "refined petroleum products" includes propane, butane, liquefied petroleum gases, naptha specialties, aviation gasoline, motor gasoline, aviation turbo fuel, kerosene, stove oil, tractor fuel, diesel fuel oil, light fuel oil (LFO), and heavy fuel oil (HFO). Refined petroleum products will be referred to hereon as RPPs. The province of New Brunswick also uses natural gas as an energy source (Statistics Canada, 1943–95). Although data on mercury concentrations within natural gas are limited, the results of other inventories (USEPA, 1993b, 1996), suggest that emissions from this source are negligible.

Although historical data for point sources such as coal-fired power plants and utility boilers are not available, their emissions have been accounted for by coal usage. This should be kept in mind when considering the estimates presented in this inventory, particularly for more recent times when control devices have increased the variability of emissions from point sources. Other regional-scale inventories such as Doiron et al. (1998), should be consulted for individual estimates of emissions from point sources of interest such as large coal-fired power plants.

Emission factors were applied to estimate mercury releases to the environment from each of the three

principal fuel types. An emission factor relates an activity (i.e. amount of fuel consumed), to an amount of contaminant released per unit of that activity. Emission factors are based on test data and direct measurements from point sources. In the case of coal, there is an extensive database on the mercury content of different coal types and associated emissions (e.g. USEPA, 1993a). Information on the mercury content of RPPs and wood is much more limited. Uncertainty in emission factors directly affects the accuracy and precision of mercury release estimates. An extensive literature search was conducted to identify the best available emission factors.

We have chosen to provide 'high' and 'low' estimates of mercury emissions from each source. These estimates do not represent the extremes of possible emission scenarios, rather they are used to represent the range in moderate estimates presented in the literature. We chose to represent moderate estimates rather than the extremes because the large variability between extreme factors would have made the reported range meaningless in practical terms. It is difficult to select emission factors without some degree of subjectivity. However, we used the following general guidelines to minimize arbitrary selection. First, we looked for emission factors with supporting documentation that had been endorsed by major government agencies [e.g. the United States Environmental Protection Agency (USEPA) and Environment Canada]. We then selected emission factors that most closely matched the activity reported in maritime Canada. Because we were interested in reporting the range in moderate emissions estimates, we eliminated the extreme high and low emission factors reported in the literature. The low estimate of emissions is based on emission factors that are more conservative than the high estimate. Recent inventories (e.g. Chevalier et al., 1996; Doiron et al., 1998), likely have applied the most conservative factors available, but the consistent application of conservative emission factors when the information on actual releases is highly uncertain probably results in underestimation of actual emissions. Thus, we have deliberately used high and low emission scenarios to represent the potential range in emission estimates than are normally presented. The methods used to estimate emissions from the three major fuel categories are presented below.

2.1. Wood combustion

Emissions of mercury from wood combustion were calculated by combining an estimated quantity of wood burned with an emission factor. Emission factors reported in the literature ranged from 3.5 to 100 g Hg/kilotonnes (KT) of wood burned. We assumed that the majority of wood burned for anthropogenic applications throughout history in Maritime Canada was for residential use. We were only able to find two emission factors that were directly measured from residential wood combustion units. Thus, our high and low estimates of 50 and 13 g of mercury/KT wood combusted were selected accordingly. Supporting documentation is available for both emission factors. The high estimate was applied in past Environment Canada inventories (Sheffield, 1983), and the low estimate was reported in the latest version of the USEPA FIRE database (FIRE version 5.1b). These emission factors were applied to annual estimates of the mass of wood burned in maritime Canada to estimate emissions.

A number of intermediate calculations were needed to estimate the yearly quantities of wood burned in maritime Canada. There were no data available on specific quantities of wood burned in this region throughout history. Hence, it was necessary to use the proportion of yearly energy consumption that was supplied by wood in Canada between 1800 and 1995 (Fig. 1) to estimate the average amount of energy consumed per capita in the form of wood (Terrajoules, TJ). A conversion factor of 18 TJ of energy released per KT of wood combusted was used to convert this quantity into a mass of wood burned per person (Statistics Canada, 1986). Historical population figures for Maritime Canada were then used to generate a yearly quantity of wood consumed (Fig. 2). Maritime Canada has a predominantly rural population base, and residential use of wood is more prevalent in rural areas. As a result, the calculated amount of wood burned per person most likely underestimates the actual quantity of wood burned. Estimates of mercury emissions from wood combustion are therefore fairly conservative. All steps used to estimate mercury emissions from wood combustion are summarized in Fig. 3.

2.2. Coal combustion

Coal is widely used as both a residential fuel source and for generation of electric power by utilities in

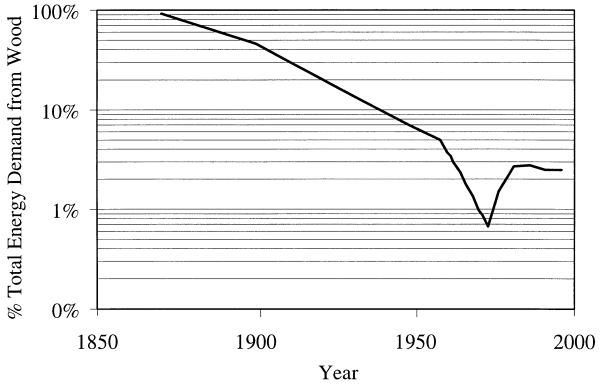

Fig. 1. Proportion of yearly energy demand supplied by wood in Canada. Statistical data adapted from Statistics Canada (1978, 1986, 1991).

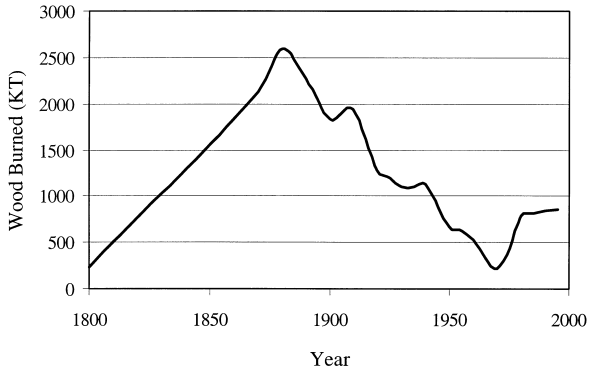

Fig. 2. Estimated quantity of wood burned in Maritime Canada between 1800 and 1995. Statistical data adapted from Leary (1983), Urquhart and Buckley (1965), Statistics Canada (1880–1997, 1978, 1986, 1991).

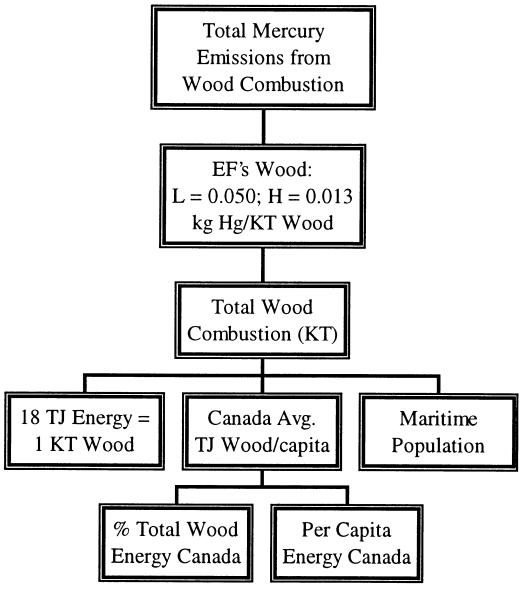

Fig. 3. Summary of methods used to estimate mercury emissions from wood combustion in Maritime Canada. EFs, emission factors; L, low estimate; H, high estimate.

maritime Canada. Mercury emissions from coal were estimated by combining reported mercury concentrations in coal with the total mass of coal consumed in Maritime Canada as considered in one of two categories, "electric power" or "residential". Residential use of coal includes all coal consumed in Maritime Canada that was not used to generate electric power. This number was calculated by subtracting the amount of coal used to generate electric power from the total amount of coal consumed (Fig. 4).

There are a number of emission factors that quantify the amount of mercury released during coal combustion. The magnitude emissions varies greatly depending on the factor selected. We used the following rationale to select emission factors for coal combustion in Maritime Canada: first, we chose emission factors that most closely represented the type of coal used in Maritime

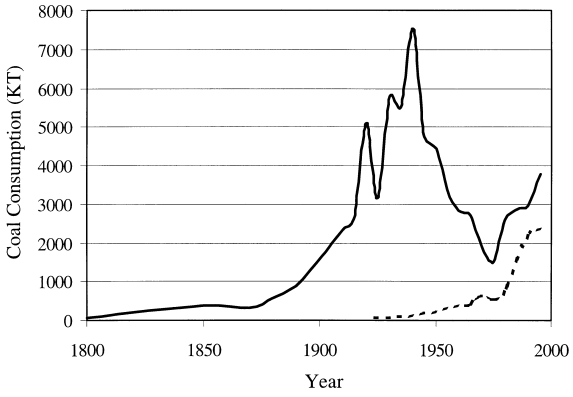

Fig. 4. Total consumption of coal in martime Canada (solid line), and proportion used to generate electric power (dashed line). Statistical data adapted from Leary (1983), Urquhart and Buckley (1965), Statistics Canada (1880–1997, 1948–61, 1950–95, 1966–77, 1992–96).

Canada. The majority of coal consumed in Maritime Canada is the bituminous type (Statistics Canada, 1880–1997, 1950–95) and concentrations of mercury in bituminous coal range from 0.01 to 3.3 ppm (Table 1). Given a reported mean of less than 0.20 ppm in several studies (the mean of means reported by Aucott and Winka, 1996; Pirrone et al., 1996; and Fire v 5.1a), we eliminated the extreme values in Table 1. Although reported mean values of mercury concentrations in bituminous coal (Table 1) tend to converge between 0.18 and 0.196 ppm, we did not want to falsely represent the precision of these estimates by selecting high and low emission factors that were so similar in magnitude, especially given the range of measured values. In addition, looking back on previous Environment Canada reports (e.g. Environment Canada, 1973, 1974) we noted that there is a history of overestimation of emissions from coal combustion. This may be due to imprecision in measurements, environmental contamination and/or low detection limits. However, given this precedent we felt it would be useful to choose a more conservative emission factor to generate the low estimate of emissions from coal combustion. Therefore, based on the study by Billings and Matson (1972), we selected a concentration of 0.10 ppm to generate our low estimate. The emission factor used to calculate our high estimate (0.19 ppm), is the mean of the means listed in Table 1. Although we acknowledge that the low value selected is a conservative estimate of the mean mercury concentration in coal consumed in Maritime Canada, we are confident that the true level of emissions falls within the range of high and low values.

We assume that all mercury contained in coal is released to the atmosphere upon combustion if no emission control devices are in place. This is supported by the analysis of Lindberg (1987) who estimated that $<1\%$ of the mercury in the original feed coal is retained in the residual ash. The emission factor for coal combustion without emission control devices can be

Table 1
Selected examples of mercury concentrations measured in coal

Source	Hg-ppm (mean)	Reference
North American coal	0.01–3.3 (0.18)	Pirrone et al. (1996)
New Brunswick coal	1.2	Sheffield (1983)
Nova Scotia coal	0.3	MacLaren (1973)
Coal combustion (general)	0.10–0.3	Billings and Matson (1972)
Bituminous coal	0.12–0.28 (0.19)	Aucott and Winka (1996)
Bituminous coal (no control)	0.196	FIRE version 5.1a
Residential coal combustion	0.23	FIRE version 5.1b

estimated directly from the mean concentration of mercury in coal. Assuming that residential coal combustion in Maritime Canada has taken place without the aid of emission control devices, the emission factors for mercury released are 190 and 100 g Hg/KT coal combusted.

Many electric power-generating facilities have emission control devices in place. It was impossible to trace the date of implementation of these control devices at each point source, and the effectiveness of such instruments varies widely (USEPA, 1993a). We assumed that there would be a substantial difference between uncontrolled and controlled emissions from point sources, and there would be an overall reduction in mercury emissions from electric power-generation facilities of 50%. The emission factors used to estimate releases of mercury through electric power generation are therefore 95 and 50 g Hg/tonne coal combusted. We estimated that mercury releases in 1995 from electric power generation were between 151 and 286 kg, which is less than a factor of two variation between independently reported emissions of 278 kg $year^{-1}$ from coal-fired power plants in the Atlantic Canada region in 1995 (Doiron et al., 1998), indicating that this is a reasonable approximation. Steps used to calculate mercury emissions from coal combustion are summarized in Fig. 5.

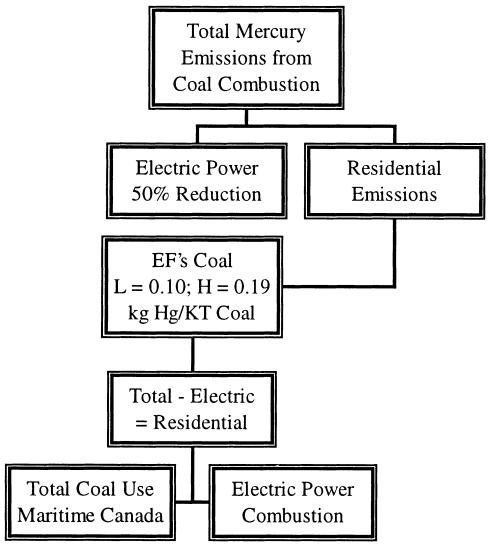

Fig. 5. Summary of methods used to estimate mercury emissions from coal combustion in Maritime Canada, EFs, emission factors; L, low estimate; H, high estimate.

2.3. RPP combustion

The major categories of RPPs include HFO and LFO, diesel oil and gasoline. We chose to use two separate methods (outlined below) to calculate emissions from RPPs due to the high degree of uncertainty surrounding estimates of emissions from this source. These methods are summarized in Fig. 6. The weaknesses and strengths of each method are discussed below.

The low estimate encompasses emissions from the combustion of HFO, LFO and diesel oil. Quantities of RPPs consumed annually in Maritime Canada were obtained from Statistics Canada (Table 2). Gasoline makes up a large component of the total RPPs consumed (Table 2). However, due to the high degree of uncertainty regarding mercury emissions from motor vehicles (USEPA, 1996), gasoline was not incorporated into our low estimate of emissions. There was no information available on consumption of specific fuel types prior to 1940. Therefore, we could not generate any estimates using this method prior to that date.

The USEPA's FIRE (V. 5.1b) database is a compilation of the most current emission factors measured with the best available technology. Emission factors from the FIRE database were applied to generate a low estimate of emissions as we felt they provided the most conservative and reliable estimates in this category. Emission factors for HFO and LFO range from 25 to 123 and 50 to 360 g Hg/1000 m^3 oil, respectively.

A different approach was used to generate the high estimate of emissions. Following the method used by Environment Canada (MacLaren, 1973; Jaques, 1987), we assumed that there is a 'crude equivalent' to the total amount of RPPs consumed. In other words, all the mercury contained in the crude oil used to manufacture the RPPs eventually makes its way into products and is released upon combustion. An emission factor of 30 g Hg/1000 m^3 crude oil, also from Environment Canada (Jaques, 1987), was used to estimate the average mercury concentration in crude oil. There is some error associated with this approach as the implementation of emission control devices in recent times has meant that not all mercury contained in RPPs is released to the atmosphere upon combustion. We feel that this method provides a rough estimate of the magnitude of emissions from this source, but perhaps overestimates emissions in the late 1980s and 1990s when emission control devices were most prominent.

National figures for consumption of crude oil in RPPs were used to estimate the amount of crude oil consumed

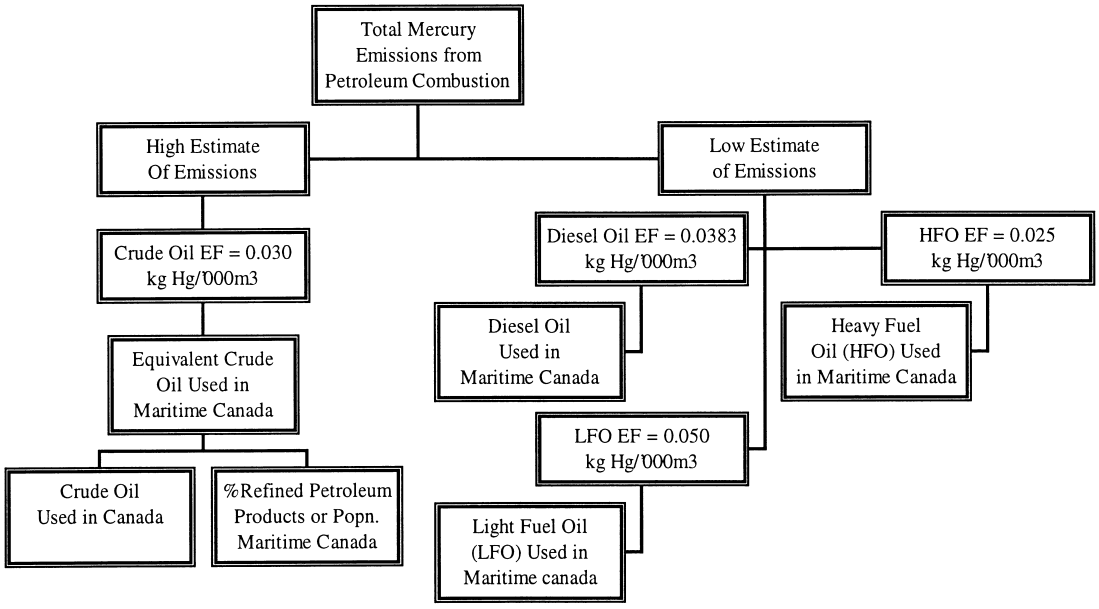

Fig. 6. Summary of methods used to estimate mercury emissions resulting from combustion of refined petroleum products. EFs, emission factors.

Table 2
Consumption of refined petroleum products (RPPs) in Maritime Canada (m^3)[a]

Year	Diesel fuel	Heavy fuel oil	Light fuel oil	Motor gasoline	Kerosene and stove oil	Total RPP use[b]
1995	1.38E+06	2.62E+06	1.39E+06	4.44E+06	6.05E+04	1.04E+07
1990	1.24E+06	3.28E+06	1.54E+06	4.43E+06	9.03E+04	1.12E+07
1985	1.44E+06	2.22E+06	1.58E+06	2.56E+06	1.58E+05	8.60E+06
1980	1.01E+06	4.17E+06	1.87E+06	2.62E+06	2.34E+05	1.03E+07
1975	9.34E+05	3.53E+06	2.00E+06	2.39E+06	4.03E+05	9.60E+06
1970	6.96E+05	2.85E+06	1.71E+06	1.77E+06	5.39E+05	8.25E+06
1965	4.95E+05	1.70E+06	1.18E+06	1.31E+06	4.65E+05	5.65E+06
1960	3.35E+05	3.41E+05	7.62E+05	1.09E+06	3.51E+05	3.07E+06
1955	2.53E+05	5.70E+05	4.10E+05	8.46E+05	2.72E+05	2.50E+06
1950	1.18E+05	3.49E+05	1.49E+05	5.79E+05	1.53E+05	1.37E+06
1945		6.01E+05	2.25E+05	3.46E+05	3.09E+04	1.24E+06
1940		5.78E+05	1.15E+05	3.01E+05	2.33E+04	1.02E+06

[a] Data from Leary (1983); Urquhart and Buckley (1965); Statistics Canada (1880–1997, 1935–95, 1943–95; 1949–72; 1966–73; 1978–95; 1992–96).
[b] Total RPP use includes propane, butane, liquefied petroleum gases, naptha specialties, avaiation gasoline, motor gasoline, aviation turbo fuel, kerosene, stove oil, tractor fuel, diesel fuel, light fuel oil, heavy fuel oil.

in maritime Canada. The amount of crude oil consumed in Canada for RPPs was obtained from Statistics Canada (Table 3). For each year of the period spanning from 1940 to 1995, a proportion of the total crude oil consumed in Canada was allocated to the maritime region according the fraction of total RPPs consumed in that region. Prior to 1940, the fraction of crude oil consumed in maritime Canada (Table 3) was estimated by the percentage of national population residing in that region.

3. Results and discussion

Mercury emissions from fuel combustion in Maritime Canada have cumulatively totaled more than 50 tonnes since 1800 (Table 4). The majority (>75%) of this total is accounted for by combustion of coal. Annual emissions of mercury from fuel combustion reached their maximum level in the 1940s, between 778 (low estimate) and 1494 (high estimate kg per year, coinciding with the maximum level of mercury emissions from coal (Fig. 7B). However, in more recent years emissions from RPPs have been comparable in magnitude to those from coal combustion (Fig., 7B, C, D). Although annual emissions of mercury from fuel combustion in 1995 were approximately 54% of the maximum reached in the 1940s (Fig. 8), there has not been a consistent decline throughout this period. Emission levels increased in the late 1970s and early 1990s. Total emissions from fuel combustion in Maritime Canada are only a fraction of the anthropogenic mercury released in

Table 3
Consumption of refined petroleum products (RPPs) and crude oil in Canada and Maritime Canada[a]

Year	RPPs Canada (m^3)	Crude Canada (m^3)	Crude maritimes (m^3)	% Canada maritimes
1995	8.45E+07	8.77E+07	1.08E+07	12
1990	8.41E+07	9.02E+07	1.20E+07	13
1985	7.80E+07	7.98E+07	8.80E+06	11
1980	1.02E+08	1.10E+08	1.11E+07	10
1975	9.01E+07	8.15E+07	8.69E+06	11
1970	6.95E+07	6.11E+07	7.26E+06	12
1965	5.32E+07	4.88E+07	5.19E+06	11
1960	2.72E+07	3.98E+07	4.49E+06	11
1955	3.02E+07	2.55E+07	2.11E+06	8
1950	1.70E+07	1.39E+07	1.13E+06	8
1945	1.05E+07	9.65E+06	1.14E+06	12
1940	7.83E+06	6.71E+06	8.76E+05	13
1935	5.53E+05	4.80E+06	6.55E+05	10
1930	4.50E+05	3.36E+06	4.67E+05	10
1925	2.30E+05	1.79E+06	3.62E+05	11
1920	1.70E+05	1.01E+06	2.04E+05	11
1915	1.06E+05	6.99E+05	1.32E+05	13
1910	3.31E+05	1.64E+05	1.16E+05	17
1900	1.19E+05	1.22E+05	2.99E+04	18
1890	1.40E+05	8.78E+04	2.42E+04	20
1880	9.32E+04	6.52E+04	1.80E+04	20

[a] Prior to 1935, percent of total crude oil distributed in Canada consumed in Maritime Canada is calculated from the percentage of Canadian population residing in Maritime Canada. After 1935, the fraction of crude consumed in Maritime Canada was estimated by the fraction of RPPs consumed in this region relative to total consumption in Canada. Data from Leary (1983), Urquhart and Buckley (1965), Satistics Canada (1880–1997, 1943–95, 1966–73, 1992–96).

Table 4
Cumulative emmisions of mercury from fuel combustion between 1800 and 1995 in Maritime Canada

	Coal	Wood	RPPs[a]	Total
High estimate (tonnes)	71	12	11	94
% Total high estimate	75	13	12	100
Low estimate (tonnes)	40	3	7	50
% Total low estimate	86	4	10	100

[a] RPPs, refined petroleum products.

Canada, which were slightly under 4 tonnes from coal-fired power plants in a single year (Chevalier et al., 1996).

Mercury emissions from fuel combustion in Maritime Canada have varied with the types and quantities of fuel used throughout history. In the 1800s, the population base of Maritime Canada was small and wood was the principal fuel source. Atmospheric mercury emissions from wood peaked in the late 1800s between 34 and 130 kg per year (Fig. 7A). Both the quantity of wood used and the proportion of total energy derived from wood declined steadily in the 1900s as coal became a more prominent fuel source (Fig. 4). Mercury emissions from coal reached a maximum level in the 1940s, between 743 and 1411 kg per year. This corresponds to the period of maximum coal use, just before the widespread introduction of petroleum-based fuel alternatives (Tables 2 and 3). In the past several decades the quantity of RPPs consumed has been quite variable, often resulting in a decline in mercury emissions from RPPs and a slight rebound in emissions from the alternate fuels, coal and wood. These fluctuations closely coincide with shifts in global oil prices such as the dramatic increases in prices following the OPEC oil crisis of 1973, and the increase in prices in the early 1980s (Helliwell et al., 1989). These trends illustrate the sensitivity of inadvertent mercury emissions from fuel combustion to changes in the prices and availability of alternate fuel sources. Mercury emissions from RPP combustion in Maritime Canada peaked in 1990 between 207 and 360 kg/year (Fig. 7C). The magnitude of emissions in 1995 was $>90\%$ of the level observed in 1990. Given the fluctuations that have occurred in the past, future increases in emissions from RPP use are probable.

Regional preferences for specific fuels also affect the magnitude of mercury emissions. The use of coal in the province of Nova Scotia is greater than the national average due to the abundance of natural mineral reserves in the region. Coal is a less efficient fuel source than RPPs (e.g. more coal must be combusted to produce equivalent energy), and results in higher levels of mercury contamination per unit of energy generated. In contrast, the province of New Brunswick uses natural gas as a principal energy source. Natural gas is a cleaner fuel than RPPs, and based on the results of other studies (e.g. USEPA, 1996), we assume mercury emissions from the combustion of natural gas are negligible. The recent development of natural gas production facilities on Sable Island off the coast of Nova Scotia could therefore have a significant impact on mercury emissions from energy generation in that province. For example, if natural gas is substituted for 75% of the coal currently consumed in the province of Nova Scotia alone, then associated emission reductions would be between 90 and 180 kg per year. This translates into $>20\%$ reduction in current emissions from all sources of fuel combustion in Maritime Canada.

The estimates of mercury emissions presented in this paper are affected by a number of sources of uncertainty. In all cases the range between high and low estimates of emissions is largely due to variability in the emission factors reported. There is a need to refine existing emission factors and develop appropriate 'standard' factors for managers interested in compiling their own emission inventories (i.e. improve the precision of emission inventories). Finally, we feel it is important to emphasize the inclusion of all sources of inadvertent mercury releases from fuel combustion in the future. Estimates of mercury emissions from gasoline combustion by motor vehicles are essential for

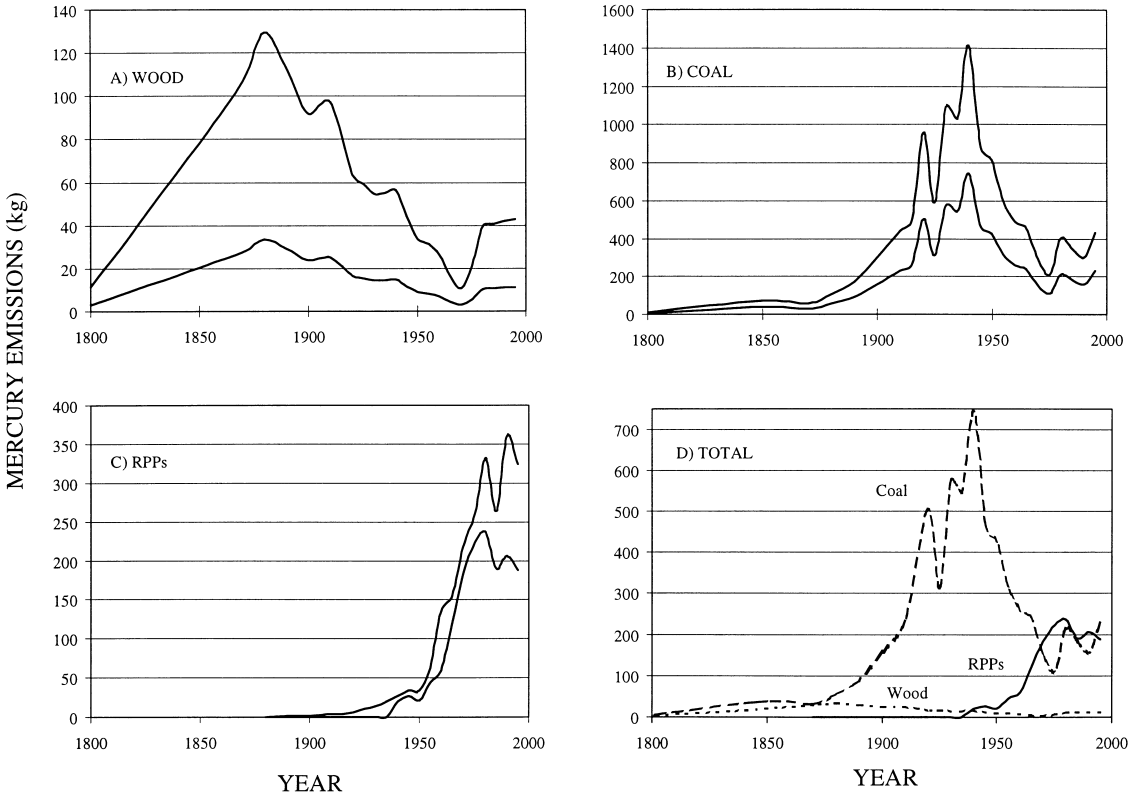

Fig. 7. Historical emissions of mercury resulting from energy generation in martime Canada. A–C show high and low estimates of mercury emissions resulting from combustion of wood (A), coal (B), and refined petroleum products (RPPs) (C). Plot (D) gives the relative magnitude of mercury released to the atmosphere from each source of fuel combustion (based on the low estimate of total emissions). Note that maximum emission levels from combustion of wood, coal and RPPs were reached in 1880, 1940, and 1990, respectively. In 1995, emissions for wood and coal were approximately one-third of their maximum level, while emissions from RPPs were >90% of the 1990 maximum.

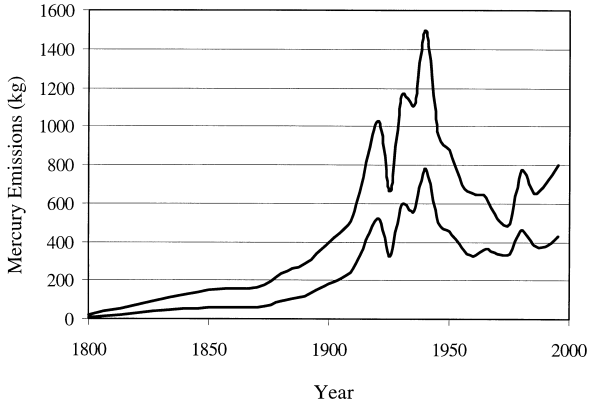

Fig. 8. High and low estimates of mercury emissions from fuel combustion in Maritime Canada. Note that annual mercury emissions in the mid-1990s were approximately 54% of the peak level reached in the 1940s. Approximately half of the current (1995) emissions are the result of combustion of refined petroleum products.

more accurate characterization of mercury emissions from RPPs. This is particularly significant given the importance of RPPs as a source of mercury emissions in more recent years.

The major factor distinguishing this study from other inventories that have been compiled in the past is its temporal scale. With the exception of Pirrone et al. (1998), there are few examples of historical inventories of anthropogenic mercury releases, and to the best of our knowledge none have been compiled on the regional scale. Naturally occurring elements such as mercury may undergo repeated deposition and re-emission once liberated from geologic materials. It is therefore important to combine estimates of current mercury releases to the atmosphere from sources such as energy combustion, with historical information on how the magnitude of these releases has changed over time. In Maritime Canada it is apparent that mercury emissions from coal combustion have declined dramatically since the 1940s, although cumulative emissions are more than double those from other types of fuel combustion (Table 4). Thus, managers considering regulating important modern sources of mercury emissions such as coal combustion must consider the extent to which enacting policies that regulate current levels of emissions will affect the overall burden of mercury in the environment, and adjust their expectations accordingly.

National-scale inventories assembled by Environment Canada in the past have included provincial estimates for fuel consumption, but the estimates of mercury emissions vary widely. Mercury emissions in Maritime

Canada are reported by Environment Canada for the years 1970 (Environment Canada, 1973) and 1978 (Sheffield, 1983), and may be compared to the results of this study. The Environment Canada estimates show a large discrepancy in mercury emissions from petroleum and coal combustion between 1970 and 1978, reporting a decline in mercury emissions of >200% for coal combustion and >400% for combustion of RPPs (Table 5). However, data collected in this study indicate that differences in the amounts of fuel consumed in 1970 relative to 1978 cannot account for the decline in mercury emissions suggested by the two Environment Canada inventories. The difference is instead a function of changes in the calculation methods used, as well as the emission factors chosen in each study. Comparison of past emission inventories that have been compiled independently for different years may lead to misinterpretation of both the success of emission reduction strategies and the absolute magnitude of mercury that has been released into the environment from anthropogenic sources. The use of consistent and explicit methods in this study prevents this type of misinterpretation.

4. Conclusion

Historical mercury emissions from fuel combustion for energy generation in Maritime Canada have responded to changes in population pressure, per capita energy demand, technological efficiency improvements and the availability and price of alternate fuel sources. The trends in both fuel consumption and mercury emissions from settlement to current times presented in this study should roughly parallel changes that took place in all industrialized countries as alternate technologies and fuel sources were introduced.

The emission inventory presented in this study has several advantages over traditional inventories. First, the time-scale of this inventory fills the temporal gaps in past inventories. This allows a more comprehensive assessment of how emissions resulting from energy generation have varied over time and allows an assessment of the cumulative releases of anthropogenic mercury from these sources. Secondly, this inventory has been compiled on a regional scale. It therefore eliminates certain approximations that may be necessary for national-scale inventories and ensures inclusion of local-scale information on consumption patterns and mercury sources. Finally, this inventory uses explicit and consistent methods to calculate emissions over time. This type of analysis helps to eliminate misinterpretation of chronological trends in emissions that may occur when relying on past inventories that do not use consistent methods and emission factors between years.

Table 5
Comparative estimates of mercury emissions in other government-sponsored inventories

	1970 (kg)	1978/80 (kg)
Coal combustion		
Environment Canada	661	282[a]
This study	153/290	212/404[b]
Petroleum products		
Environment Canada	1900	419[a]
This study	184/218	237/338[b]

[a] 1978 Statistic, includes the province of Newfoundland.

[b] 1980 Statistic, Environment Canada data from Environment Canada (1973), Sheffield (1983).

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