Atmospheric Mercury Deposition Impacts of Future Electric Power Generation

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Final Report
submitted to the
Commission for Environmental Cooperation
under project numbers:
111.02.01 and 311.03.03
N/D: 241.01614.037

NOAA Reference Numbers:
(50-22) MAC000/RR1JMA60

December 8, 2003

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* This report was developed for discussion purposes. The opinions, views or other information contained herein do not necessarily reflect the views of the CEC, Canada, Mexico or the United States

1. INTRODUCTION

Mercury contamination in the Great Lakes and many other ecosystems is increasingly being recognized as a serious environmental concern. The dominant route of human exposure to mercury is through fish consumption, and significant portions of the general population are believed to be consuming toxicologically significant levels of mercury (e.g., National Research Council, 2000). Historical discharges – e.g., from chlor-alkali production using the mercury-cell process – are believed to have caused large accumulations of mercury in sediments in Lake Erie and Lake Ontario (Marvin *et al.*, 2003). As these discharges have been substantially reduced, atmospheric deposition is now believed to be a more significant loading pathway for these lakes. Mass balance calculations for Lake Michigan (Mason and Sullivan, 1997) and Lake Superior (Dolan *et al.*, 1993) indicate that atmospheric deposition accounts for approximately 75 percent of the overall mercury loading to these lakes.

To effectively address mercury issues in the Great Lakes (or any other receptor), it is important to know the relative importance of sources of the contamination, as advocated in Annex 15 of the Great Lakes Water Quality Agreement (IJC, 1987) and the Clean Air Act (U.S. EPA, 1990). In a recent publication (Cohen *et al.* 2003), such *source-receptor* information was presented for the Great Lakes, based on a 1995-1996 mercury emissions inventory for the United States and Canada.

The present study extends this earlier work in three significant ways. First, a number of additional receptors are analyzed, in addition to the Great Lakes. The receptors chosen for the present study are shown in Figure 1. Second, emissions estimates for major point sources in Canada have updated based on 2000 data submitted to Environment Canada's National Pollutant Release Inventory (NPRI). Finally, a number of scenarios for future emissions from coal-fired electricity generation plants in the United States and Canada were examined. The emissions inventories and scenarios are described in the next section.

2. EMISSIONS INVENTORIES and SCENARIOS

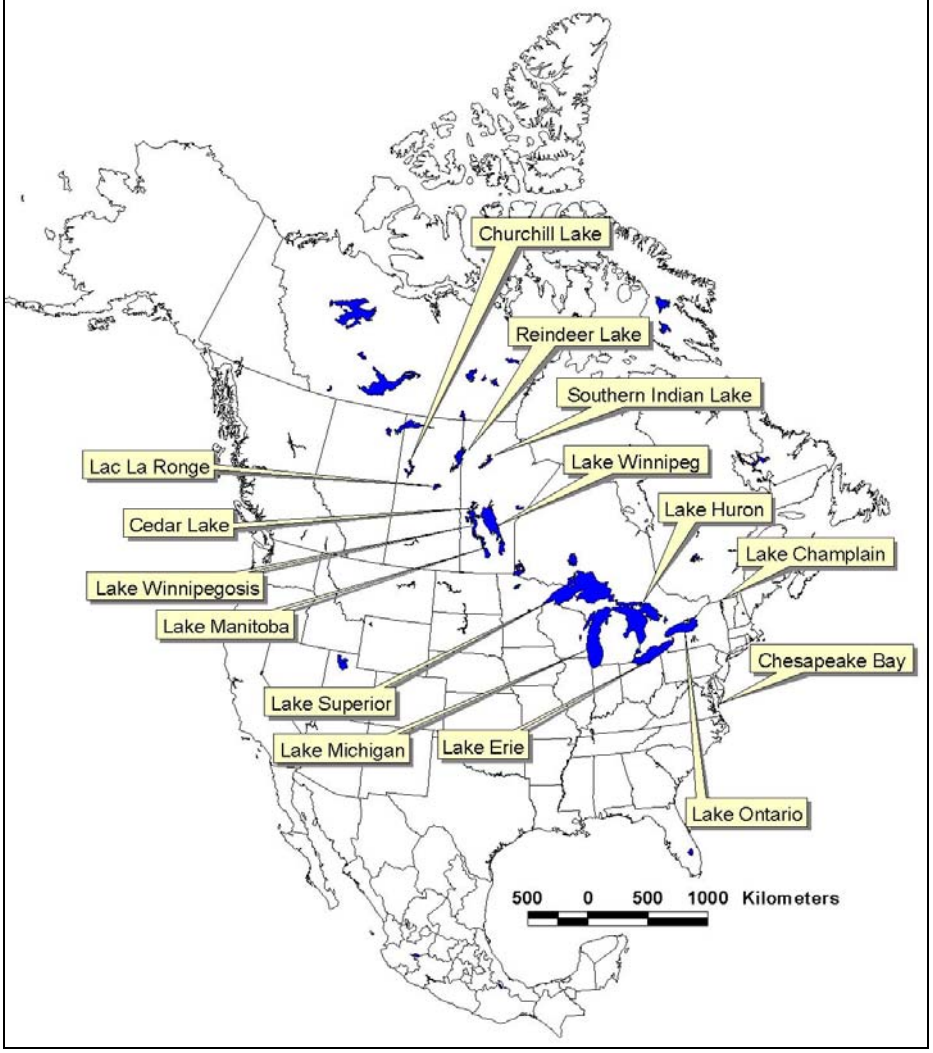
2a. Baseline 1995-2000 Inventory

Ideally, a baseline inventory would be created for one particular year, e.g., 2000. However, data for any one particular year are not available for all sources in the United States and Canada. Therefore, a hybrid 1995-2000 inventory was created, as described below. The only difference between this baseline inventory and that used in the Cohen *et al.* (2003) analysis is that major Canadian point sources have been updated from 1995 to 2000.

First, a mercury emissions inventory for the United States was obtained from the U.S. EPA (Ryan, 2001). The inventory contained annual emissions estimates for most anthropogenic sources of mercury. For coal-fired electricity generation, municipal waste incinerators, and medical waste incinerators, the estimates in this inventory were for 1999, while the remainder were reported to be representative of 1996 emissions.

The U.S. inventory was modified in recognition that one source category – coal combustion in commercial, industrial, and institutional boilers and process heaters (a different category of source than coal-fired electricity generation) – appeared to be under-represented in this inventory, and so data from an alternative 1995-1996 U.S. EPA inventory (Bullock 2000;

Figure 1. Receptors Considered in this Analysis



U.S. EPA 1997) was utilized for this source type. This alternative inventory was also used for U.S. municipal waste incinerators and medical waste incinerators, because significant reductions in emissions from these source categories occurred between 1996 and 1999 (Mobley, 2003), and the goal of the earlier study was to analyze data for 1996.

The coal-fired electricity generation emissions estimates for 1999 were retained because they were based on a significant amount of source testing and were estimated with a much more sophisticated approach than used in previous inventories. Emissions from this source category were fairly similar in 1996 and 1999, at least in total (Mobley 2003).

The U.S. inventory contained a total of 17,513 discrete point sources with specific locations. As is common practice in emissions inventories, certain source categories (e.g. mobile sources, residential fuel consumption, fluorescent lamp breakage) – called *area* sources – were not estimated at precise locations but were estimated at the county level.

Emissions estimates for major Canadian point sources were updated to the year 2000 based on annual mercury emissions information reported by the sources to Canada's NPRI. Significant emissions changes occurred for a few of these sources between the 1995 inventory used in Cohen *et al.* (2003) and this 2000 inventory. For example, mercury emissions from the Teck Cominco smelter in Trail, British Columbia were estimated to have dropped from 1800 kg in 1995 to 150 kg in 2000. This facility's emissions represented a sizable fraction (~17%) of all estimated Canadian mercury emissions in 1995, and so this change was very significant. Discussions were held with a representative of this facility to confirm that these reductions had indeed taken place (Edwards, 2003). The reason for the significant decrease in emissions was that the facility introduced new lead-smelting technology, and the emissions from that new process are now treated with a gas-cleaning system already being utilized by the facility's zinc processes. This pollution control equipment includes a dedicated mercury removal step.

In this Canadian inventory, there were approximately 600 *point* sources. As there were no updates available, the 1995 *area* source inventory used in Cohen *et al.* (2003) was utilized. There were no estimates of emissions from mobile sources available for inclusion in the Canadian inventory.

Emissions from many significant current anthropogenic sources, such as coal-fired electrical utilities or municipal or medical waste incinerators, are generally mixtures of Hg^0, Hg(II) and Hg(p). Because of the distinct atmospheric deposition behavior of these different forms, estimates of the amounts of each form emitted from each source is needed. The U.S. inventory contained such information, albeit on a fairly approximate basis, as the estimates were based on very few measurements for most source categories. Analogous speciation data for the Canadian emissions were not available. Therefore, estimates for the proportions of the different mercury forms emitted from Canadian sources were made by assuming they were similar to the corresponding source-categories in the U.S.

Only *direct anthropogenic* emissions from the reference year(s) have been explicitly included in this analysis. As discussed in Cohen *et al.* (2003) *natural* and *re-emissions* of mercury were approximately accounted for by assuming that they roughly balanced the deposition of *elemental* mercury from direct anthropogenic emissions. In addition, only sources in the U.S. and Canada were included. Other modeling exercises have suggested that the contribution of sources outside the U.S. and Canada to atmospheric deposition to the Great Lakes accounts for ~13% (Shannon and Voldner, 1995) to ~20% (Dastoor, 2003, personal communication) of the total deposition. Inclusion of emissions from Mexico, Europe, Asia, and the rest of the world in this modeling methodology is planned for the future.

There are many uncertainties in both the U.S. and Canadian inventories, and in the application of such inventories in this modeling analysis. First, there have been relatively few measurements of the proportion of the three forms of mercury emitted from various source categories, and so this aspect of the inventory is particularly uncertain. As is discussed throughout this chapter, the atmospheric fate of the different emitted forms is quite distinct; accordingly, source-receptor relationships are strongly dependent on the emissions profile of each source. Second, some potentially significant sources (e.g. electric arc furnaces) were not included. Third, while emissions from some source categories (e.g. coal-fired power plants) have been measured with some regularity, other categories have been rarely measured. As a result, the annual emissions estimates are uncertain for many sources.

Finally, information on the temporal variation of emissions was not available, and so all emissions sources in the inventories were assumed to have been continuous and constant throughout the year. This is probably a reasonable assumption for coal-fired power plants (the largest-emitting source category in the inventory), but may be less appropriate for many other source categories. Even for sources that were relatively continuous, data for episodes such as maintenance or upset-related shut downs were not included in the inventory. Weather patterns can be highly episodic and significantly affect source-receptor relationships; these temporal uncertainties will certainly compromise the accuracy of the estimated concentrations or deposition at any particular location and time. However, this analysis has been conducted over the course of an entire year (and primarily, annual estimates have been generated), and this will likely reduce the overall uncertainty introduced by this variability.

2b. 2020 Emissions Scenarios for Coal-Fired Electricity Generation Facilities in Canada

Two future inventories for Canadian mercury emissions in the year 2020 from coal-fired electricity generation facilities were developed based on electricity generation scenarios from Canada's National Energy Board (NEB) (2003). The NEB developed the scenarios as two different plausible energy futures for Canada, although NEB intended neither to represent a more probable or more desirable energy outcome. The NEB also does not represent the scenarios as forecasts, but as a framework for public discussion on emerging issues and trends. Building upon this intent, we used the NEB scenarios as basis for modeling future mercury deposition scenarios in the receptor lakes of this study.

The first inventory – based on the NEB *supply-push* scenario – represents a world in which technology advances slowly and Canadians take limited action with respect to the environment. The second inventory – based on the NEB *techno-vert* scenario – represents a world in which technology advances rapidly and Canadians take broad action with respect to the environment and the accompanying preference for environmentally-friendly products and cleaner-burning fuels.

Information on the projected locations of additional or reduced generating capacity in these scenarios was provided only by province. Since actual locations are needed to conduct the atmospheric source-receptor modeling, assumptions regarding locations had to be made. To estimate the locations for changes in coal-fired electricity generation capacity in any province, it was assumed that changes were made *proportionally* at each of the *existing* facilities in the province. For example, if a province's 2020 coal combustion was estimated to be 15% more than 2000 levels, coal combustion at each of the province's coal-fired electricity generation facilities was increased 15%. Mercury emissions in the these scenarios were estimated assuming the same emissions factors used in the 2000 emissions estimates. That is, the amount of mercury emitted from each facility *per metric ton of coal burned* in 2020 was assumed to be the same as that in 2000. Because British Columbia had no coal power plants in 2000, but the 2020 supply push scenario includes new coal combustion in that province, we estimated the mercury emissions using the average emission factor derived from neighboring Alberta. It is recognized that these assumptions regarding locations and emissions factors are somewhat arbitrary and other assumptions could be examined in future work.

2c. 2020 Emissions Scenarios for U.S. Coal-Fired Electricity Generation Facilities

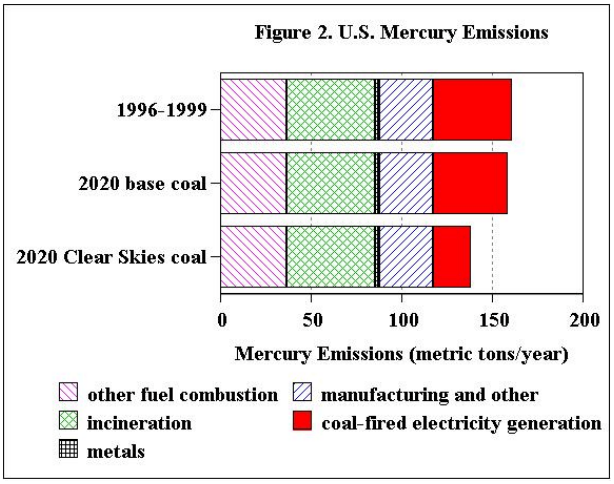
Two scenarios for U.S. mercury emissions in the year 2020 from coal-fired electricity generation facilities were created using publicly available future power plant generation scenarios from the U.S. EPA (2003). The EPA developed the scenarios for the year 2020 using the Integrated Planning Model (IPM) that is designed to analyze a wide range of issues related to the electricity generation sector. The first inventory – a *2020 base case* inventory – represents the estimated emissions from U.S. facilities if no new regulatory limitations were imposed beyond existing programs to cap and trade emissions of sulfur dioxide and nitrogen oxides. The generating capacity in this scenario is estimated primarily on economic and demographic factors, and assumes power plants will not be subject to any mercury controls. The second inventory represents emissions in 2020 from coal-fired electricity generation facilities in the U.S. if the proposed *Clear Skies* legislation is adopted and implemented. For mercury, it represents a presumptive cap of 15 tons (~14 metric tonnes) on mercury in 2018 versus the base 1999 U.S. emissions of about 48 tons (~43 metric tonnes) from coal burning. In the IPM scenario we use for 2020 Clear Skies, however, the total mercury emissions are actually 23 tons (~21 metric tonnes) in 2020 due to provisions in the legislation that would allow "banking" of early excess emission reductions that can be used later under a trading program. Therefore, the 15 ton cap can be exceeded in the early years as sources make use of emissions allowances accumulated from early excess reductions, and this type of compliance strategy is incorporated into the IPM scenario.

Information regarding *specific* facilities is provided in these inventories based on the IPM scenarios, and so assumptions regarding locations were not required. Moreover, the IPM scenarios provided information regarding the emissions from specific facilities, so no assumptions regarding the allocation of emissions increases or reductions were required.

Unfortunately, speciation profiles were not provided in these inventories. To estimate the speciation profile of emissions in these 2020 inventories, the following procedure was used. First, information on speciation profiles for 1999 base-line emissions were examined. It was determined from this examination that the most significant factor affecting speciation appeared to be the presence or absence of scrubbers (wet or dry). On average, when scrubbers are present, the proportion of Hg(II) is much less than when scrubbers are absent. Information on projected pollution control changes at each facility – available in the inventories – was then considered with this factor in mind. If a wet or dry scrubber was predicted to be added to an existing facility, then the speciation profile for that facility's mercury emissions was estimated based on the average profile for facilities with that type of scrubber. For new facilities with scrubbers, the same average speciation profiles were used. For new facilities without scrubbers, the average profile of similar facilities (i.e., those without scrubbers) in the 1999 base-line inventory was used.

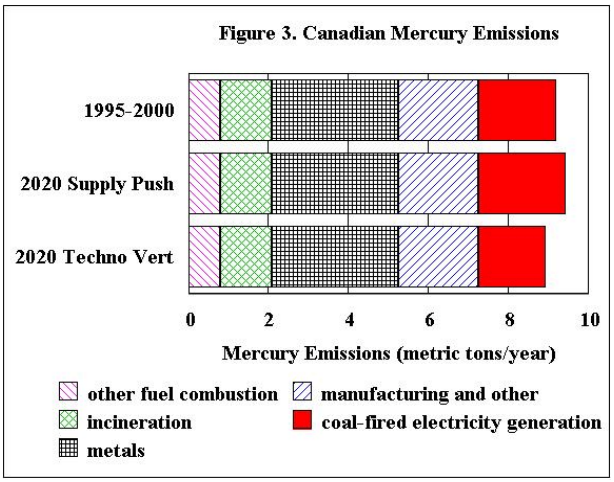
We note that while our 2020 base case inventory assumes no national mercury controls in the U.S., this is a point of some contention. At the time of this analysis, the U.S. EPA was under a court order to develop mercury emission standards for U.S. power plants under an existing provision of the U.S. Clean Air Act. This provision would require the implementation of Maximum Achievable Control Technology (MACT) standards on all affected coal power plants in the U.S. Therefore, even without the passage of the Clear Skies legislation, it has been argued that the MACT standards, if promulgated by the EPA under the court order, would result in

significant mercury reductions from U.S. coal power plants under existing law. For example, a group of Northeastern U.S. States estimated mercury emissions from U.S. power plants after 2007 would be down to about 7 tons (~6 metric tonnes) annually if MACT rules were adopted (NESCAUM 2003). We were unable at the time of this analysis to develop a "MACT scenario" comparable to the 2020 base case and 2020 Clear Skies scenarios generated from the EPA IPM scenarios because we were unable to locate any similar type of electricity sector scenario for 2020 incorporating assumptions of MACT standards for power plants.



2d. Summary of Emissions Inventories Utilized

To summarize the inventories used in this modeling analysis, Figures 2 and 3 show the total anthropogenic mercury emissions in the U.S. and Canada, respectively, broken down into five general source categories: (1) all fuel combustion *except* coal-fired electricity generation; (2) waste incineration; (3) metallurgical processes; (4) manufacturing (e.g., chlor-alkali) and miscellaneous (e.g., lamp breakage) sources; and (5) coal-fired electricity generation. It should be noted that mercury emissions from U.S. medical and municipal waste incinerators have been significantly reduced since the date of this inventory.



3. ATMOSPHERIC MODELING METHODOLOGY

The atmospheric modeling methodology used is described in Cohen *et al.* (2003). Essentially the same methodology was utilized in this analysis, with a few minor differences. The most significant difference was that more *standard source locations* were used in the present analysis – 293, as compared to only 84 in the previous analysis – to provide improved accuracy in the spatial interpolation procedures for the new receptors that were added. Receptors other than the Great Lakes were not considered in the previous analysis, and so the number and spatial distribution of the standard source locations were optimized to provide an accurate estimate for the Great Lakes alone. In the present analysis, the new standard source locations were added to allow accurate interpolations for the new receptors. Please see Cohen *et al.* (2003) for additional explanation of this aspect of the analysis.

4. RESULTS and DISCUSSION

The analysis produces the estimated wet and dry deposition of each form of mercury arising from emissions from *each* source in the various inventory files to *each* of the receptors in the analysis. Thus, there is a tremendous amount of data to synthesize, and, there are many ways to display the results. In Table 1, the total model-estimated atmospheric mercury deposition flux (g/km^2-yr) to each study receptor arising from emissions from each inventory data set is shown. It can be seen from Table 1 that there are often significant differences among different receptors, and among different inventory data sets. This is not surprising, given the different relative efficiencies of transport from sources in a given data set to a given receptor – due to different speciation and spatial distribution – and of course, due to the fact that there are wide variations in the magnitude of emissions in the different inventory data sets.

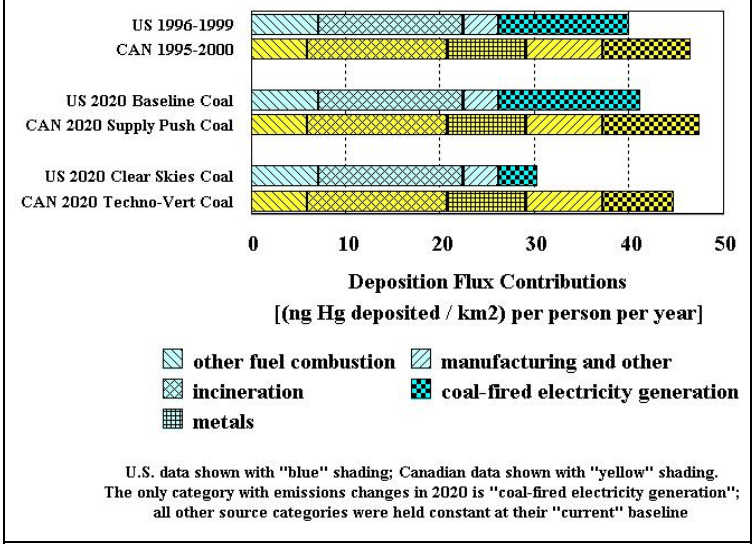
Table 1. Model-Estimated Mercury Deposition Flux to Each Study Receptor (g Hg/ km^2-year)

	Canada					United States				
	"Current"			Future		"Current"			Future	
	1995	2000	2000	2020		1996	1996	1999	2020	
RECEPTOR	area sources	point sources*	**COAL**	**COAL** *supply push*	**COAL** *techno vert*	area sources	point sources*	**COAL**	**COAL** *base*	**COAL** Clear Skies
Lake Erie	0.17	0.29	**0.24**	**0.26**	**0.20**	2.99	5.70	**6.01**	**5.22**	**1.66**
Lake Michigan	0.05	0.09	**0.06**	**0.07**	**0.05**	2.45	6.40	**3.27**	**2.95**	**1.54**
Lake Superior	0.06	0.10	**0.09**	**0.10**	**0.08**	1.13	2.21	**1.34**	**1.27**	**0.68**
Lake Huron	0.14	0.21	**0.11**	**0.12**	**0.09**	1.66	3.70	**2.58**	**2.25**	**0.95**
Lake Ontario	0.47	0.65	**0.28**	**0.31**	**0.23**	2.03	4.92	**3.68**	**3.99**	**1.10**
Lk Champlain	0.43	0.26	**0.05**	**0.06**	**0.05**	1.84	4.88	**2.03**	**1.96**	**0.65**
Chesapeake Bay	0.04	0.05	**0.03**	**0.03**	**0.02**	3.56	12.65	**8.04**	**6.39**	**1.91**
Ches. Bay Watershed	0.04	0.06	**0.03**	**0.03**	**0.02**	2.66	14.80	**3.98**	**3.42**	**0.94**
Lake Winnipeg	0.06	0.30	**0.11**	**0.14**	**0.11**	0.50	0.78	**0.47**	**0.47**	**0.30**
S. Indian Lake	0.03	0.21	**0.07**	**0.08**	**0.06**	0.18	0.28	**0.17**	**0.16**	**0.10**
Lake Manitoba	0.07	0.17	**0.14**	**0.19**	**0.14**	0.59	0.89	**0.53**	**0.53**	**0.35**
Lk. Winnepegosis	0.06	0.31	**0.14**	**0.19**	**0.14**	0.41	0.63	**0.38**	**0.38**	**0.24**
Cedar Lake	0.05	0.56	**0.12**	**0.15**	**0.11**	0.35	0.55	**0.33**	**0.33**	**0.21**
Reindeer Lake	0.03	0.25	**0.08**	**0.10**	**0.07**	0.15	0.24	**0.13**	**0.13**	**0.08**
Lac La Ronge	0.04	0.23	**0.11**	**0.14**	**0.10**	0.18	0.31	**0.16**	**0.16**	**0.10**
Churchill Lake	0.04	0.12	**0.11**	**0.14**	**0.10**	0.12	0.23	**0.11**	**0.11**	**0.07**

* All point sources *except* coal-fired electricity generation facilities

As can be seen from Table 1, other source categories besides coal-fired electricity generation can be significant contributors to any given receptor. An illustrative example of the relative importance of other source categories is shown in Figure 4 for Lake Ontario. In interpreting this figure, note that U.S. incinerator emissions [and therefore their impacts on Lake Ontario] are reported to have decreased significantly relative to these 1996 data.

Figure 4. Per Capita Contributions to Lake Ontario from All Sources



U.S. data shown with "blue" shading; Canadian data shown with "yellow" shading. The only category with emissions changes in 2020 is "coal-fired electricity generation"; all other source categories were held constant at their "current" baseline

All things being equal, the closer the facility is to a given receptor, and the more the prevailing winds blow from the facility to the receptor, the greater will be the source's impact on the receptor. Many of the U.S. sources are *downwind* of the Canadian lakes, but are *upwind* of some or all of the other receptors. Facilities in central and western Canada are the principal facilities *upwind* of the Canadian lakes.

In this analysis, we found the receptors could be divided into two general categories:

(1) the **Great Lakes, Lake Champlain, and Chesapeake Bay** – relatively large deposition fluxes, predominantly influenced by U.S. emissions due to their proximity and for which the impacts of Canadian emissions are relatively small;

(2) the **eight Canadian lakes** studied – relatively small deposition fluxes, influenced by both Canadian and U.S. emissions.

As shown in Figures 5 and 6, the contributions from the U.S. are much, much greater than Canadian contributions for the first group. For the group of Canadian lakes, the absolute contribution of the two countries are roughly comparable.

As shown in Figures 7 and 8, we note that if the contributions were estimated on a *per-capita* basis – with the population of the U.S. being approximately 10 times that of Canada, the contributions from the U.S. would still be greater than Canadian contributions for the first group, while for the group of Canadian lakes, the *per-capita* contributions from Canada would be significantly greater than that from the U.S.

Figure 5. Deposition Flux from Coal-Fired Electricity Generation to "U.S. influenced" receptors

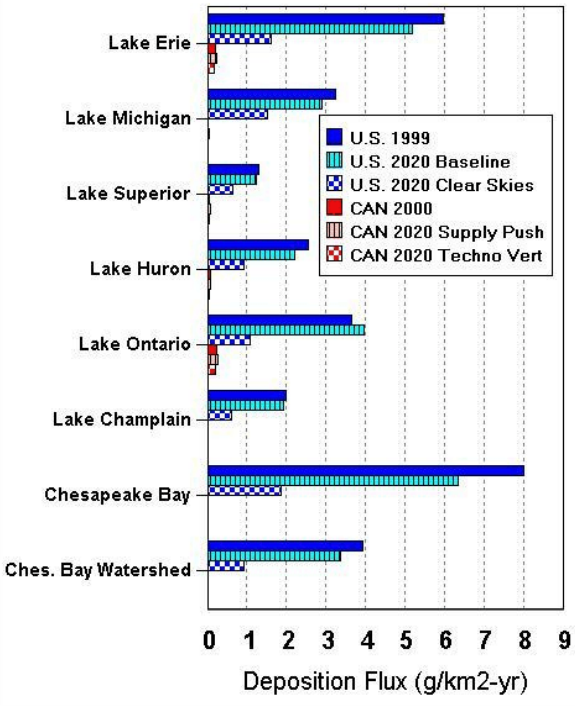


Figure 6. Deposition Flux from Coal-Fired Electricity Generation to "Canada & U.S. influenced" receptors

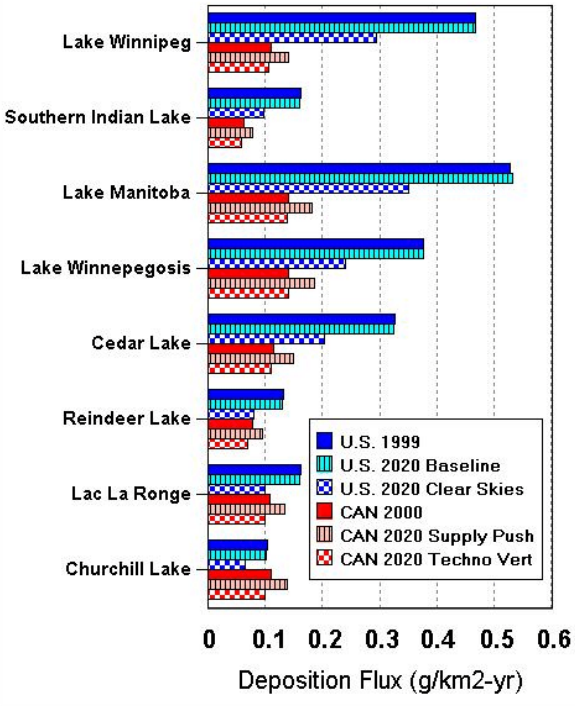


Figure 7. Per Capita Deposition Flux from Coal-Fired Electricity Generation to "U.S. influenced" receptors

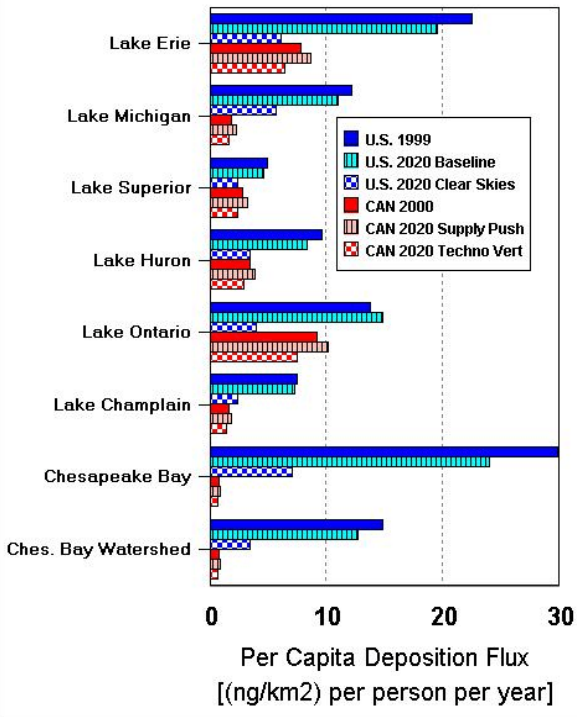
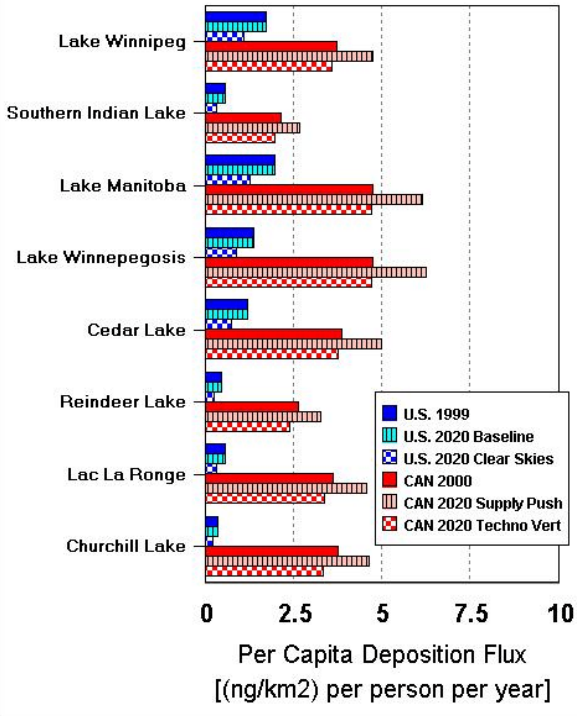


Figure 8. Per Capita Deposition Flux from Coal-Fired Electricity Generation to "Canada and U.S. influenced" receptors



5. SUMMARY

The amount of coal combustion in the U.S. is much greater than the amount of coal combustion in Canada. Mercury emissions from coal combustion in the U.S. are correspondingly much greater than that from Canada. Even on a per-capita basis, coal combustion and mercury emissions from coal combustion are approximately three times greater in the U.S. than in Canada.

Coal-fired electricity generation facilities are not the only sources of atmospheric mercury emissions. Other significant source categories include metallurgical operations and waste incineration. For Canada, metallurgical operations were more significant than coal-fired electricity generation for most of the receptors studied. For the U.S., there have been large reported decreases in emissions from incineration from 1995-2000. Thus, the use of a 1996 U.S. emissions inventory for this source category probably overstated its current impact. Thus, although this analysis found that U.S. waste incineration was more important for most receptors than U.S. coal combustion, this is probably not the case now.

There are three general forms of mercury emissions – elemental mercury, reactive gaseous or ionic mercury (RGM), and particulate mercury. Each of these different forms has a different atmospheric behavior. RGM is much more vulnerable to local and regional deposition. Particulate mercury is less vulnerable. Elemental mercury is much less likely to be deposited locally or regionally. So, knowledge of the speciation of mercury emissions is very important for predicting the deposition to local, regional, and other receptors. Unfortunately, speciation is not well known for many source categories. Also, no speciation data were available for Canada, and data from the U.S. was applied in an approximate manner to the Canadian sources.

The studied receptors appeared to fall into two qualitatively different groups. For the first group – the Great Lakes, Lake Champlain and the Chesapeake Bay – the deposition was relatively high and was dominated by U.S. emissions (even on a *per-capita* basis). For the second group (the eight Canadian lakes), the deposition was somewhat lower, and both Canadian and U.S. emissions sources contributed significantly. On a *per capita* basis, Canadian sources generally contributed significantly more than U.S. sources for this second group.

The 2020 scenarios chosen were not really comparable between the U.S. and Canada. For the U.S. the Clear Skies scenario represented a moderately significant reduction in emissions, while the Canadian Techno-Vert scenario did not represent comparable emissions reductions for Canada. Indeed, the variation in emissions in the Canadian scenarios was not very large. These 2020 scenarios were only one of many possible futures; there are many other scenarios that would be interesting to consider. For example, additional displacement and/or pollution control could be envisioned, such as MACT standards in the U.S., leading leading to much lower Hg emissions from coal-fired power plants than are considered here. Or, less restrictive regulations could lead to higher emissions than considered here.

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