

NUMERICAL SIMULATION OF ATMOSPHERIC LOADINGS OF MERCURY FROM A COAL FIRED POWER PLANT TO LAKE ERIE

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ABSTRACT: Loadings of atmospheric mercury to Lake Erie were numerically simulated with the use of speciated Hg emission scenarios from a coal fired power plant on the shore of Lake Erie in Ontario.

Three-dimensional numerical modeling experiments were conducted using the BLFMAPS-a Mesoscale Boundary Layer forecast and Air pollution prediction system. The modeling system was utilized to simulate meteorology and the air concentration, dry deposition, wet deposition and air-water exchange of Hg species. Simulations were done for Hg containing particulates with three aerodynamic particle diameters of small (0.25 μ m), medium (4 μ m), and large (20 μ m). The numerical experiments exhibited the different characteristics of Hg concentration and deposition patterns of particulate Hg (P-Hg), gaseous elemental Hg (GEM) and reactive gaseous Hg (RGM).

For three out of four emission scenarios RGM is found to be the dominant contributor of the three species of Hg to the Lake Erie loading. The contribution of particulate Hg to the net loading, is relatively small with coarser particles having a stronger deposition rate than finer particles. Fine particles have a longer lifetime in the atmosphere and transport over long distances. 28% of the coarse particle and 7% of fine particle emissions were deposited within 100 km of the power plant. Our experiments also suggest that a case with a larger GEM portion of emission (about 90% of total Hg emission) will have the least amount of total Hg loading to the Lake Erie.

Comparison of model results of surface air concentration and total loading of Hg to Lake Erie with the CAMNeT (The Canadian Atmospheric Mercury Network) and MDN (Mercury Deposition Network of US National Atmospheric Deposition Program) measured data suggest that the power plant has a potential impact of loading of less than 15% of the total atmospheric deposition.

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1. INTRODUCTION:

Mercury is a well-known toxic substance to the aquatic and human life and as an air pollutant it has both natural and anthropogenic sources. Mercury exists in the atmosphere in three major forms: gaseous elemental mercury (GEM), reactive gaseous mercury (RGM) and particulate mercury (P-Hg). GEM is quite volatile, relatively insoluble in water and with weak deposition has an atmospheric lifetime of the order of 1 year, it transports on continental to global scale. The reactive form of Hg (all Hg compounds) RGM is highly susceptible to deposition is short lived (<2d) and transports over local and short range. The P-Hg deposits to the surface through dry and wet deposition processes, more efficiently through wet deposition. The deposition depends primarily on the particle size, land use and boundary layer meteorology. Hence relatively coarser particles travel short distance, whereas fine particles travel a longer distance with residence time of 7 to 10 days. In the presence of air pollutants such as O₃, Cl₂, H₂O₂, and hydroxyl (OH) compounds and in aqueous, aerosol abundant states of the atmosphere the Hg species transform from one form to the other almost continuously in a very complex way. Some of the mechanisms are still not fully understood to explain speciation shift over several hours of transport (Lohman 2006).

The objective of this study is to investigate the impact, namely the atmospheric Hg loading to Lake Erie due to the coal fired power plant that is located at Nanticoke on the northern shore of Lake Erie in Ontario.

With the intent of knowing the impact to the immediate surroundings and the Lake Erie, a circular area with a radius of 100 km will suffice, that is a travel time of only 4 to 6 hours. We assume the concentration of the plume species while undergoing transport, would reduce only through deposition processes and not by chemical transformation. Thus this exercise without explicitly modeling chemical transformation of the plume species should work as a "screening tool" to identify whether deposition from the plant is

significantly above background values before a full blown Hg modeling including chemistry is attempted

2. STUDY AREA:

The coal-fired power plant is located at Nanticoke on the northern shore of Lake Erie in Ontario. The modeling study area occupies 400 km by 400 km (Fig.1a) with Lake Erie is in the middle of domain and the NPG (Nanticoke Power Generating station) is marked on all figures for convenience. The concentric circles with 25, 50 and 100-km radii from NPG are also depicted. The northern region to the power plant is mainly wooded area with mixed broad leaf and needle leaf trees, some agricultural crop lands. South of Lake Erie the land use category is mainly wooded area with deciduous broad leaf trees. Urban centers are about 100 km away from the NPG station (Fig.1b). The land use category data (LUC) are from USGS (US Geological Survey) Global Land Cover characteristic data at 1km resolution. The categories are as given in Table 1.

Table 1. Land use categories (LUC) used in the model for boundary layer dynamics and deposition/leak note that categories 8, 9, 12, and 14 do not exist in the study area.

LUC	Description
1	Evergreen-needle leaf trees
2	Evergreen broadleaf trees
3	Deciduous needle leaf trees
4	Deciduous broadleaf trees
5	Mixed broadleaf and needle leaf trees
6	Grass
7	Crops, mixed farming
8	Desert
9	Tundra
10	Shrubs and interrupted woodlands
11	Wet land with plants
12	Ice cap and glacier
13	Inland water
14	Ocean
15	Urban

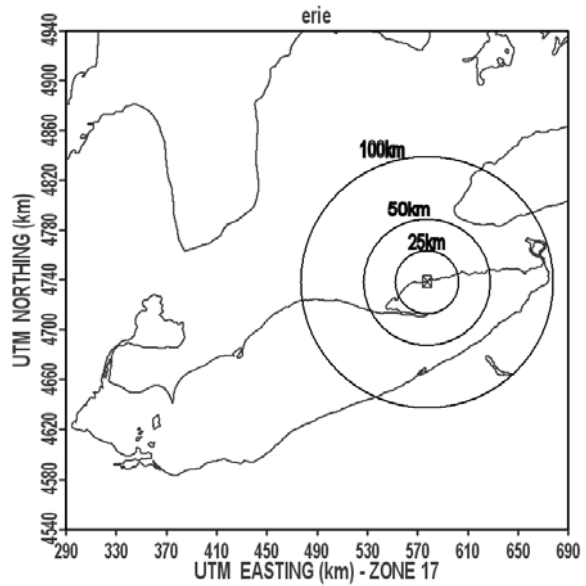


Figure 1a. Model domain with concentric circles of 25, 50 and 100 km radii from NPG at X

BLFMAPS

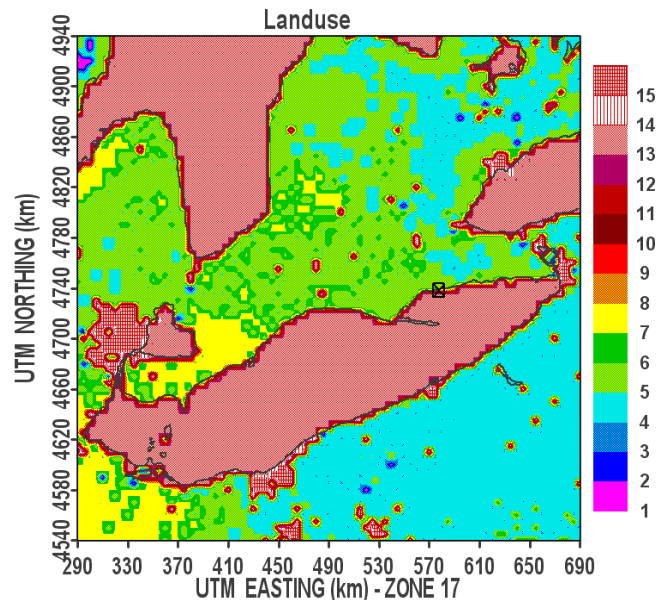


Figure 1b. Land-use category distribution (1 to 15) over the model domain as per USGS data. NPG is at X

3. EMISSION RATES

The annual average total Hg emission rate for the Nanticoke power plant varies between 4 to 8 mg/s (i.e., 130 to 240 kg/y) from NPRI data (National pollutant release inventory, see reference). In the year 2005 the annual average emission rate is 5 mg/s. The speciated Hg emission rate distributions for the set of four scenarios considered here for study are shown in Table 2. Incidentally these scenario distributions have in agreement with the coal fired power plant stack and plume measurements elsewhere in Europe, USA and Canada (Banic et al 2006, Edgerton et al 2006, Pacyna and Pacyna 2005). Scenario C has been observed in an aircraft experimental plume study in 2000 for the Nanticoke coal fired power plant (Banic et al 2006 and Banic personal communication). Hg species measurements of coal fired power plant (CFPP) plumes in USA (Edgerton et al., 2006) showed that on average 2% or less of P-Hg and 84% of GEM and 14% was RGM. It was closer to the scenario C. The US EPA ICR (Information collection request) study showed on average for CFPP the species

distribution was 20% GEM, 78% RGM, and 2% P-Hg a case closer to scenario B (Edgerton et al 2006). However some individual plume events from EPA-ICR and EPRI-ICR data show equal fraction of RGM (45%) and GEM (45%) as in our scenario A and other cases as in scenarios B and C, but in most cases P-Hg was always less than 10%. Lohman et al (2006) indicated of nine CFPP events in U.S. that P-Hg was of 2%, RGM and GEM varying about 50% each a scenario close to A. Similarly stack measurements of speciated Hg emissions from five CFPP in Southeastern US show 30 to 90% of total Hg was in GEM, 11 to 70% was RGM, and about 2% as P-Hg (Seigneur et al (2006). Hg emissions through combustion of fossil fuels in Europe show 50% of GEM, 30% of RGM and 20% of P-Hg (Petersen et al 1995). Thus our scenario D with higher P-Hg fraction of 45% happens to be a rare case. These scenarios presumably cover a range of possible combinations of Hg species in the Nanticoke power plant plume that was undergoing chemical transformation during its transport over 8 to 10 hours over the study domain (travel distance of 200 km from the source).

Table 2. Proportionate Hg species emission rates.

Scenario	GEM	RGM	P-Hg
A	45%	45%	10%
B	5%	90%	5%
C	90%	5%	5%
D	10%	45%	45%

3.1 P-Hg Size Distribution

The particle size distribution of Hg containing particles was grouped into three size bins of large particles of size range >10µm, medium size particles of range 2.5 to 10µm, and small particles of size range < 2.5 microns. 80% of the mass of total particulate Hg is assumed to be in the large size bin, 5% in the medium and 15% of the total particle mass in the small size bin. The aerodynamic diameters selected to represent these bins are 20µm, 4 µm and 0.25 µm. The above scenarios of P-Hg emissions are proportionally distributed to the three size ranges. Table 3 gives emission rates for individual Hg species according to the four scenarios.

Table 3. Emission rates for four scenarios of Hg species.

Assumed speciated Hg emission rates (mg/s) for April and May 2005					
Scenarios with 5 mg/s of total Hg emission					
Scenario	GEM	RGM	P-Hg		
			Large	Small	Medium
A	2.25	2.25	0.4	0.025	0.075
B	0.25	4.5	0.2	0.0125	0.0375
C	4.5	0.25	0.2	0.0125	0.0375
D	0.5	2.25	1.8	0.1125	0.3375

4. MODEL DETAILS IN BRIEF

The BLFMAPS is a combination of a mesoscale meteorological boundary layer forecast model (BLFM, Daggupaty et al 1994) and a set of air pollution transport, dispersion and deposition (APS) modules (Daggupaty et al.2006). The BLFM is a 3D Eulerian model, it utilises twice daily objectively analysed weather data from the Canadian Meteorological Centre (CMC) and will predict meteorological parameters for 12 hours with a five minute time step on a fine horizontal area of 400 km x 400 km with grid spacing of 5 km and over a 10-layered (terrain following) vertical domain (0, 1.5, 3.9, 10, 100, 350, 700, 1200, 2000 and 3000 m above ground level) in this case over Southwestern Ontario (Fig.1a). The three dimensional atmospheric pollutant transport, and dispersion equation with source and sink terms is solved numerically by a finite difference approximation and operator splitting scheme. The horizontal advection terms are solved by an efficient modified Bott's scheme. The predicted meteorological variables, mixed layer depth and turbulent parameters as function of three-dimensional space and time are used in the air pollution modules to predict hourly concentration and deposition as function of time in the three-dimensional model domain. The combined modelling system was originally designed for passive pollutants, and particulate matter. Here it is modified to suit some specifications for Hg species simulations.

4.1. Dry Deposition

Dry deposition formulation follows multi-resistances in the boundary layer. Particle deposition containing Hg is not different from any other metal containing particle. Hence the use of BLFMAPS formulation was followed (Daggupaty et al 2006). Effective dry deposition velocity (V_d^{eff}), which accounts for the influence of sub-grid scale heterogeneous land-type effects, was modeled as in Ma and Daggupaty (2000). Resistance formulation for gaseous pollutants of Zhang et al (2001, 2003) was adopted with modification for effective V_d . The land use information on 1 km sub-grid spacing was used for the effective V_d formulation over the study area.

Following Ma and Daggupaty (2000) and Zhang et al (2001, 2003) effective dry deposition velocity is given as.

$$V_d^{eff} = \frac{1}{R_a^{eff} + R_d^{eff} + R_c} + v_g$$

Ra the bulk aerodynamic resistance is function of boundary layer parameters u^* , Z_0 , L. R_d is quasi-laminar or surface layer resistance, R_c is overall canopy resistance and v_g is gravitational settling velocity.

For gaseous Hg deposition Zhang et al (2003) method was used. The model simulates for different land use classes and shows reliable temporal variation of dry deposition velocity (V_d). For RGM the deposition process is similar to nitric acid and thus, as surrogate, HNO_3 was used. The model predicted maximum V_d over water surfaces is ~ 3 cm/s and over forested regions is ~5 cm/s. These values agree with earlier studies (Berg et al 2001; Ryaboshapko et al 2004). Further a measurement over wetlands for RGM was < 7 cm/s (Poissant et al 2004). GEM deposition is lower than RGM but relatively significant over forested regions and negligible over water and other surfaces and during night time. In the model we selected a surrogate substance C7A carbonyls and adjusted predicted maximum V_d values to ~0.00004 cm/s for water, 0.001 to 0.003 cm/s over vegetated surfaces and 0.04 cm/s over forests. These values agree with some earlier mercury studies (Petersen et al 2001; Ryaboshapko et al 2004; Lindberg and Straton, 1998).

The dry deposition flux ($g\ m^{-2}\ s^{-1}$), is given as
 $F_d(x, y) = c(x,y,z_{1.5}) V_d^{eff}(x, y)$

where $c(x,y,z_{1.5})$ is pollutant concentration in air at 1.5 m above ground level.

4.2. Wet Deposition

The wet deposition flux was calculated as product of the vertically integrated concentration (C), normalised scavenging coefficient and precipitation rate as follows,

$$F_w(x,y) = \Lambda * I(x,y) * \int C(x,y,z) dz;$$

where $F_w(x,y)$ is the wet deposition flux ($g\ m^{-2}\ s^{-1}$), Λ is the normalised scavenging coefficient ($s^{-1}mm^{-1}\ hr$) and I is precipitation intensity (water equivalent in $mm\ hr^{-1}$).

Presence or absence of Hg within a particle and its small contribution to particle mass do not change its aerodynamic properties. Hence as we did earlier for lead and other metals (Daggupaty et al 2006) we follow as in BLFMAPS for particulate

Hg also. For summer season Λ is taken as 1.4×10^{-5} , 2.2×10^{-4} and 1.8×10^{-3} for small, medium, and large particles respectively. During winter, the corresponding values for snow are 4.7×10^{-6} , 7.3×10^{-5} and 6.0×10^{-4} respectively (Schwede and Paumier, 1997, Gatz, 1975, Slinn, 1977).

As GEM has low solubility and RGM is highly soluble, the wet deposition of gaseous Hg is dominated by RGM. We assigned the washout ratio for GEM as 1×10^4 (i.e., $\Lambda = 3.0 \times 10^{-6}$). RGM properties are close to that of nitric acid, the washout ratio as of HNO_3 is 1.6×10^6 (i.e., $\Lambda = 6 \times 10^{-4}$). These are in agreement with Berg (2001), Petersen et al., (1998) and Ryaboshapko et al (2004).

4.3 Adsorption

At the air-water interface there happens bi-directional gas exchange, this process is important in estimating the overall fate of mercury. However in the case here we are interested only in the downward atmospheric flux of gaseous Hg into the lake. Compared to wet and dry deposition processes this exchange is relatively insignificant in the case of GEM and small in the case of RGM.

For GEM the flux is $F(\text{GEM}) = K_w C/H$

where K_w is water side transfer velocity, C is GEM surface air concentration and H is Henry's Law constant (Liss and Slater 1974).

A seasonal average value for spring of 9 (cm/h) is used for K_w (Lai et al 2007).

For RGM, the flux is $F(\text{RGM}) = K_a C$

K_a is transfer velocity to the water and C is surface air concentration of RGM. K_a is 2.0 cm/s (Mason and Sullivan 1997, Lo (1996)).

5. RESULTS AND DISCUSSION

The BLFMAPS model was run for the period of April and May 2005 with all the four scenario emissions from NPG source for individual Hg species as given in Table 4. The hourly averaged concentration and accumulated deposition fluxes to the surface for GEM, RGM and particulate Hg of small, medium and large particle sizes were evaluated over the circular areas surrounding NPG and particularly over the area of Lake Erie.

5.1 Concentration and Deposition Distribution:

Figures 2 a, b display the model computed daily averaged surface concentration distribution for

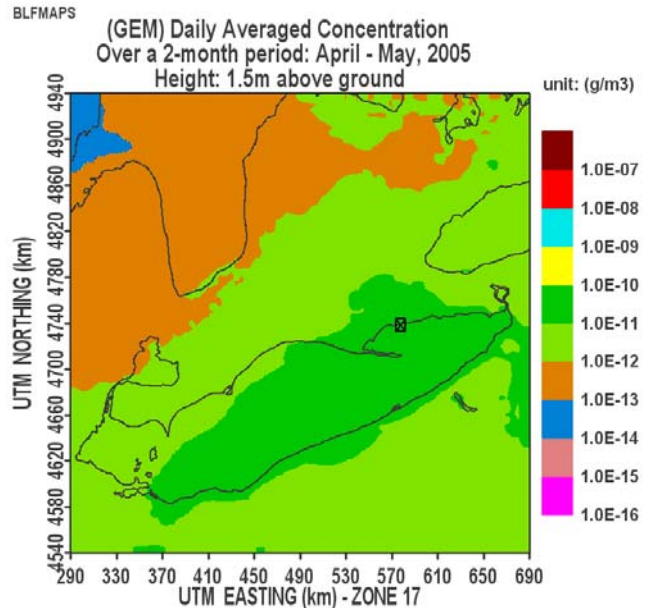


Figure 2a. Model computed daily averaged surface air concentration distribution of GEM with emission rate of 4.5 mg/s (i.e., scenario C).

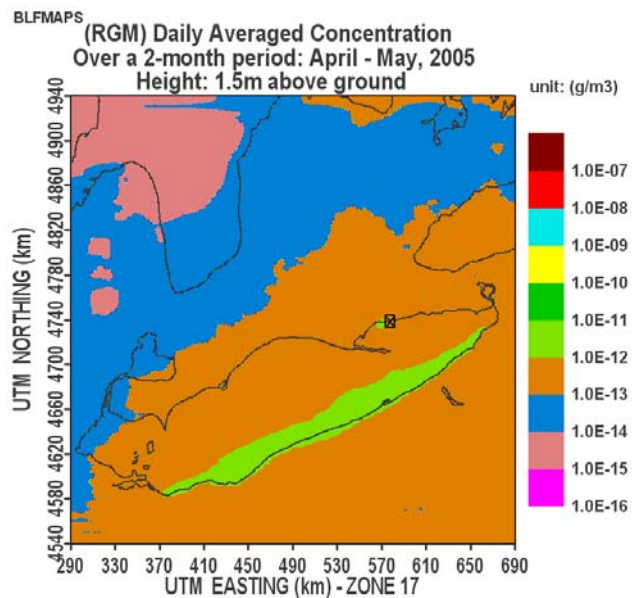


Figure 2b. Model computed daily averaged surface air concentration distribution of RGM with emission rate of 0.25 mg/s (i.e., scenario C).

GEM and RGM with emission rates 4.5 mg/s and 0.25 mg/s respectively and it corresponds to the scenario C. The modeled surface concentration of GEM over Lake Erie seems to be 0.01 to 0.1 ng/m³. The scenario C configuration puts RGM values at 0.1 to 1 pg/m³ over the lake. From

CAMNet (Canadian Atmospheric Mercury Network) monitored data the average surface air concentration for TGM (total gaseous Hg is GEM + uncertain part of RGM) for the study period for Egbert and for (July, August and September 2005) Lake Ontario buoy locations is 1.45 ng/m³. The model simulated average surface air concentration of GEM+RGM due to scenario C emissions from Nanticoke for Egbert is 0.0006 ng/m³, and for near about buoy is 0.003 ng/m³. The model simulation indicates that mercury emitted from Nanticoke has a small influence on the observed air concentration of TGM.

For ease of noting how the individual species behave, model simulations were run with emission rate of 1g/s for each of GEM and RGM. Figures 3 a and 3 b show accumulated total deposition due to dry, wet deposition and adsorption over the two month period. It is quite evident that for equal source strength deposited RGM is almost two orders of magnitude higher than that of GEM in the vicinity of the NPG and about one order more than that of GEM over Lake Erie. It is also remarkable that the higher contoured deposition pattern quickly tapers off away from the source in the case of RGM.

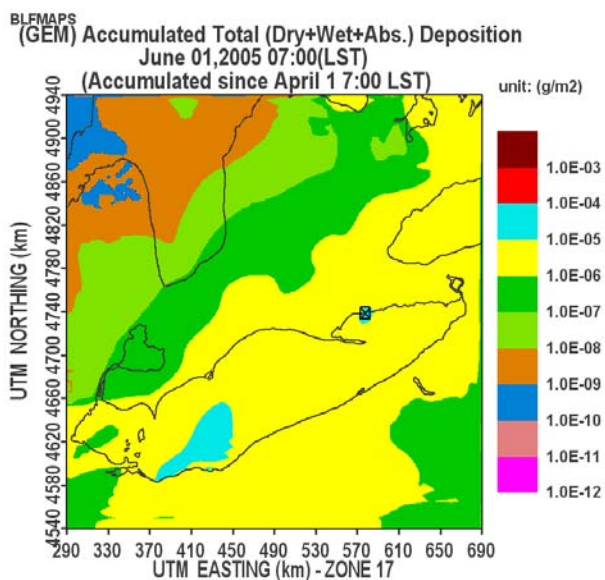


Figure 3a. Accumulated total deposition (dry + wet + adsorption) over two months (April, May 2005), for GEM with emission rate of 1g/s.

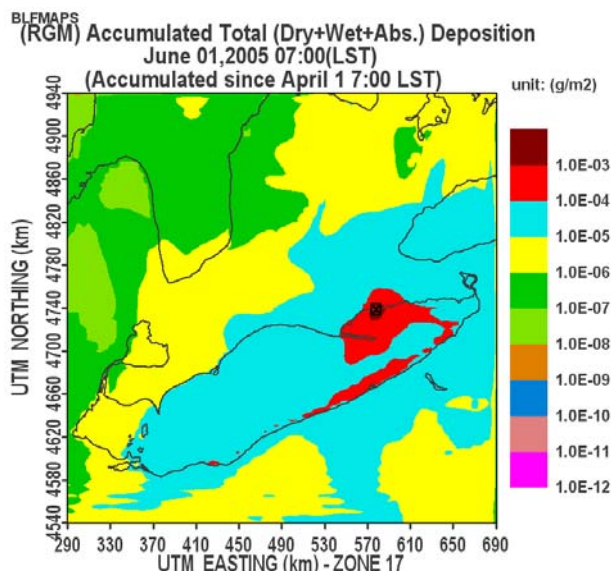


Figure 3b. Accumulated total deposition (dry + wet + adsorption) over two months (April, May 2005), for RGM with emission rate of 1g/s.

5.2 Deposition over Lake Erie

In order to examine the contribution of Hg species to Lake Erie we accumulated the modeled deposition for April and May 2005 over the entire area of Lake Erie. The accumulated deposition is expressed as percentage of the emission of individual species. For example from Figure 4a about 25% large Hg particulate emission was deposited to the lake whereas only about 8% of small particulate emission entered the lake. Further most of the deposition (about 23% large particulate emission and 7% fine particulate emission) was through wet deposition. In the case of GEM Fig. 4b) dry deposition is almost negligible while the total through wet deposition process about 3% of GEM emissions contributed to the lake. The gaseous exchange at the interface was only very little. In the case of RGM with its high solubility and strongly susceptible to deposition over water (Fig.4 c) 25% of RGM emissions over the two months are deposited into the Lake Erie. Again major contributing process is through wet deposition.

Fig. 4a.

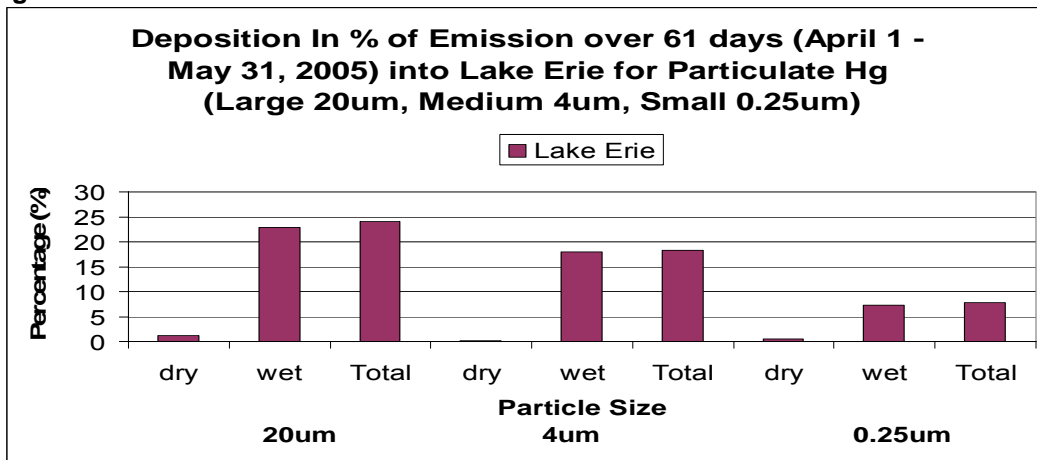


Fig. 4b.

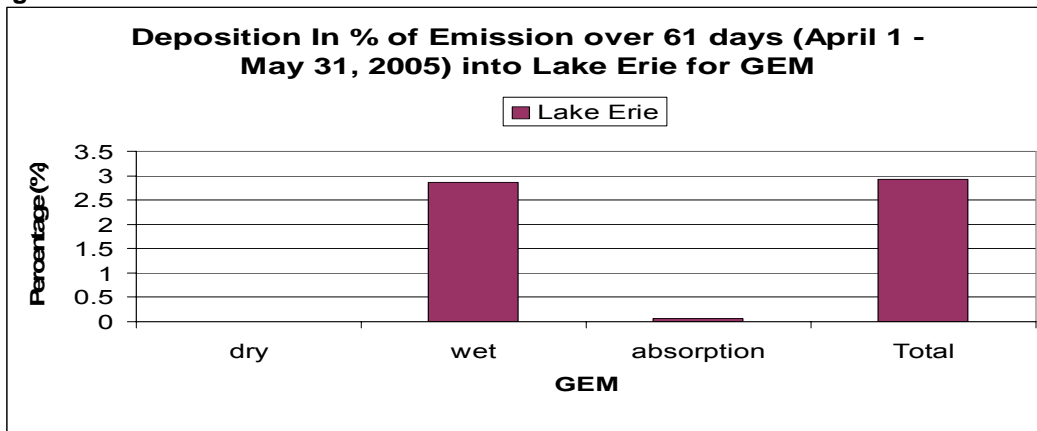


Fig. 4c.

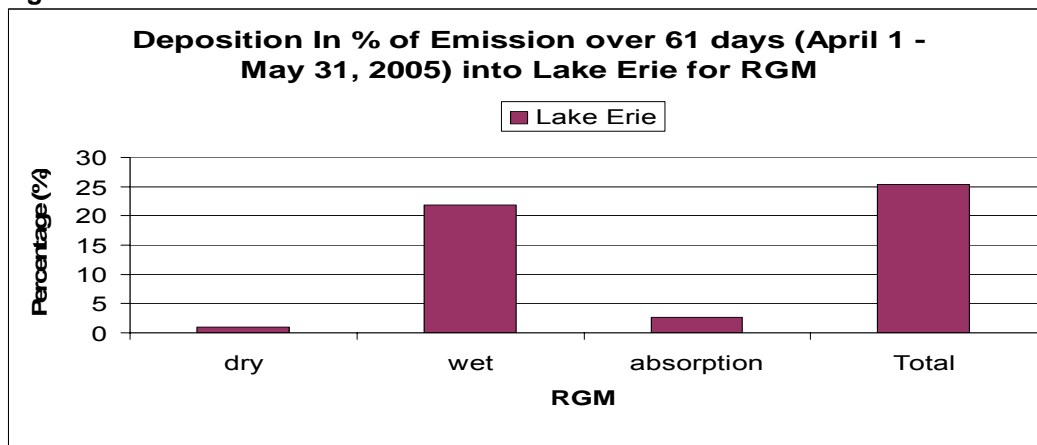


Figure 4a. Accumulated Deposition in percentage of emission for April and May 2005, for particulate Hg (large, medium and small particle sizes) to Lake Erie, through dry, wet, and absorption processes.

Figure 4b. Same as in fig.4a but for GEM.

Figure 4c. Same as in fig.4a but for RGM.

Figure 5a.

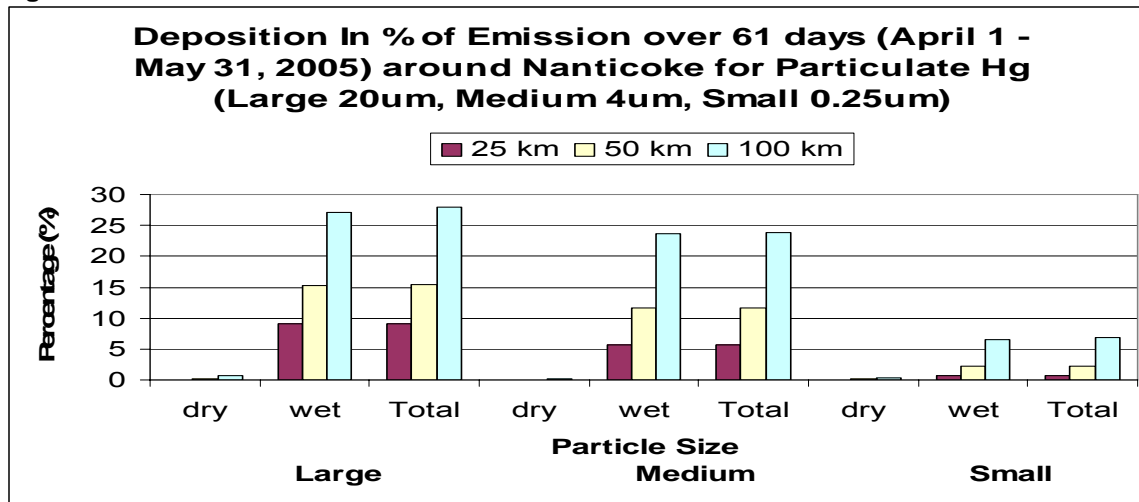


Figure 5b.

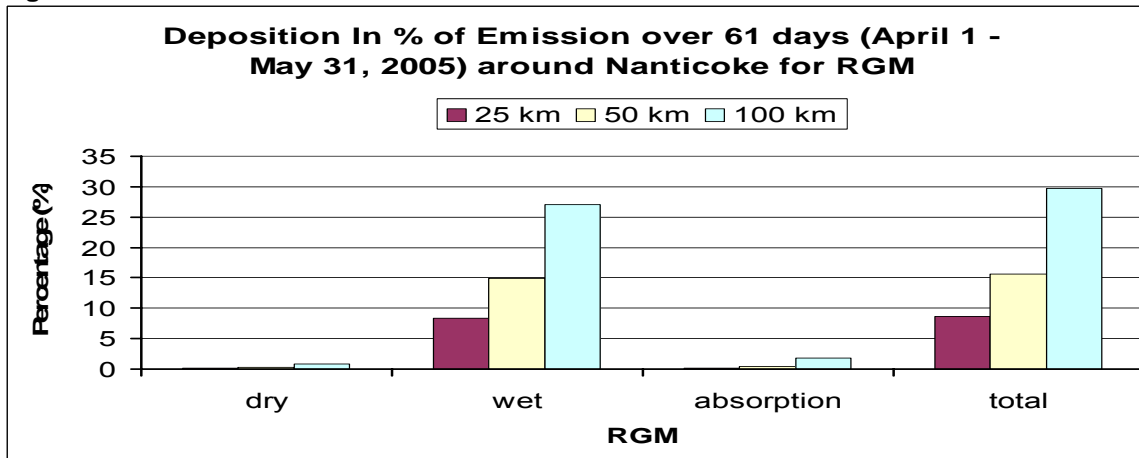


Figure 5c.

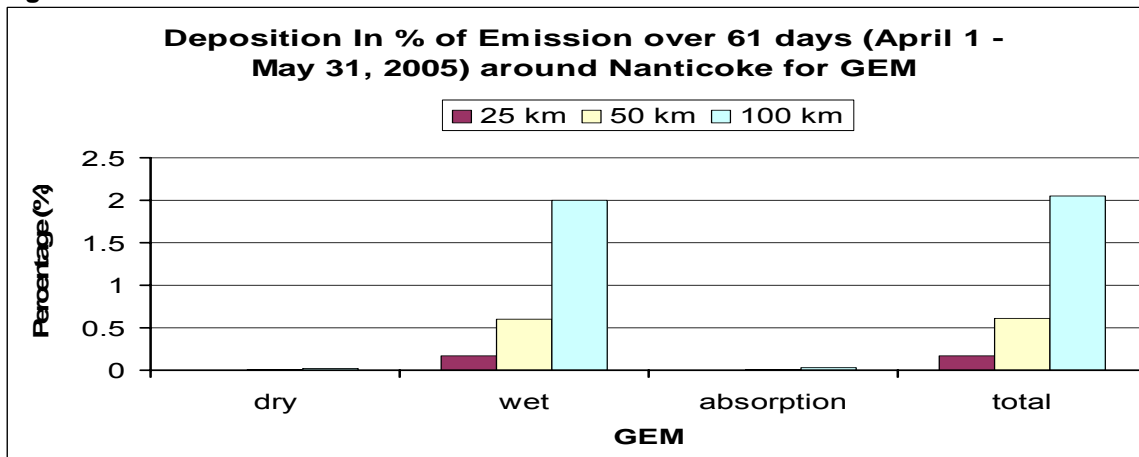


Figure 5a. Accumulated Deposition in percentage of emission for April and May 2005, for particulate Hg (large, medium and small particle sizes) to circular areas around NPG with 25, 50 and 100 km radii through dry and wet processes.

Figure 5b. Same as in fig. 5a but for RGM.

Figure 5c. Same as in fig. 5a but for GEM.

5.3 Deposition surrounding NPG

Similarly the evaluation of deposition in the circular areas around the power plant with 25, 50 and 100 km radii shows that 30% RGM emissions were deposited within 100 km circular area of which 27% was by wet deposition (Fig 5b) process. GEM emissions of 2% only were deposited within 100 km area, attesting that 98% of GEM was transported out of the area (Fig. 5 c). As generally expected elemental Hg is not prone to strong deposition, has the character for long residence time in the atmosphere and subjected to long range transport. The particulate Hg with ability to deposit efficiently through dry and wet processes, it was significantly removed by wet deposition to the extent of 28% of large particle deposition (Fig. 5 a). Whereas only 7% of small particle emissions were deposited within 100 km distance, thus small particle emissions of 93% were also available for long range transport. Hence main deposition contributors within 100 km of the power plant were RGM and Large particulate Hg.

The depositions over the lake and over the areas around NPG mentioned in the sections 5.2 and 5.3 are expressed in percentage of the emissions; hence the remarks are valid for April and May 2005 for all possible emission rates.

5.4 Mercury species loading to Lake Erie

In order to examine quantitatively the actual impact and total loading of mercury species to the Lake Erie due to the power plant we have analyzed the modeled data for all the four scenarios. The following Table 4 gives the details. Major contributing scenario was scenario B with 90% of emissions was in the form of RGM and P-Hg of 5%. As we have seen earlier the species prone to large deposition to the lake are RGM and particulate Hg. The second highest scenario with loading was scenario D with equal contribution of 45% emissions of RGM and P-Hg. The total loading to the lake through scenario B for April and May 2005 was about 9 kg. The least contributing scenario was scenario C with 90% of emission was in GEM phase, the total loading was 1.5 kg. For all Hg species wet deposition was the dominant process that contributes to the loading. From MDN data for 2005 the Erie monitoring site that was representative of Lake Erie has $9 \mu\text{g}/\text{m}^2$

of annual total Hg wet deposition. From our model values of Table 5, Scenario B gives $1.4 \mu\text{g}/\text{m}^2$ per year; it is about 15% of MDN value. The other scenarios A, C and D were 10%, 3% and 15% respectively. One interesting thing to note was increase of GEM emissions (scenario B to C) will reduce wet deposition and also the total loading to the lake. In the same token increase of RGM (scenario C to B) will increase loading. This is obviously due to strong deposition character of RGM and the lake is in the close proximity of the power plant. Further an increase in P-Hg fraction of emission (scenario A to D) lead to increase in contribution through wet and dry deposition processes and thereby increase in the loading. For Nanticoke to have the minimum effect on the loading of mercury to Lake Erie, the emissions would need to have a greater fraction of GEM and a lesser fraction of Hg contained in large particles.

6. CONCLUSIONS

For three out of four emission scenarios RGM is found to be the dominant contributor of the three species of Hg to the Lake Erie loading. Large particle Hg and RGM were the main contributors for total deposition within the area with 100 km radial circle of the plant. 98% of GEM emissions and 93% fine particulate Hg were transported out of the circular area with 100km radius from the power plant. The scenario B emissions give highest loading and it was about 15% of observed MDN value. Our experiments also suggest that a case with a larger GEM portion of emission (about 90% of total Hg emission) will have the least amount of total Hg loading to the Lake Erie. It is prudent to mention that out of the four scenarios the aircraft measured Hg species configuration in the plume is that of scenario C and it has least impact with 3% of the observed total Hg wet deposition to Lake Erie. The chemical transformation of the plume species over the travel period of study domain is hard to anticipate and is not modeled in this study. Thus the plume could be undergoing different combinations of species strength over the travel period. The different scenarios considered here may play a role in deposition as the plume evolves in time. However, it appears that Nanticoke power plant has maximum potential impact of about 15% of the atmospheric Hg loading to the Lake Erie.

Table 4. Mercury loading to Lake Erie as per scenarios A to D.

Hg loadings (g) to Lake Erie for April and May 2005									
Scenarios with 5 mg/s of total Hg emission	GEM	RGM	GEM+RGM	Hg-p			Hg-p Total	Total Hg (GEM+RGM+Hg-P)	
				Large	Medium	Small			
A	Dry	0.0016	473.4	473.4	91.1	0.3	2.2	93.6	567.0
	Wet	332.3	2797.8	3130.1	646.5	24.3	28.5	699.3	3829.4
	Adsorption	7.33	865.5	872.8					872.8
	Total	339.6	4136.7	4476.3	737.6	24.6	30.7	792.9	5269.2
B	Dry	0.00018	946.7	946.7	45.6	0.2	1.1	46.9	993.6
	Wet	36.9	5595.5	5632.4	323.2	12.1	14.2	349.5	5981.9
	Adsorption	0.81	1731.0	1731.8					1731.8
	Total	37.7	8273.2	8310.9	368.8	12.3	15.3	396.4	8707.3
C	Dry	0.0032	52.6	52.6	45.6	0.2	1.1	46.9	99.5
	Wet	664.5	310.8	975.4	323.2	12.1	14.2	349.5	1324.9
	Adsorption	14.6	96.2	110.8					110.8
	Total	679.2	459.6	1138.8	368.8	12.3	15.3	396.4	1535.2
D	Dry	0.00036	473.4	473.4	410.2	1.6	9.9	421.7	895.1
	Wet	73.8	2797.8	2871.6	2909.1	109.4	128.2	3146.7	6018.3
	Adsorption	1.63	865.5	867.2					867.2
	Total	75.5	4136.7	4212.2	3319.3	111.0	138.1	3568.4	7780.6

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http://www.ec.gc.ca/pdb/npri/npri_home_e.cfm. **NPRI** (Environment Canada's National Pollutant Release Inventory).