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Atmospheric gaseous elemental mercury in downtown Toronto

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Abstract

Continuous measurements of gaseous elemental mercury (GEM) in downtown Toronto were conducted from June 2001–February 2002. Ambient air was sampled at 3.5 and 7 m above ground in a canopy over a sidewalk. The quarter hourly mean concentrations were found at $2.39 \pm 2.05\,ng\,m^{-3}$ ($N = 10{,}658$) for the lower sampling level, and $2.57 \pm 2.39\,ng\,m^{-3}$ ($N = 10{,}628$) for the upper sampling level. The average of both sampling levels ($2.48\,ng\,m^{-3}$) was 60% higher than the average of Canadian background emission ($1.55\,ng\,m^{-3}$, Alert). Seasonal concentrations showed the effects of the temperature on the volatilization of Hg^0. Higher GEM was found in the summer time (20 June–19 September) at $3.03 \pm 2.13\,ng\,m^{-3}$ (lower level) and $3.31 \pm 2.56\,ng\,m^{-3}$ (upper level) while lower values were observed in the fall (20 September–19 December) at $1.84 \pm 0.87\,ng\,m^{-3}$ (lower level) and $1.97 \pm 1.38\,ng\,m^{-3}$ (upper level). Log normal probability distribution of the GEM concentration was verified and the double peak hourly average diurnal distribution of GEM concentration was observed in our study.

Keywords: Mercury; Atmosphere; Gaseous elemental mercury (GEM); Urban environment

1. Introduction

Mercury is a highly toxic element that is found both naturally and as an introduced contaminant in the environment. Atmosphere is the major pathway of transporting mercury from its sources of emission to deposition (Lu and Schroeder, 2004; Schroeder and Munthe, 1998). Among all the mercury species in the atmosphere, gaseous elemental mercury (GEM) is proven to take more than 98%, under normal environmental conditions, with a residence time of approximately 0.5–2 years (Poissant, 2000). Once emitted into the atmosphere, elemental mercury (Hg^0) may be transformed to oxidized compounds (Hg(II)) that are readily deposit by dry and wet processes to land and aquatic surface (Edney, 2001). Much more toxic organic species of mercury would then be formed through chemical and biological processes in the natural environment. These toxic mercury species can be bio-accumulated in aquatic food chain more than a million fold (Edney, 2001). Consequently, human consumption of fish and shellfish containing high levels of mercury can result in significant adverse health effects (e.g., neurological damages, preclinical changes in kidney function, delayed development and cognitive changes in children, etc.) because methyl mercury can break the blood–brain barrier and penetrate into the placenta (Clarkson, 1997).

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Research on the concentration status of elemental mercury in the atmosphere is important in understanding the fate of this element in the environment. It is also fundamental for predicting ambient concentrations of mercury species formed through certain atmospheric chemical mechanisms (Edney, 2001). Measurements of atmospheric elemental mercury have been made worldwide at urban (Carpi and Chen, 2002; Dvonch et al., 1995; Fang et al., 2004; Feng et al., 2003; Kim and Kim, 2000, 2001, 2002; Liu et al., 2002; Lynam and Keeler, 2004; Nadim et al., 2001; Sakata and Marumoto, 2002), rural (Banic et al., 2003; Blanchard et al., 2002; Chen et al., 2004; Iverfeldt et al., 1995; Kellerhals et al., 2003; Lee et al., 1998; Poissant, 2000; Schmolke et al., 1999; Slemr and Scheel, 1998), remote (Berg et al., 2003; Golubeva et al., 2003; Poissant and Pilote, 2003; Schroeder et al., 1998), and marine/costal (Gardfeldt et al., 2001; Iverfeldt et al., 1995; Lamborg et al., 1999; Lee et al., 2000; Slemr and Langer, 1992; Temme et al., 2003; Weiss-Penzias et al., 2003) locations. These studies showed the complicated concentration distribution and fate of mercury around the world. A global increase in mercury between 1977 and 1990 was observed (Slemr and Langer, 1992; Temme et al., 2003) and a decreasing trend from European background emission since 1990 was revealed (Iverfeldt et al., 1995; Slemr et al., 2003). Among those GEM measurements, the results from some large cities (urban environment) are summarized in Table 1. The GEM concentrations from these locations ranged from 1.2 to 18.4 ng m^{-3} with standard deviations ranging from 0.7 to 24.8 ng m^{-3}. The values from the locations in Asia (especially China) are higher than those from Europe and North America. These results will be compared with the results obtained from this study.

As for Canada, it is estimated that total annual natural source emissions of Hg was 1.1×10^6 kg in 1989 (Richardson et al., 2003). The Canadian Atmospheric Mercury Measurement Network (CAMNet) was established in 1994. It consists of 11 sites across Canada territory and provides valuable rural and background total gaseous mercury (TGM) data ever since. GEM is believed to consist more than 98% of TGM (Poissant, 2000). The GEM concentration in Canadian urban atmosphere, on the other hand, is less known throughout all these years, because there is no urban site in the CAMNet. This research was carried out to characterize the ground-level atmospheric GEM concentrations and to study the effect of canopy on the GEM concentrations in a typical urban area in Canada.

Table 1
Comparison of GEM concentrations among urban cities

Area	Country	City	Exp. year	Average[a] (ng m^{-3})	Std. dev. (ng m^{-3})	Reference
Asia	China	Beijing	1998	8.3–24.7	3.1–24.8	Liu et al. (2002)
	China	Changchun	1999–2000	18.4	/	Fang et al. (2004)
	China	Guiyang	2001–2002	8.40	/	Feng et al. (2003)
	Korea	Seoul	1987–1988	14.4	9.56	Kim and Kim (2002)
			1999–2000	5.34	3.92	
	Japan	Tokyo	2000–2001	2.70	3.59	Sakata and Marumoto (2002)
Europe	France	Bordeaux[b]	1995–1996	2.7	/	Pecheyran, et al. (2000)
North America	USA	Four sites in Connecticut[c]	1997–1999	2.19–2.69	0.66–1.72	Nadim et al. (2001)
	USA	Broward County	1993	2.8–3.3	/	Dvonch et al. (1995)
	USA	Chicago	1994–1995	3.6	2.9	Landis et al. (2002)
	USA	Detroit	1999–2002	1.17–40.33	/	Lynam and Keeler (2004)
	USA	New York	2000	3.84	/	Carpi and Chen (2002)
	Canada	Toronto	2001–2002	2.48	2.22	This study

[a]Range values are presented when measurement took place in several sites.
[b]Low temperature/GC/ICP/MS method.
[c]Bridgeport, East Hartford, Old Greenwich, Waterbury.

2. Experimental

2.1. Sampling site

The sampling site was located on Ryerson University campus, an area surrounded by commercial, public, and private buildings in downtown Toronto (population ~3,000,000), Ontario, Canada (Fig. 1). The latitude and longitude of the sampling site are 43°40′N and 79°24′W, respectively.

2.2. Instrumental

Two automated mercury analyzers were used in this study. Gardis (Gardis-1A$^+$, Lithuania) (Urba et al., 1995) was used from 20 June 2001 to 09 January 2002 and Tekran mercury vapor analyzer (Model 2537A, Tekran Inc., Toronto, Canada) was used from 14 January to 28 February 2002 after the breakdown of the Gardis analyzer. Both analyzers have a built-in air pump for air sampling and employ gold traps for mercury pre-concentration and thermal desorption for sample preparation. The differences between these two analyzers are: (1) Gardis has two gold traps arranged in series, one of which is a mercury collector and the other an analytical trap. The Tekran mercury vapor analyzer has a dual gold cartridge design, arranged in parallel, thus allowing continuous measurements of mercury in the air sample; and (2) Gardis uses cold vapor atomic absorption spectrometry (CVAAS) while Tekran uses cold vapor atomic fluorescence spectrometry (CVAFS) for mercury detection. It has been demonstrated that the results generated by these two instruments are comparable (Munthe et al., 2001).

Both instruments were calibrated by manual injection of known volumes of air saturated with elemental mercury at a selected temperature. In addition, the Tekran vapor analyzer was calibrated

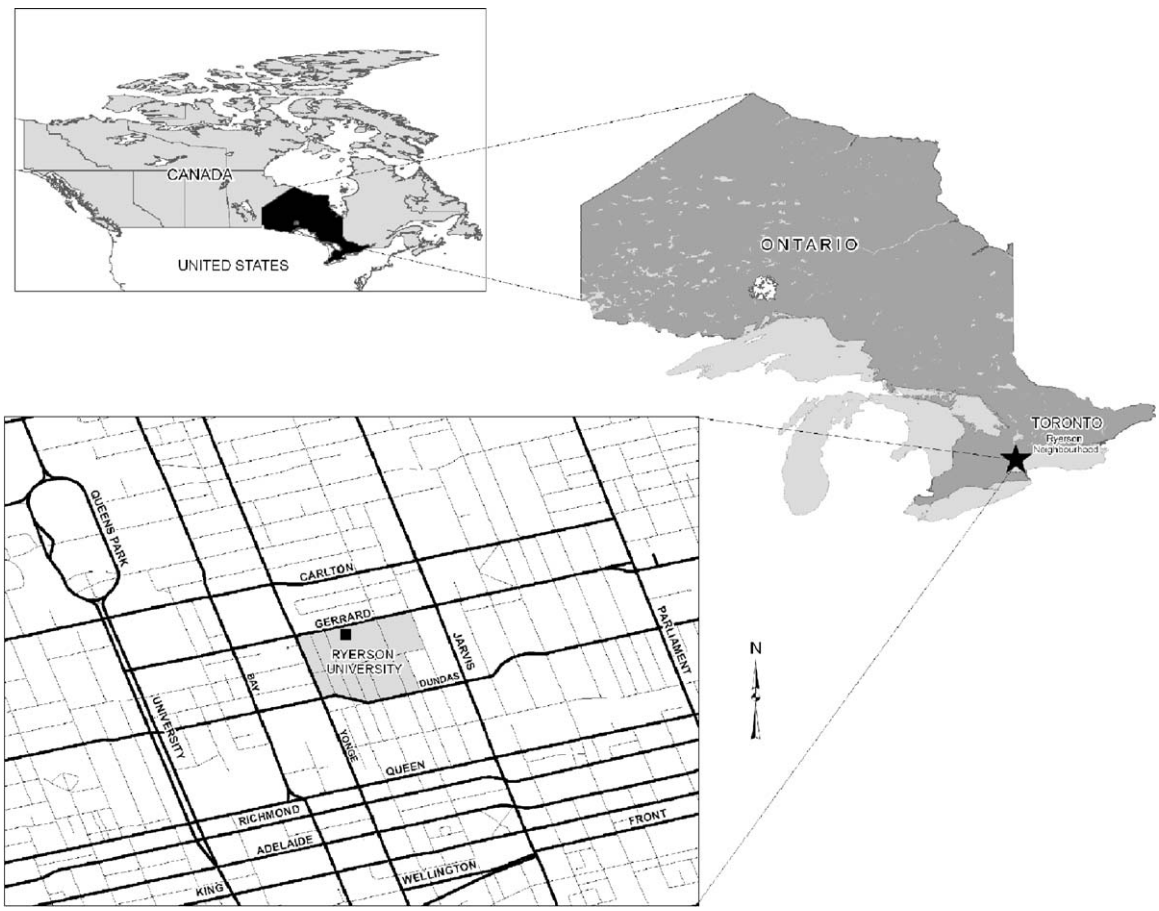

Fig. 1. Sampling location in downtown Toronto, Ontario, Canada.

automatically through a built-in permeation mercury source. The differences in the results generated from the two gold cartridges in the Tekran analyzer were found to be less then 2% during the study period.

2.3. Sampling

The whole sampling period can be divided into three seasons for seasonal distribution analysis. Summer was considered as 20 June through 19 September, while fall was 20 September through 19 December and winter was 20 December through 19 March.

During sampling, air was pulled into the analyzer from 3.5 m and 7 m, respectively, above ground in a canopy over a sidewalk. A solenoid valve (model 3-124A1, Furon, California, USA) controlled by a timer (model XT-4F, ChronTrol co., California, USA) was used to alternate the air flow from the two levels at 15 min intervals. A Teflon-coated membrane filter with 0.2 μm pore size and 47 mm diameter was placed in the sampling path prior to the analyzer to remove particulate matter in the air stream. The filter was replaced every 2 weeks. While the Tekran 2537A analyzer was set in continuous monitoring mode at $1.5\,L\,min^{-1}$ flow rate with 15 min sample integration time for each cartridge, the Gardis-1A^{+} was operated at a flow rate of $0.45\,L\,min^{-1}$ and in 15-min cycles, during which 11 min were used for sampling, 1 min for heating the Hg-collecting gold trap, 1 min for heating the analytical trap, and 2 min for cooling.

2.4. Meteorological parameters

Meteorological parameters (surface air temperature, wind speed, wind direction, and relative humidity) that were measured at Toronto island, which is about 4 km away from our sampling site, were obtained from Environment Canada. Only surface air temperature data were used in this study.

3. Results and discussion

The time series of GEM concentrations from both sampling levels are shown in Fig. 2. The interruptions occurred during certain periods are due to instrument maintenance. A large variability of the temporal distribution patterns between seasons as well as in the same season was detected. Frequent and high spikes of mercury concentration were observed during our study period, indicating the effect of anthropogenic sources at the sampling site and leading to high standard deviation values for both sampling levels. This phenomenon was previously observed in other studies (Carpi and Chen, 2002; Dommergue et al., 2002; Kim and Kim, 2000, 2002). Table 2 presents the statistical summary of our GEM results. The GEM concentration values ranged from 0.36 to $128.33\,ng\,m^{-3}$ for the lower sampling level and from 0.41 to $145.64\,ng\,m^{-3}$ for the upper sampling level. The average values during the experimental period (from 20 June 2001 to 28 February 2002) were $2.39 \pm 2.05\,ng\,m^{-3}$ for the lower sampling level and $2.57 \pm 2.39\,ng\,m^{-3}$ for the upper sampling level. Fig. 2c shows a good correlation ($y = 1.0678x$, $R^2 = 0.788$) between the concentrations from the two sampling levels. Canopy effect is suggested to contribute the difference between the two sampling levels (Lindberg et al., 1998). The highest seasonal average GEM value was observed in the summer months at $3.17\,ng\,m^{-3}$. The average value for the winter season was $2.16\,ng\,m^{-3}$ while the lowest value of $1.90\,ng\,m^{-3}$ was observed for the fall season. The higher values in summer might be a result of higher Hg emission from the surface due to higher temperature in the season. There was higher average GEM concentration in winter than in fall probably because of more energy consumption associated with heating in the winter season. Seasonal modulations show that the daytime average values are always higher than nighttime average values from both sampling levels and that the difference between the sampling levels during daytime is greater than that of nighttime (Fig. 3). Solar radiation leads to higher surface temperature and the higher level of biological activities during daytime, thus higher ground and foliar emission of mercury. Interestingly, the difference in Hg concentration between the two levels in winter was at the similar level as that in summer season. The reason for this is not understood at this point.

Probability distribution of GEM concentrations in downtown Toronto revealed that the majority of the GEM values lied in the range of 2–4 $ng\,m^{-3}$ for both sampling levels. The log normal distribution is apparently well applied to the result of our study. Four Canadian rural-affected sites in the CAMNet all had right-skewed probability distributions that approach log normality (Kellerhals et al., 2003). This matches the theoretical distribution of pollutant concentrations for a passive pollutant that is

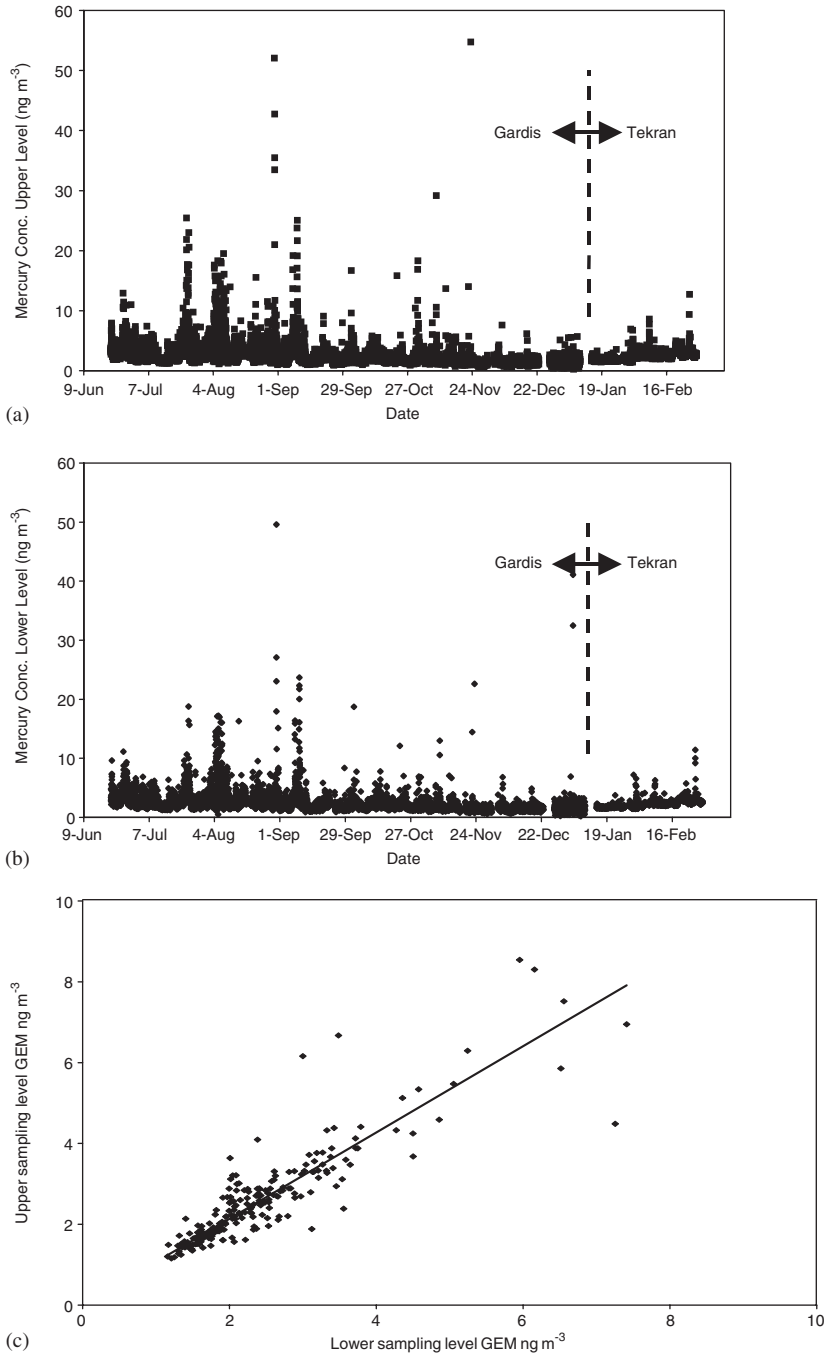

Fig. 2. Temporal variability of GEM concentrations in downtown Toronto Ontario, Canada, from June 2001 to February 2002: (a) upper sampling level, (b) lower sampling level, and (c) correlation between the upper and lower level (daily average).

emitted and then subjected to successive random dilutions (Ott, 1995). This log normal distribution pattern is observed in our study, suggesting GEM as a passive pollutant at our monitoring site with the effect of nearby anthropogenic sources by means of air mixing. In addition, the results show when the mercury concentrations are lower than $3\,ng\,m^{-3}$, the concentrations at the lower intake are higher than those at the upper intake, while when the concentrations are higher than $3\,ng\,m^{-3}$, the

Table 2
Statistical summary of quarter hourly interval GEM concentration (ng m^{-3}) in Toronto from June 2001–February 2002 and from the CAMNet (Kellerhals et al., 2003)

	All data		Seasonal data						Data from CAMNet (exclude Alert), 1997–1998	Data from CAMNet Alert, 1997–1998
			Summer		Fall		Winter			
	Lower level	Upper level	Lower level	Upper level	Lower level	Upper level	Lower level	Upper level		
N	10658	10628	4194	4174	3861	3852	2589	2588	6971–15866[a]	15021
Average	2.39	2.57	3.03	3.31	1.84	1.97	2.11	2.21	1.33–1.90[a]	1.55
Std. dev.	2.05	2.39	2.13	2.56	0.87	1.38	1.19	0.77	0.22–0.40[a]	0.39
Median	2.01	2.10	2.48	2.64	1.69	1.74	2.01	2.10	1.32–1.83[a]	1.59
Min.	0.36	0.41	0.47	0.46	0.67	0.64	0.36	0.41	<0.05	<0.05
Max.	128.33	145.64	49.62	145.64	22.59	54.76	128.33	12.71	10.19	3.04

[a]Range values for the sites (exclude Alert) in the CAMNet.

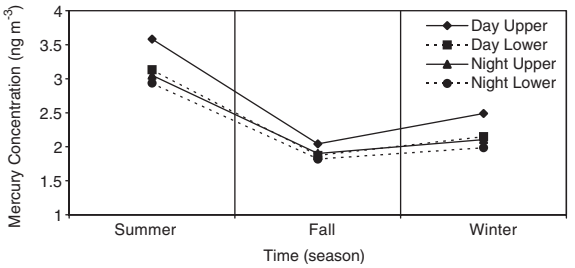

Fig. 3. Seasonal variation of average GEM concentrations modulated by daytime and nighttime in Toronto from June 2001 to February 2002.

concentrations are higher at the upper intake relative to the lower intake. This suggests that during the low concentration periods, the vertical concentration profile is mainly affected by surface emission while the high Hg concentrations are caused by nearby anthropogenic emissions.

Diurnal distribution of GEM for the time series was also analyzed in the study. The seasonal modulations of the diurnal distribution of GEM concentrations are presented in Fig. 4. The summer months displayed peaks of GEM concentration at 6 a.m. followed by at 11 a.m. for both the lower and upper sampling levels, while in the fall months, the peaks occurred at 9 a.m. for the lower sampling level and at 9 a.m. followed by at 11 a.m. for the upper sampling level. The winter season had the peak of GEM concentrations at approximately 12–1 p.m. for both the lower and upper sampling levels. This is a different diurnal pattern from other studies conducted in the mid-1990s in the United Kingdom (Lee et al., 1998) and in late 1990s in Asia (Kim and Kim, 2001, 2002) in which diurnal graphing showed highest GEM values during the nighttime. Our result was in general agreement with the results from CAMNet (Kellerhals et al., 2003). Reasons for higher GEM concentrations during the day could be related to the solar radiation. Solar radiation can lead to/enhance photochemical reduction of Hg^{2+} species, photo-induced biological processes (e.g., photosynthesis), and increase surface temperature. These factors, alone or in combination, could lead to the release of elemental mercury to the atmosphere (Feng et al., 2003; Poissant, 2000; Kim et al., 1995; Ariya et al., 2004). The comparison between the hourly temperature and corresponding GEM concentration suggested that ground emission of elemental mercury played a role in all the seasons (i.e., higher Hg^0 concentrations in daytime). The effect of photochemical reduction and biological

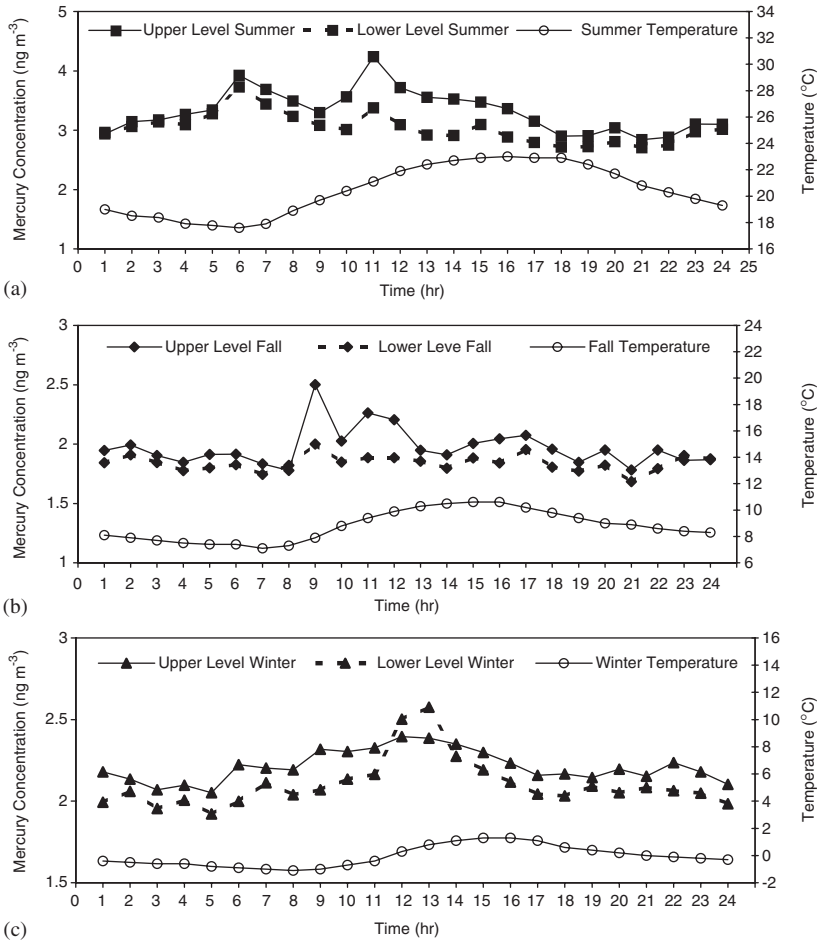

Fig. 4. Seasonal diurnal variability of GEM concentrations in downtown Toronto: (a) summer (20 June to 19 September 2001), (b) fall (20 September to 19 December 2001), and (c) winter (20 December to 28 February 2002) compared with seasonal average ambient surface air temperature (from Environment Canada).

processes is more pronounced in summer when solar radiation is more intense and the ground is not covered by snow. The sunrise time in summer at our sampling site was between 5:36 a.m. (20 June 2001) and 7:01 a.m. (19 September 2001), which correlated well with the first peak in Fig. 4a. When the second peak occurred, the difference between the upper level and the lower level was greater. According to Lindberg and co-workers (1998), although dynamic exchange of mercury occurs all the time over forests, whether the foliar surfaces function as a source or sink for mercury depends on many factors such as leaf temperature, leaf surface conditions, soil gas mercury level, atmospheric oxidants, and biological factors. Our data (Fig. 4a) suggested that stronger solar radiation around 11 a.m. in summer time enhanced the biological processes, leading to foliar emission of mercury. In winter season when the sunlight was less intense, the ground surface was covered with snow most of the time, and the level of biological activities was low (e.g., there were no leaves on the trees thus the photosynthesis ceased), the diurnal pattern of GEM concentrations is similar to those of the annual and seasonal surface air temperature. This suggests that the temperature-driven surface emission of mercury predominates in winter. As a result, the GEM concentrations at the lower level were higher than those at the upper level between 12:00 and 14:00 (Fig. 4c).

The results from this study and those from the CAMNet are compared in Table 2. The GEM concentrations from the CAMNet-Alert station in the Arctic are considered as background values and the mean concentration was reported as 1.55 ± 0.39 ng m^{-3} (Kellerhals et al., 2003). The GEM concentration of 2.48 ng m^{-3} (calculated as the average of

the values from the two sampling levels) in downtown Toronto is 60% higher than that from Alert and 55% higher than the average value ($1.60\,ng\,m^{-3}$) of the rural sites in CAMNet (1997–1998). It is worth to note that the standard deviation values were much higher from the measurements in Toronto, compared with those from the sites in CAMNet. The high standard deviation was a result of the effect of anthropogenic emissions of mercury in urban environment. Compared with the values collected from other urban areas worldwide (Table 1), the average value from Toronto is, in most cases, lower than those from other urban cities in North America. Data from Asian area, especially from China, are significantly higher. It is good to see the decrease of mercury in some Asian areas, such as Seoul, during the last decade (Kim and Kim, 2002). Intense campaigns were carried out across Europe during the past years (Schmolke et al., 1999; Munthe et al., 2003) to estimate the overall distribution of mercury species across Europe. Only few data, however, were reported on mercury concentrations in urban environment in Europe. The results in Table 2 show that the GEM concentrations in the urban atmosphere are characterized by high standard deviations. As discussed above, this statistical parameter basically shows the effects from human activities rather than poor reproducibility of the analytical instrument thus can be considered as a unique characteristic of urban atmospheric mercury concentrations.

4. Conclusion

This study revealed that the concentrations of GEM in the urban atmosphere (Toronto) are elevated, compared with those from rural and remote locations in Canada, and are highly variable, due to anthropogenic emission, canopy and urban, and temperature effects. Studies are needed to identify the sources of emission thus to develop emission control strategies. Information on mercury species in the urban atmosphere is urgently needed to assess the transport, transformation, deposition, and health effect of mercury.

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