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TAO, Vol. 15, No. 2, 261-267, June 2004

NOTES AND CORRESPONDENCE

The Signature of Diversification of Source Processes in Controlling Atmospheric Mercury Levels in East Asia

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(Manuscript received 3 October 2003, in final form 26 May 2004)

(Key words: Mercury, Anthropogenic, Emission, Source processes, Asia)

1. INTRODUCTION

The unique physicochemical properties of total gaseous mercury (TGM: primarily in its elemental form, Hg^o), characterized by strong stability, and long atmospheric residence time of approximately 1 yr, permit analysis of its temporal distribution patterns to be a potential indicator of air-quality change and associated source processes (Schroeder and Munthe 1998). In fact, large differences in concentration levels of Hg exist in many countries depending on their level of development and approaches to air quality management. In many Western countries, where significant efforts have been made in air quality control, the presence of moderately low Hg concentration values around 2.0 ng m^{-3} can readily be observed, even in locations adjacent to major metropolitan areas, Perch River area in New York, USA (Ames et al. 1998) or at certain source processes; for example, a coal-fired power station in Harwell, UK (Lee et al. 1998). This trend, however, contrasts sharply with what is observed in some Asian countries.

As many studies conducted on the Asian continent have focused on rapid urbanization, these differences in observed Hg concentration levels appear to be highly significant. For example, the presence of unusually high Hg concentrations, levels above 10 ng m^{-3} have been observed in a number of urban locations in China with relatively high frequency (Tan et al. 2000; Liu et al. 2002). In studies by Liu et al. 2002, the Hg values determined from eight different locations within Beijing City were highly variable depending on type of land-use and the temporal scale (seasonal and/or diurnal). Other Asian countries have also shared the presence of unusually high Hg data during different periods, such as Japan in the early 80s (Nakagawa 1995) and Korea in the late 80s (Kim and Kim 2000). Hence, the results of those

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studies confirm unique aspects of mercury's geochemical behavior on the Asian continent, especially in terms of its relationship with anthropogenic processes. In light of complexities involved in the Hg distribution patterns across the Asian countries, the signature of different Hg source processes is discussed in terms of its relationship with spatial and/or temporal variability.

2. RESULTS AND DISCUSSION

In Table 1, the results of many studies conducted in a number of important locations in Asia are tabulated. As Liu et al. (2002) measured temporal and spatial variabilities of Hg from eight different sites in Beijing city, their results may be used as a good reference point to contrast the effects of different anthropogenic or natural source processes under varying environmental settings. In Fig. 1, the measurement data of Liu et al. are hence plotted to compare both seasonal and diurnal distribution patterns among their selected study sites within the city of Beijing. To account for the relative enhancement in summertime Hg levels observed in one urban downtown (Tiananmen Square: TS) and one suburban industrial station (Shijingshan: SS), Liu et al. pointed out surface temperature as the controlling mechanism of Hg levels; this is important because the evaporation of surface-bound Hg can be accelerated with increasing

Table 1a. A statistical summary of the mean concentrations of atmospheric Hg determined from a number of monitoring sites located in the East Asian countries.

	Data Group	Industrial	Residential	Commercial	Rural	Remote
Korea	1		5.25[1)*]	5.34[2)]		4.47[3)]
	2		5.26[4)]	6.54[5)]		7.03[6)]
	3					3.15[7)]
Japan	1	31.6[8)]	10.3[9)]		5.73[10)]	3.4[11)]
	2		16.5[12)]		42.4[13)]	
China**	1	6.75[14)]	16.7[15)]	10.5[16)]	3.75[17)]	3.35[18)]
	2		8.47[19)]			

* For detailed information of superscripts, refer to reference numbers provided in Table 1b.

** Except for reference 18, the mean values for each individual area in China were derived by summing up various mean values representing either daytime or nighttime for each season.

Table 1b. Source information for the Hg data provided in Table 1a. All the reference nos. are identical to those given in Table 1a.

Ref. No.	Site	City/Province	Study period	Number of data*	Author
A. Korea					
1	residential	Kwa Chun	99~00	1992	Kim and Kim 2001b
2	Han Nam	Seoul City	99~00	2576	Kim and Kim 2001b
3	13 Mountains	Nationwide, Korea	87~93	32	Kim and Kim 1996
4	Yang Jae	Seoul City	99~00	11572	Kim and Kim 2001a
5	3 Terminals	Seoul City	Mar. 98	349	Kim and Kim 2001c
6	2 Mountains	Korea	97/98	358	Kim and Kim 2001c
7	Hari	Kang Hwa Island	01/02	323	Kim et al. 2003.
B. Japan					
8	urban	Chiba and two others	91~96/94	40	Nakagawa and Hiromoto 1997
9	urban areas	Chiba and three others	91~96/95	216	Nakagawa and Hiromoto 1997
10	suburban	Kushiro and two others	91, 94, 95	31	Nakagawa and Hiromoto 1997
11	Oceans	Japan sea/Pacific	91	4	Nakagawa and Hiromoto 1997
12	rural city	Hayama and two others	91~96, 95, 96	64	Nakagawa and Hiromoto 1997
13	Farmland	Tukui and two others	95/93	15	Nakagawa and Hiromoto 1997
C. China					
14	Shijingshan	Beijing	Feb. & Sept. 98	12 days	Liu et al. 2002
15	Xuanwu	Beijing	Jan. & Sept. 98	9 days	Liu et al. 2002
16	Tiananmen Sq.	Beijing	Feb. & Sept. 98	8 days	Liu et al. 2002
17	two rural sites	Beijing	Feb. & Sept. 98	9 days	Liu et al. 2002
18	Mountain sites	Guizhou	Unreported	122	Tan et al. 2000
19	residential	Beijing	Jan. & Feb. 98	15 days	Liu et al. 2002

* For data provided by Liu et al., all information is given as number of days.

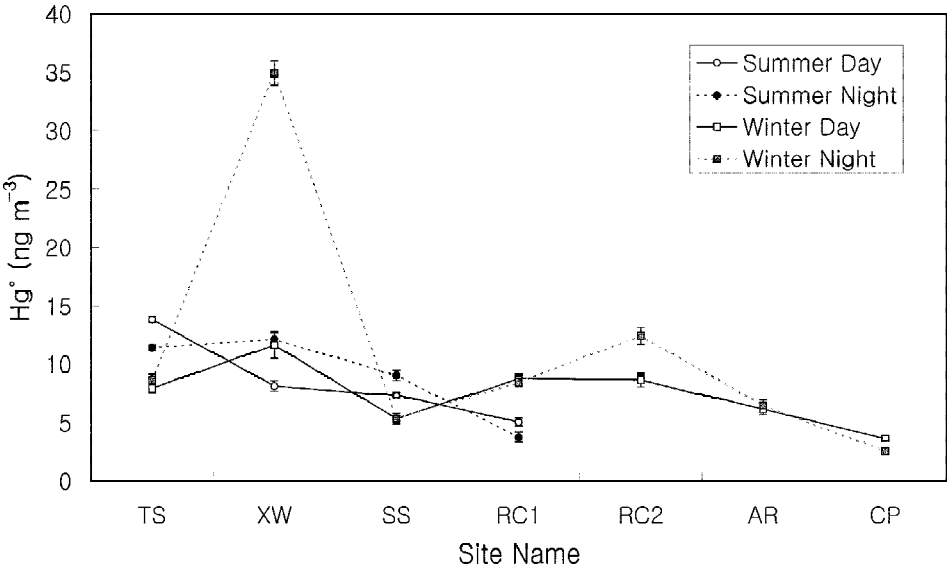

Fig. 1. Comparison of Hg concentrations determined from eight different locations of Beijing city, China (Liu et al. 2002). Results are plotted to compare changes across both seasonal (summer vs. winter) and diurnal scale (day vs. night). Error bars represent relative standard deviation. Actual names for each study site compared in this Figure are as follows: Tiananmen Square (TS), Xuanwu (XW), Shijingshan (SS), Research Center 1 (RC1), Research Center 2 (RC2), Atmospheric Research Institute (AR), Changping (CP), and Huairou County (HC).

temperature. However, based on the findings of enhanced Hg concentrations in the downtown area (TS) relative to the industrial area (SS), they suggested that anthropogenic processes were more prominent in the downtown area. To support their assertion, they quoted the results from a few urban areas in Asia wherein similar seasonal patterns were monitored: Seoul, Korea (Shon et al. 1993), and three urban sites in Tokyo/Sagami Bay, Japan (Nakagawa 1995). Temperature effect alone may have been important enough to explain the temporal patterns of Hg at those Beijing sites. This type of approach, however, is too simple to explain seasonal distribution patterns of Hg in urban areas, for there is a broad spectrum of anthropogenic processes.

It is in fact noted that a considerably different pattern exists simultaneously in a residential area of Beijing. Although these authors did not provide the full explanations for the different temporal patterns in the Xuanwu (XW) area, its results contrast sharply with those of the two other areas. The pattern at the XW site is characterized by the presence of an excessively high mean concentration recorded during a winter nighttime period (34.9 ng m^{-3}); the site clearly exhibited relative enhancement during winter as apposed to summer, and nighttime as

apposed to daytime levels. Although air pollution can be promoted under such meteorological conditions as winter and nighttime, the observed patterns in the residential area of Beijing are undoubtedly indicative of unusually strong anthropogenic source processes. It is striking to find that the patterns observed in the residential area of XW in Beijing are exceptionally compatible with what were seen in Seoul during the late 80s.

The previous studies of Kim and Kim (2000) offer some insights into the influences of anthropogenic processes on the distribution characteristics of TGM. They conducted a detailed analysis of the Hg data collected from several locations in Seoul during the late 80s, when the consumption of coal (e.g., anthracite) peaked. This research was continued further in the late 90s by reinvestigating the Hg levels at some of the previous study sites in Seoul (Kim and Kim 2002). A series of comparative analyses made over a decade demonstrated tight relationships between the Hg levels and anthracite consumption patterns. It was thus concluded that the presence of exceptionally high Hg concentrations, those above 20 ng m^{-3} should be an indicator of such strong source processes. The temporal patterns of Hg distribution under the influence of those source processes were in fact systematic enough to distinguish between seasons and between concentration ranges, as those exceptional values were observed most intensively during winter with the highest seasonal mean. The patterns, however, were reversed completely after excluding those exceptional, upper-bound concentration data so that spring (and summer) recorded the highest seasonal mean. Unlike the results seen in those previous years, there are many indications that the present temporal patterns in Seoul (in the late 90s) have changed significantly due possibly to diversification of source processes (Kim and Kim 2002). It should be addressed that the present time Hg, approximately 5 ng m^{-3} while being significantly lower than those levels observed in the late 80s (> 10 ng m^{-3}), exhibits indistinguishable differences in its seasonal mean values. Hence, one may expect that the effects of source processes, even anthropogenic ones, can vary through time.

It is well known that chlor-alkali plants belonged to the predominant anthropogenic source of Hg emissions in many industrial countries prior to 1970s (Schroeder and Munthe 1998). However, relative dominance of anthropogenic sources changed very rapidly with the implementation of various regulating controls. Along with changes in fuel consumption patterns, various man-made activities are pointed out to be important sources of Hg in urban air. For instance, it was reported that the gaseous Hg^o emissions from municipal solid waste (MSW) incinerators were one of the important sources of Hg (Sakata and Marumoto 2002); they found that such a Hg source could account for the dominant fraction of particulate Hg levels occurring in the Tokyo metropolitan area. The significance of a series of metal handling processes (such as refining, smelting, etc) has also been reported (Nriagu and Pacyna 1988).

Considering that the urbanization and industrialization of China are progressing rapidly along with notable increases in coal consumption (Xiao et al. 1998), the simple explanation of Liu et al. (such as the temperature effect) may not be sufficient in explaining the observed differences in urban areas of various characteristics. A line of evidence including an exceptionally wide range of the mean concentrations such as from 2.5 (winter nighttime in a rural area) to 34.9 ng m^{-3} (winter nighttime in a residential area) and diverse features of temporal variability in the city of Beijing suggests the possibility that the source processes of Hg in the urban areas of Asia are extremely diverse.

Acknowledgements This research was supported by a Korean Science and Engineering Foundation (KOSEF) Grant (R02-2002-000-00055-0).

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