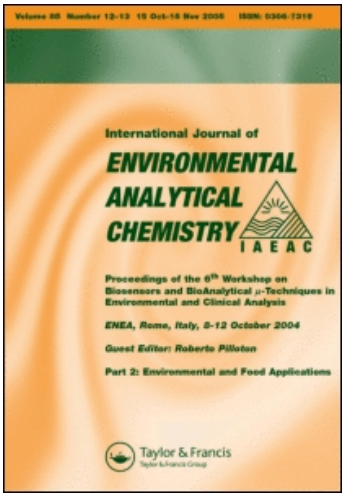

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VERTICAL DISTRIBUTION OF PARTICULATE MERCURY AS MEASURED ON A METEOROLOGICAL OBSERVATION TOWER (213 m)

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The concentration of mercury in airborne particles was determined in samples collected at heights of 1 m and at 175 m up the Meteorological Observation Tower (213 m) at the Meteorological Research Institute, Tsukuba between August 1985 and April 1986. The concentrations of particulate mercury at the two heights were compared with gaseous mercury levels at a height of 10 m and with other chemical components in the particles. The concentrations of particulate mercury at both heights showed no positive correlation with those of gaseous mercury, total suspended particles, sulfate, chloride, sodium or calcium. However, there was some positive correlation between particulate mercury and nitrate concentrations especially at 175 m; the concentrations of these components were found to be high when the wind direction was southwesterly. These results suggest that these components were transported from the Tokyo Metropolitan area.

KEY WORDS: Particulate mercury, airborne particles, vertical distribution, meteorological observation tower (213 m).

INTRODUCTION

The concentration of mercury in ambient air has been intensively studied by Nakagawa[1–3] throughout Japan to determine the geochemical cycles or environmental burden of anthropogenic and/or natural sources. Particulate mercury was found to account for up to 10% of total mercury.[3] The role of particulate

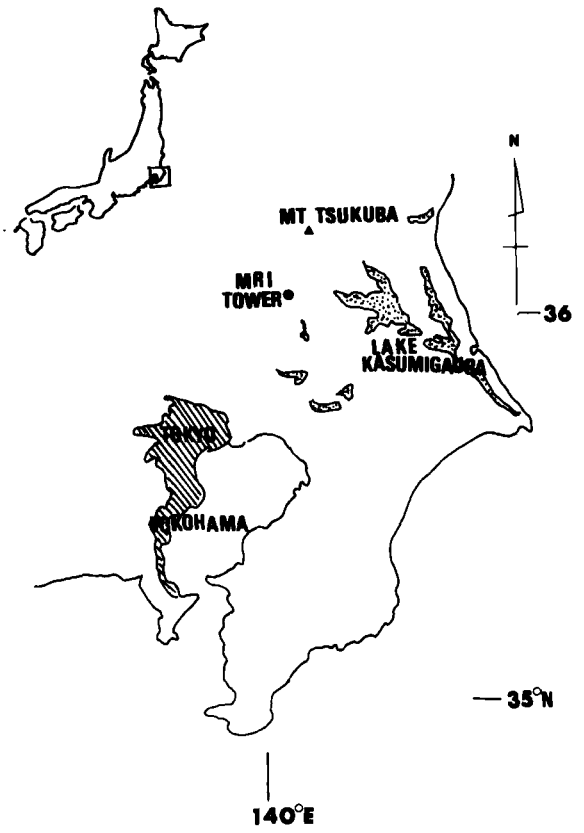

Figure 1 Location of the sampling site.

mercury, however, is as yet not well understood. Temperature has been suggested to be one of the most decisive factors in partitioning particulate and gaseous mercury, owing to the high vapour pressure of the element. Almost no information, however, is available for the vertical distribution of particulate mercury at remote sampling sites, nor the seasonal variations in particulate mercury levels.

In this study, in order to determine the vertical distribution of particulate mercury at various seasons, a simultaneous sampling by two high volume air samplers was performed at the 175 m and 1 m stages of the Meteorological Observation Tower (213 m) of the Meteorological Research Institute at Tsukuba City, from August 1985 to April 1986. The location of the sampling site is shown in Figure 1. The role of particulate mercury was discussed in relation to the meteorological conditions. It was also compared to those of other major chemical components of particulate matter as well as that of gaseous mercury in the surface air near the sampling site.

EXPERIMENTAL

Sampling of Suspended Particles

Two Shibata HV 1000 high volume air samplers were operated: one at the 175 m stage of the Meteorological Observation Tower and the other in the observation field (the height of the sampler was about 1 m) in the same campus of Meteorological Research Institute, which is located in Tsukuba City. Suspended particles smaller than 10 μm were collected on Gelman Microquartz filters. The sampling intervals were 3–5 days and the sampling air flow rate was 1000 l/min.

Sampling of Gaseous Mercury

The concentration of gaseous mercury was monitored on the roof (10 m high) of the National Institute for Environmental Studies, which is located about 300 m west of the tower. The sampling apparatus was similar to that reported by Matsumoto *et al.*,[4] based on a low volume ($\sim$0.5 l/min) air trap on gold wire after passing a water trap, followed by an atomic absorption determination.

Determination of Mercury in the Suspended Particles

A portion of the filter (1/8) was cut into small pieces and moderately mineralized in a long necked flask of volume 100 ml by adding H_2SO_4 (x mol/l, 2 ml) and HNO_3 (y mol/l, 8 ml) on a hot plate of 120 °C, for 2 h. The mineralization period was previously determined by using several portions from the same filter, which showed the same results for mineralizations of more than 1 h.

The digested solution was made up to 200 ml with demineralized water and then 20 ml of 5% (M/V) $SnCl_2$ was added to reduce mercury to atoms; the solution was then directly introduced to a quartz cell of pathlength 100 mm fixed on the atomizer of an atomic absorption spectrometer (Hitachi AAS 170–30) for the determination of the absorption at 253.7 nm. The sensitivity and the reproducibility of this method has been previously investigated with satisfactory results for the determination of mercury in airborne particles.

Determination of other Chemical Components in the Suspended Particles

The concentration of the major chemical components in the suspended particles collected on the filter was determined by ion chromatography, atomic absorption and emission spectrometry, the details of which have been reported elsewhere.[6]

RESULTS AND DISCUSSION

The results for mercury are shown together with other major chemical components

Table 1 Concentrations of mercury and other chemical components in airborne particles ($< 10\,\mu m$)

Sample		*Hg*	Cl^- (ng/m^3)	NO_3^-	SO_4^{2-}	NH_4^+	Ca^{2+}
0 meter							
Aug.	5–8, 1985	0.045	140	650	1670	8	120
	8–12	0.039	110	400	1500	3	70
	12–16	0.034	80	370	2420	40	80
	16–20	0.024	15	190	2190	70	60
	20–24	0.025	10	270	5600	1020	80
Oct.	1–4	0.103	200	2360	4360	850	320
	4–7	0.114	1040	9320	6300	4240	230
	7–12	0.071	109	1240	3170	520	200
	12–14	0.079	460	1050	2410	20	250
	14–18	0.082	120	2160	3270	540	200
Dec.	12–19	0.209	3910	5660	3810	2920	240
	19–23	0.149	5690	4070	3690	4320	240
	23–26	0.198	2050	2860	3060	2030	250
Jan.	16–20, 1986	0.089	3140	4860	5480	3060	220
	20–24	0.042	2450	4390	4110	2760	240
Mar.	31–Apr. 8	0.102	440	2360	5780	1910	500
Apr.	8–14	0.098	470	2560	5850	1330	830
	14–18	0.061	560	1470	2550	1150	100
	18–21	0.093	880	2520	4550	1450	180
	21–28	0.056	260	1530	3470	880	190
175 m							
Aug.	5–8, 1985	0.016	225	800	2010	5	130
	8–12	0.021	216	580	1630	2	70
	12–16	0.034	30	840	2970	140	90
	16–20	0.037	40	430	2770	20	70
	20–24	0.016	10	130	5920	1060	80
Oct.	1–4	0.133	70	1920	4940	890	180
	4–7	0.223	30	4490	5260	2190	230
	7–12	0.058	10	750	3030	340	90
	12–14	0.059	650	1260	2770	20	70
	14–18	0.089	90	340	3550	550	220
Dec.	12–19	0.127	280	3210	1950	1300	150
	19–23	0.068	840	2750	2990	1480	220
Jan.	16–20, 1986	0.042	640	1990	2590	90	180
	20–24	0.062	470	2320	2340	90	160
Mar.	31–Apr. 8	0.061	140	740	2360	530	290
Apr.	8–14	0.164	180	2510	3580	1150	300
	14–18	0.065	410	1290	2910	840	80
	18–21	0.029	880	1390	2640	1140	60
	21–28	0.039	450	2020	3950	1290	180

in Table 1. Figure 2 shows the seasonal variations in the concentrations of particulate mercury at heights 175 m and 1 m. As can be seen from Figure 2, the concentration of particulate mercury at 1 m was found to be highest in winter, lowest in summer, and intermediate in spring and autumn. Total suspended

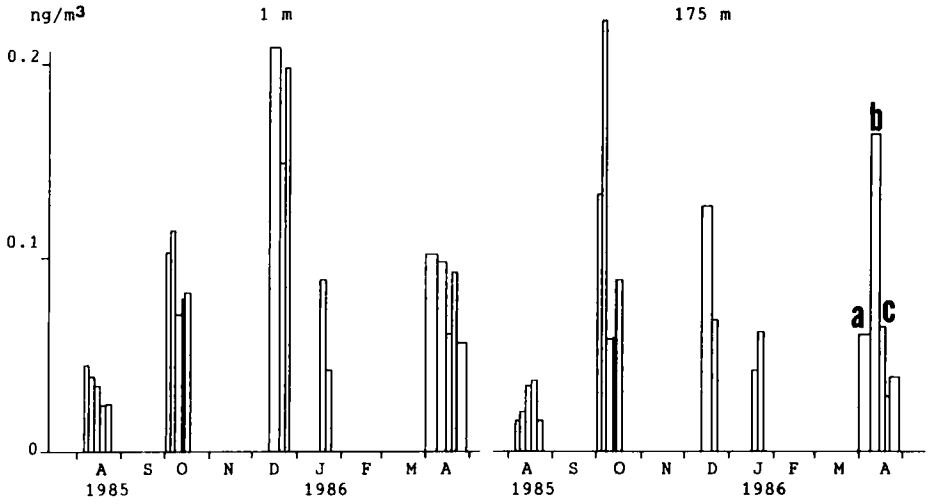

Figure 2 Concentration of particulate mercury at Tsukuba as measured on the Meteorological Observation Tower (213 m).

particles, and some major chemical components (such as sulfate) which were assumed to have no particular local sources, showed a similar tendency. At 175 m, however, the concentration of particulate mercury showed peaks higher in spring and autumn than in winter. The seasonal pattern of particulate mercury levels at 175 m was different from those of total suspended particles, particulate sulfate, chloride, and calcium. The pattern showed a similar tendency to that of nitrate.

At 1 m, the seasonal concentration pattern was similar for total suspended particles, sulfate, and calcium. These patterns showed high concentrations in winter (when the air was stable) and low concentrations in summer (when the air was comparatively well mixed). At 175 m, these patterns showed less typical seasonal variations. These data suggest that the stability of the air was one of the main factors determining the distribution of these components.[6] The concentration patterns of mercury and nitrate, however, were different from those of total suspended particles, sulfate, and calcium showing some high peaks at 175 m in spring and autumn sampling periods; these peaks could not be explained simply in terms of the stability of the air.

The concentration of gaseous mercury in surface air at Tsukuba was monitored at the same time, on the roof of the National Institute for Environmental Studies (NIES, about 10 m height); this building is located about 300 m west of the Meteorological Observation Tower. The daily concentrations of gaseous mercury are summarized in Figure 3, showing the average values for every seven days. Figure 3 indicates that the concentration of gaseous mercury was high in summer and lower in other seaons, but that seasonal differences were not as pronounced as those of particulate mercury. The data for the concentration of gaseous mercury in October 1985 were not available, because the monitoring program was set at a different site. Generally the concentration of gaseous mercury near the surface was

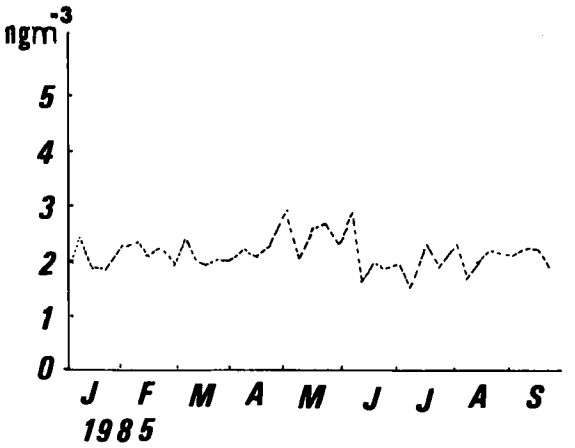

Figure 3 Concentration of gaseous mercury at Tsukuba as measured on the roof of NIES building (~10 m).

Table 2 Correlation coefficients between particulate mercury and other components

	TSP	Cl^-	NO_3^-	SO_4^{2-}	NH_4^+	Ca_a^{2+}	*Hg(gas)*[a]
1 m	0.586	0.661	0.580	0.251	0.600	0.348	−0.523
175 m	0.465	−0.221	0.763	0.486	0.037	0.599	(−0.497)

[a]Measured on the roof of NIES building (~10 m).

about 2 ng/m^3 (range 0.9–2.5 ng/m^3); i.e. about 10 to 50 times the concentration of particulate mercury. This concentration of gaseous mercury is comparable to that of unpolluted areas of Japan as reported by Nakagawa.[2]

The correlation coefficients between particulate mercury and other components are shown in Table 2. Only nitrate showed some correlation with mercury at 175 m. Figure 4 shows the relation between particulate mercury and nitrate, at 175 m. Figure 5 shows the relation between particulate and gaseous mercury. Data for gaseous mercury were obtained only on the roof of NIES building, so Figure 5 shows the relationship between the gaseous mercury and particulate mercury at 1 m. No positive correlation was found between these two forms of mercury; in fact a somewhat negative correlation was found between the two parameters, due to the higher concentration of gaseous mercury and lower concentration of particulate mercury in summer. These results may indicate some gas to particle conversion, depending upon the ambient temperature, but the conversion rate is estimated to be slow.

In our recent work, high concentrations of particulate nitrate at Tsukuba have been found in some cases to correlate with strong southwesterly winds, suggesting that polluted air from the Tokyo Metropolitan region, including the Keihin–Keiyō industrial area, might be the main source.[6]

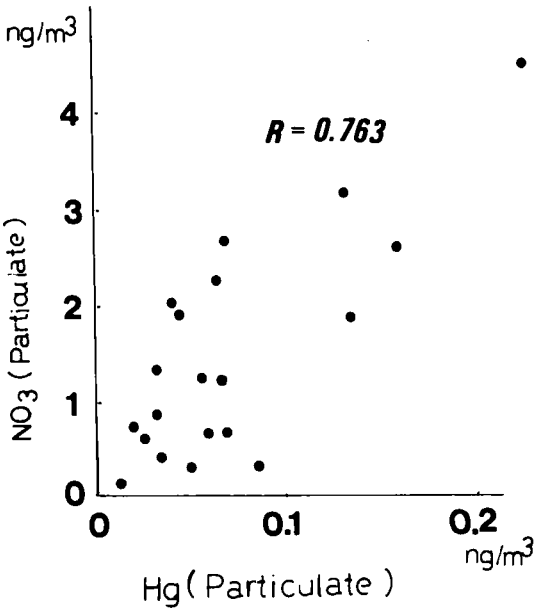

Figure 4 Relation between particulate mercury and particulate nitrate at 175 m.

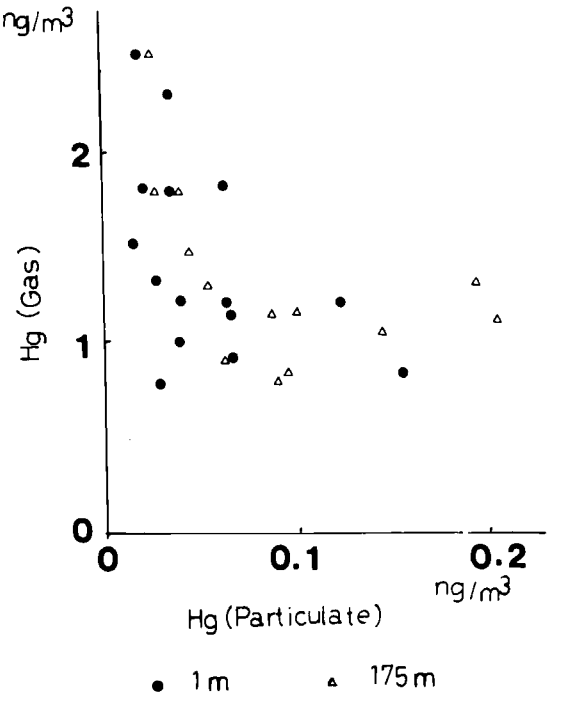

Figure 5 Relation between particulate and gaseous mercury.

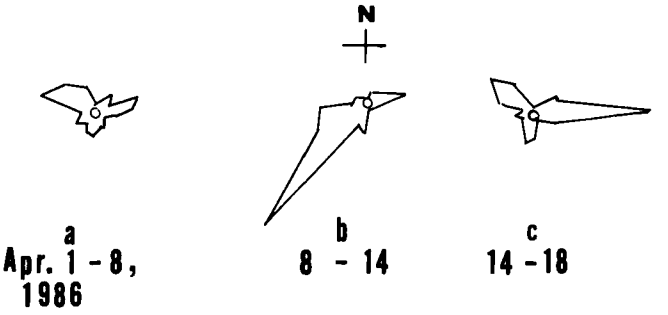

Figure 6 Frequency distribution of wind direction at 200 m in April 1986. Note a, b and c correspond to the peaks a, b and c in Figure 2.

The wind direction and speed at 200 m height was examined in relation to incidences of high concentrations of particulate mercury and nitrate. As an example, Figure 6 shows the frequency distribution of wind direction for each sampling period in April 1986. Figure 6 clearly shows that southwesterly winds predominated in the period April 8–14, when the concentration of particulate mercury and nitrate was high (Figure 2). The concentration of gaseous mercury at the surface, however, remained almost constant, in the range 0.9–1.2 ng/m^3, in this sampling period, indicating that this difference might not be caused by gas to particulate conversion of mercury at the sampling site.

These results suggest that particulate mercury might be transported from a polluted area by the local wind system. The chemical or physicochemical conversion of gaseous to particulate mercury, however, should be investigated further and attempts made to determine the rate constants. These reactions may take place in the course of transportation of mercury in air which has a high concentration of suspended particles together with a high concentration of nitrate or other chemically active matter.

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