

## 4.5 Quantifying Sources of Mercury Using Hg/CO Ratio in Gaseous Substances Observed at the MFRS

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### 1. Introduction

Mercury, the only metal element that is liquid at ambient temperature and pressure, has a low boiling point (357 °C). Therefore, mercury evaporates easily under ambient conditions and is readily emitted from various sources. In 2010, the majority of global anthropogenic emissions of mercury to the atmosphere were associated with artisanal and small-scale gold mining (37.1%) and stationary combustion of fossil fuels (24.7% total; 24.2% from coal combustion) (UNEP, 2013).

### 2. Atmospheric mercury

In the atmosphere, mercury is assumed to be in the form of gaseous elemental mercury (GEM, Hg(0)), reactive gaseous mercury (RGM, Hg(II)) or gaseous oxidized mercury (GOM, Hg(II)), and particle-bound mercury (Hg(p)). The sum of GEM and RGM is commonly known as total gaseous mercury (TGM). In terms of relative abundance, Hg (0) accounts for more than 95% of atmospheric mercury, while Hg (II) and Hg (p) make up the remainder (Lindqvist et al., 1991).

Hg (0) released to the atmosphere is oxidized and settles out of the atmosphere as Hg (II) and Hg (p), eventually ending up in waterways or water bodies. It is then methylated as a result of the metabolic activity of microorganisms and bioaccumulated through the food chain. For this reason, there are serious concerns about the ecological impact of mercury on humans and wildlife.

According to regional estimates by the United Nations Environment Programme (UNEP), East and Southeast Asia account for 39.7% of the global anthropogenic emissions of mercury. Because the Japanese archipelago is downwind of these regions, Japan is strongly affected by the long-range transport of mercury from East and Southeast Asia. Nagafuchi (2000) has already pointed out the adverse health and environmental effects of air pollution in the Kyushu region of Japan. Hence,

understanding the dynamics of long-range transport of mercury is important. However, relatively few studies of the long-range transport of mercury have been conducted in Japan.

### 3. Japanese monitoring sites

In Japan, air pollutants have been monitored at 158 sites since 1997. Sampling is conducted for 24 h once a month at each of the sites. The sites include general environmental sites (111 sites), roadside sites (30 sites), and emission source sites (17 sites). Results from roadside and emission source sites, which account for approximately 30% of the sampling sites, may be affected by regional pollution sources. It is difficult to exclude the impact of local sources at monitoring sites. Therefore, in evaluating the long-range transport of mercury, it is important to select monitoring sites where the effects of regional sources are minimal.

Monitoring at mountainous sites is well suited for evaluating the long-range transport of mercury because (1) there are no nearby pollution sources, (2) the vertical profile of atmospheric mercury can be determined, and (3) atmospheric air in the free troposphere is not easily affected by friction against the ground and air masses that are easily transported at mountainous sites. Therefore, atmospheric Hg concentration has been measured around the world in this way.

### 4. Observation at Mt. Fuji

As such, monitoring at mountainous sites plays an important role in the evaluation of long-range transport of mercury. Here, we provide a summary of the results of monitoring on Mt. Fuji (3776 m). Given that most mountains do not offer access to commercial power, mountain monitoring of mercury typically relies on active or passive supply of electricity through solar batteries or fuel cells. In the case of Mt. Fuji, however, we were able to carry out continuous mercury monitoring using commercial power (summer only).

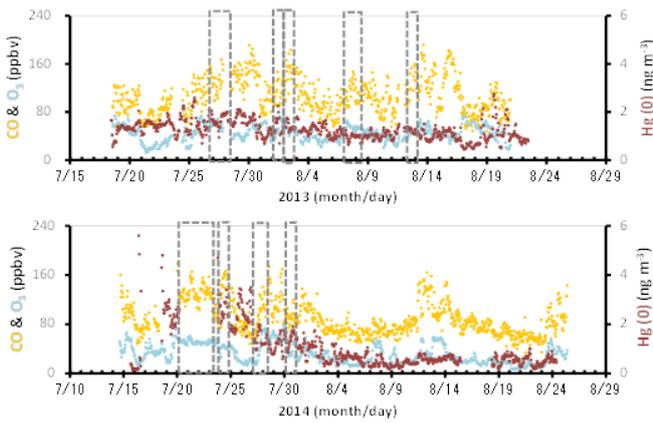


Fig. 4.5-1: Change in Hg(0), CO, and ozone concentrations during the monitoring periods in 2013 and 2014 at Mt. Fuji

The Hg/CO ratio was used to determine whether the mercury was anthropogenic in origin or due to biomass burning. As can be seen in Table 4.5-1, the  $\Delta \text{Hg}(0) / \Delta \text{CO}$  ratios measured on Mt. Fuji in 2013 and 2014 were one order of magnitude higher than those measured in Okinawa and Mt. Bachelor (Oregon, US). Although a simple comparison of results is not possible given that the  $\Delta \text{Hg}(0) / \Delta \text{CO}$  ratios for Okinawa and Mt. Bachelor are based on year-round monitoring, the results demonstrate the possibility of using data from Mt. Fuji to identify contaminant sources. Mercury emissions calculated via  $\Delta \text{Hg}(0) / \Delta \text{CO}$  ratios and CO emissions from Asia were substantially higher than previous estimates.

Table 4.5-1:  $\Delta \text{Hg}(0) / \text{CO}$  ratios for Mt. Fuji in 2013 and 2014, and a comparison with previous values

Emission type	Source region	Measured	CO emission,	GEM emission,	Previous reported		
		TAM/CO, this work (mol mol <sup>-1</sup> )	from literature (mol y <sup>-1</sup> )	this work (metric t y <sup>-1</sup> )	Total Hg	GEM	
Anthropogenic industrial	China	$5.16 \times 10^{-7}$	$6.00 \times 10^{12}$	620	536 604	300 320	Steets et al., 2005 Pacyna et al., 2006
Biomass burning	Global	$1.36 \times 10^{-7}$	$24.6 \times 10^{12}$	670	600 1000	—	Pacyna et al., 2006, Andreae and Merlet, 2001 Brunke et al., 2001
Biomass burning	Boreal forests	$1.07 \times 10^{-7}$	$1.07 \times 10^{12}$	29 Alaska 168 all boreal forest.	22.5 Global boreal 53-341		Sigler et al., 2003 Turetsky et al., 2006
Anthropogenic industrial	China	$11.2 \times 10^{-7}$	$6.00 \times 10^{12}$	1350			This study

\*Table 4.5-1 is modified from Peter Weiss-Penzias (2007)

Figure 4.5-1 shows the change in Hg (0), CO, and ozone concentrations during the monitoring periods in 2013 and 2014 at Mt. Fuji. The values in the figure show the impact of pollution events as determined according to criteria proposed by Weiss-Penzias et al. (2007) and Jaffe et al. (2005). Specifically, a change was deemed a contamination pollution event if (1) CO enhancement was more than 15% above the mean for the survey period for at least 12 hours, (2) there was enhancement in at least one other species (O<sub>3</sub>, Hg), and (3) an ensemble of back trajectories showed a consistent pattern of transport from one of the high Hg emission sources on the Asian continent.  $\Delta \text{Hg} / \Delta \text{CO}$  ratios were calculated for the 9 pollution events identified as satisfying any of the above criteria.

## 5. References

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