Four-year Measurements of Wet Mercury Deposition at a Tropical Mountain Site in Central Taiwan

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ABSTRACT

Rainwater samples were collected between 2010 and 2013 at Lulin Atmospheric Background Station (LABS) to study the distribution and characteristics of wet mercury (Hg) deposition, and possible driving mechanisms. Sample Hg concentrations ranged from 0.8 to 35.1 ng L⁻¹ with an overall volume-weighted mean (VWM) concentration of 9.2 ng L⁻¹. Annual wet Hg deposition fluxes ranged between 24.4 and 48.9 µg m⁻² with a mean value of 32.3 µg m⁻². This mean annual wet flux was about 1.5–6.0 times the values measured at 15 sites in the U.S. and 4–16 times the values reported from mountain and high-elevation sites in China. Both rainwater Hg concentration and rainfall amount contributed to this geographical difference, but rainfall amount played a more important role. This indicated that tropical mountains in East and Southeast Asia, especially the windward maritime slopes with abundant rainfall, could be hot spots of wet Hg deposition. Wet Hg deposition flux was high in summer because of elevated rainwater Hg concentrations and high rainfall. The seasonal pattern of rainwater Hg concentrations was different from that of the East Asian air pollutant export, indicating other factors, e.g., rainfall type, were also influencing rainwater Hg concentrations. A clear difference in seasonal frequency distribution of rainfall types was observed, with rain events associated with the Pacific high pressure type (PH) occurring more frequently in summer months. PH rainfall type had the highest VWM Hg concentration (13.5 ng L⁻¹), 2.3–6.2 ng L⁻¹ greater than those of the other rainfall types. Because of intense surface heating under summer PH conditions, precipitation systems usually form locally due to strong convection, resulting in afternoon shower. Therefore, the elevated rainwater Hg levels in summer at LABS were likely due to the scavenging of free tropospheric gaseous oxidized Hg (GOM) by deep convection.

Keywords: East Asia; Rainwater; Rainfall type; Scavenging.

INTRODUCTION

Mercury (Hg) is a persistent and bioaccumulative toxicant of global concern with widespread anthropogenic emission sources (UNEP, 2013). The most important source of inorganic Hg to many terrestrial and aquatic ecosystems is deposition from the atmosphere, especially in remote areas and the open oceans where local anthropogenic Hg emission sources are scarce (Mason and Sheu, 2002; Keeler *et al.*, 2005; Sunderland *et al.*, 2007; Mason, 2013). Both wet and dry deposition processes are important (Zhang *et al.*, 2017). However, compared to dry deposition, sampling and quantification of wet deposition are more accurate and hence more popular worldwide (Cole *et al.*, 2014; Weiss-Penzias *et al.*, 2016; Wright *et al.*, 2016; Sprovieri *et al.*, 2017).

Measurements of wet Hg deposition have been conducted at sites in major continents, such as North America (Civerolo *et al.*, 2014; Prestbo and Gay, 2009), South America (Fostier *et al.*, 2000; Hansen and Gay, 2013), Europe (Fu *et al.*, 2015; Sprovieri *et al.*, 2017), South Africa (Brunke *et al.*, 2016), and East Asia (Sakata and Marumoto, 2005; Seo *et al.*, 2012; Sheu and Lin, 2013; Fu *et al.*, 2016; Nguyen *et al.*, 2016). For instance, wet Hg deposition fluxes ranged from 3.0 to 25.0 µg m⁻² yr⁻¹ at various Mercury Deposition Network (MDN) sites over the U.S. between 1996 and 2005, and higher wet Hg fluxes were observed along the Gulf Coast (Prestbo and Gay, 2009). Wet Hg deposition fluxes of 5.8–17.7 µg m⁻² yr⁻¹ have been reported from 10 sites across Japan (Sakata and Marumoto, 2005). Fu *et al.* (2016) reported annual wet-only Hg deposition fluxes ranging from 1.8 to 7.0 and from 13.4 to 56.6 µg m⁻² for remote and urban sites in China, respectively. Sheu and Lin (2013) reported an annual wet Hg deposition flux of 10.2 µg m⁻² to Pengjiayu, a remote islet in the Northwest Pacific Ocean. However, most of these measurements were conducted in the temperate region. Measurements conducted in the tropical region are not as common.

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Measurement and model studies suggested that the tropical areas could be hot spots for wet Hg deposition because of the high rainfall (Hansen and Gay, 2013; Shanley *et al.*, 2015; Horowitz *et al.*, 2017). For instance, Shanley *et al.* (2015) measured high wet Hg deposition of $27.9 \mu\text{g m}^{-2} \text{yr}^{-1}$ at a tropical wet forest site in Puerto Rico, which was driven in part by high rainfall (2855 mm yr^{-1}). This suggested that the other tropical areas, where high rainfall occurs, maybe hot spots for wet Hg deposition as well (Shanley *et al.*, 2015). The average rainfall over East and Southeast Asia tropical climate zone is approximately 2000 mm yr^{-1} and could be greater than 3000 mm yr^{-1} on windward maritime slopes (Celâl Şengör *et al.*, 2019). However, wet Hg deposition measurements are scarce in the tropical region of East and Southeast Asia (Sprovieri *et al.*, 2017; UN Environment, 2019). Therefore, measurement data of Hg wet deposition to the tropical region of East and Southeast Asia are crucial to improve our understanding of the regional and global Hg cycles (Horowitz *et al.*, 2017; Sprovieri *et al.*, 2017).

Wet Hg deposition could be affected by several factors, including rain depth, atmospheric chemistry and transport, distribution of emission sources, and rainfall types (Gratz *et al.*, 2009; Holmes *et al.*, 2016; Kaulfus *et al.*, 2017; Mao *et al.*, 2017a, b). Meteorological conditions, such as rain depth and rainfall types, have been demonstrated to play important roles in controlling wet Hg deposition (Gratz *et al.*, 2009; Sheu and Lin, 2013; Shanley *et al.*, 2015; Mao *et al.*, 2016). Annual and interannual patterns of wet Hg deposition were found to be determined by rainfall amount (Gratz *et al.*, 2009; Mao *et al.*, 2016). Besides, the occurrence of deep convection has been suggested to contribute to the elevated rainwater Hg concentration in summer (Sheu and Lin, 2013; Shanley *et al.*, 2015). Recent studies also indicated

that thunderstorm, a rainfall type due to strong convection, can increase rainwater Hg concentration as well as wet Hg deposition over the U.S. (Holmes *et al.*, 2016; Kaulfus *et al.*, 2017). For example, Holmes *et al.* (2016) found thunderstorm can increase rainwater Hg concentration by 50% as compared to other rainfall types. These were attributed to scavenging of upper-altitude GOM by deep convection (Selin and Jacob, 2008). Results of these studies indicated the connection between wet Hg deposition and rainfall type.

The relationship between anthropogenic Hg emissions and wet Hg deposition has been documented by numerous studies (e.g., Fu *et al.*, 2016; Mao *et al.*, 2016; Weiss-Penzias *et al.*, 2016; Sprovieri *et al.*, 2017). Asia is the greatest contributor of anthropogenic Hg to the atmosphere (UNEP, 2013; Zhang *et al.*, 2016b) and the exports of East and Southeast Asia Hg emissions have been identified in the downwind regions (Friedli *et al.*, 2004; Chand *et al.*, 2008; Sheu *et al.*, 2010; Sheu and Lin, 2011; Sheu *et al.*, 2013; Jen *et al.*, 2014; Chen *et al.*, 2016; Nguyen *et al.*, 2019; Sheu *et al.*, 2019). In addition, various modeling studies indicated that Hg emissions from East Asia significantly contribute to Hg deposition over the downwind regions (e.g., Lin *et al.*, 2010; Chen *et al.*, 2014; Chen *et al.*, 2018). Therefore, atmospheric Hg deposition studies conducted in the downwind regions of East Asia are valuable for verifying the modeling results and thus contributing to improve our understanding in atmospheric Hg cycling.

Taiwan is located between the East and Southeast Asia continent and the west Pacific Ocean (Fig. 1). In this region, southwest monsoon prevails in summer, which brings a large amount of moisture, whereas northeast monsoon prevails between late fall and spring (Sheu *et al.*, 2010; Sheu and Lin, 2011; Lin *et al.*, 2013). In this study, we reported the four-year (2010–2013) measurements of wet

Fig. 1. Location of Taiwan and Lulin Atmospheric Background Station (LABS).

mercury deposition at the Lulin Atmospheric Background Station (LABS), a tropical mountain site in central Taiwan. The primary objective of this study is to characterize the wet Hg deposition to a tropical mountain site with high rainfall. The seasonal pattern of wet Hg deposition and the association of wet Hg deposition with rainfall type are also discussed, and possible driving mechanisms are discussed. Results of this research also contribute to an improved understanding of the influence of East and Southeast Asian Hg emissions to the wet Hg deposition in the downwind mountain areas.

SITE AND METHODS

Site Description

Lulin Atmospheric Background Station (LABS; 23.47°N 120.87°E, 2862 m above sea level) is a two-story building on the summit of Mt. Front Lulin in Yu-Shan National Park in central Taiwan (Fig. 1). LABS was established in late 2005 and its official operation began since April 2006 to study the impact of regional and long-range transported air pollutants on the mountain environment and ecosystems in Taiwan (Sheu *et al.*, 2010). There are no significant anthropogenic emission sources of Hg at the summit or in the surrounding areas. LABS is unique because its location and altitude can complement the global network of Global Atmospheric Watch (GAW) in the East Asia region where no high-elevation baseline station is available (Sheu *et al.*, 2010). According to the meteorological information collected at LABS from January 2007 to December 2017, the overall mean temperature is 10.5°C with the highest mean temperature in July (14.1°C) and the lowest in January (5.6°C). The average annual rainfall is 3178 mm, with about 50% occurring in summer (1582 mm, June–August) because of typhoons and thermal convection induced afternoon showers.

Rainwater Sampling and Hg Analysis

Collection of rainwater samples at LABS for total Hg analysis began since 2009 and is still continuing. In this study, the first complete four-year (2010–2013) of data were reported and analyzed for characterizing the wet Hg deposition at LABS. Weekly rainwater samples were collected using an automated wet-only rain collector and an acid-cleaned sampling apparatus (Landis and Keeler, 1997; Sheu and Lin, 2013). The apparatus comprised of a glass funnel connected to a 1L Teflon bottle by a Teflon adaptor. A glass vapor lock was used to prevent Hg evaporation from rainwater after collection. Bottles, glassware, and adaptors were acid-cleaned, rinsed with deionized water, and then air-dried before use. Samples were retrieved and the sampling apparatus was changed every Tuesday at 9 am. Samples were double-bagged before being shipped back to National Central University (NCU) for Hg analysis. Total Hg in unfiltered samples was analyzed by dual gold trap amalgamation and cold vapor atomic fluorescence spectrometry (CVAFS) after BrCl oxidation, hydroxylamine hydrochloride neutralization, and SnCl₂ reduction (U.S. EPA, 2002; Sheu and Lin, 2013). To assure

the data quality, various QA/QC procedures were routinely performed, such as duplicate analysis, ongoing precision and recovery, matrix spike, bottle blanks, transport blanks, and field blanks. Results of these QA/QC procedures well met the criteria requested by the US-EPA Method 1631, Revision E (U.S. EPA, 2002). Details of the cleaning procedure, rainwater samples analysis, QA/QC procedures can be found in Sheu and Lin (2013).

Between 2010 and 2013, amounts of annual rainfall ranged between 3173 and 3736 mm, with an average of 3421 mm (Tables 1 and 2). A total of 150 samples were collected, representing around 98% of the total rainfall. Missing samples occurred mainly due to system malfunction or very low weekly rainfall, resulting in no or insufficient rainwater amount for Hg analysis. Summer rainfall (mean = 1703 mm) contributed about 50% of the annual rainfall.

Wet Hg Deposition Flux

Weekly wet Hg deposition flux was defined as the product of the sample Hg concentration and the weekly rain depth. Monthly and yearly wet deposition fluxes were estimated by using monthly/yearly VWM Hg concentration and monthly/yearly cumulative rain depth.

Backward Trajectory Analysis

Backward trajectories (BWTs) were used to identify the origins and transport paths of air masses arriving at LABS. BWTs were calculated using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Draxler and Rolph, 2013) with meteorological data from the NCEP Global Data Assimilation System. Five-day backward trajectories arriving at 2862 m above sea level (i.e., LABS altitude) at 8 am local time were computed for each day in 2010–2013.

RESULTS AND DISCUSSION

Rainwater Hg Concentrations

Rainwater Hg concentrations ranged from 0.8 to 35.1 ng L⁻¹ (Table 1), with an overall VWM concentration of 9.2 ng L⁻¹. There was a weak but statistically significant inverse correlation between the rainwater Hg concentration and weekly rainfall (Fig. 2; $R^2 = 0.036$, $p = 0.022$), indicating rainwater Hg concentrations decreases with increasing rainfall. This result is similar to previous studies that also observed a significant inverse correlation between rainfall and rainwater Hg concentration (Prestbo and Gay, 2009; Seo *et al.*, 2012; Wang *et al.*, 2012; Hansen and Gay, 2013; Sheu and Lin, 2013). This was possibly due to the scavenging of atmospheric oxidized Hg (Hg(II)) to rainwater during the initial period of a rain event and the dilution effect caused by substantial rainfall (Selin and Jacob, 2008; Seo *et al.*, 2012; Sheu and Lin, 2013).

Table 2 summarized the VWM Hg concentrations in rainwater and wet Hg fluxes measured at LABS and numerous sites worldwide with different site characteristics. The VWM Hg concentration of LABS (9.2 ng L⁻¹) was close to the values of various rural and remote sites, such as Puerto Rico (9.8 ng L⁻¹) and Pengjiayu, Taiwan (8.9 ng L⁻¹),

Fig. 2. Relationship between rainwater Hg concentration and rainfall.

much lower than the urban sites, such as Kathmandu, Nepal (18.3 ng L^{-1}), Seoul, Korea ($10.1\text{--}16.3 \text{ ng L}^{-1}$), and Chongqing, China (30.7 ng L^{-1}), but higher than the values reported from mountain or high-elevation sites in China (Table 2; $3.7\text{--}7.7 \text{ ng L}^{-1}$). The value of LABS was approximately 2.4 times the values of Mt. Leigong (4.0 ng L^{-1}), Mt. Damei (3.7 ng L^{-1}), and Mt. Ailao (3.7 ng L^{-1}), and was about 1.3 times the values of Mt. Changbai (7.4 ng L^{-1}) and Mt. Waliguan (6.9 ng L^{-1}). It should be noted that this comparison may be affected by the mismatch in monitoring time and duration. Difference in rain depths among regions is often considered as an important factor to influence the rainwater Hg concentration because of the below-cloud scavenging and dilution effects (Prestbo and Gay, 2009). However, in spite of the fact that annual rainfall at LABS (3421 mm) was significantly greater than that of the mountain sites in China ($290\text{--}1931 \text{ mm}$), the VWM Hg concentration at LABS was still higher than those of the Chinese mountain sites (Table 2). These results, therefore, suggested that other factors, such as meteorological conditions, rainfall types, atmospheric chemistry, and Hg emissions and transport (Prestbo and Gay, 2009; Shah *et al.*, 2016; Mao *et al.*, 2017a, b; Shah and Jaeglé 2017; Sprovieri *et al.*, 2017) could also contribute to the difference in rainwater Hg concentrations between the LABS and the Chinese mountain sites. In summary, these comparisons indicated that the LABS rainwater Hg levels were higher than those of the mountain sites in China, but were lower than those of urban sites.

Wet Hg Deposition Fluxes

Weekly wet Hg deposition fluxes ranged from 0.01 to $7.6 \mu\text{g m}^{-2}$ in 2010–2013, with a mean weekly wet deposition flux of $0.82 \mu\text{g m}^{-2} \text{ week}^{-1}$. The greatest rainfall event occurred during 06/05/2012–06/26/2012 with a cumulative rainfall up to 812 mm (639 mm during 06/05–06/12). This

event was caused by the combination of the southwest monsoon, Meiyu rainfall, and typhoon TALIM (06/19–06/21). Rainwater Hg concentration of this event was 9.4 ng L^{-1} , comparable to the overall VWM Hg concentration (9.2 ng L^{-1}) in 2010–2013. This event alone contributed about 16% ($7.6 \mu\text{g m}^{-2}$) of the total wet Hg deposition flux in 2012 ($48.9 \mu\text{g m}^{-2}$). More than 27% of the weekly wet fluxes were greater than $0.82 \mu\text{g m}^{-2} \text{ week}^{-1}$, the mean weekly wet flux, and mainly occurred from May to August. All of these high wet flux samples were associated with rainfall greater than 45 mm . The strong fluctuation pattern in the weekly Hg deposition fluxes at LABS is similar to those MDN sites in the U.S. and some sampling sites in Mexico, where thunderstorms and hurricanes can bring a large amount of rainwater to samples in a short period (Prestbo and Gay, 2009; Hansen and Gay, 2013).

A significant positive correlation between the weekly rainfall and wet Hg deposition flux was observed (Fig. 3(A); $R^2 = 0.67$, $p < 0.01$), indicating that wet Hg flux increased with increasing rainfall at the LABS. About 67% of the variation in wet Hg deposition flux could be explained by the variation in rainfall. This result agreed well with those reported from sites in China, Japan, and Taiwan (Sakata and Marumoto, 2005; Sheu and Lin, 2013; Fu *et al.*, 2016), where rainfall explained about 70% of the wet Hg flux in various sites across Japan (Sakata and Marumoto, 2005) and more than 80% at four mountain sites in China (Fu *et al.*, 2016). On the other hand, the correlation between sample Hg concentration and wet Hg deposition flux was weak, with a R^2 value of 0.032 (Fig. 3(B); $p = 0.033$).

Annual wet Hg deposition fluxes at LABS ranged from 24.4 to $48.9 \mu\text{g m}^{-2}$ (Table 1) with a four-year average of $32.3 \mu\text{g m}^{-2}$ (Table 2). Wet Hg flux at LABS was greater than those observed at the remote and rural sites in China, Italy, Tibetan Plateau, Brazil, and Mexico, mainly due to higher rainfall (Table 2). The wet Hg deposition flux at

Fig. 3. Relationships between weekly wet Hg deposition flux and (A) rainfall (B) rainwater Hg concentration.

Table 1. Summary of rain depths, rainwater Hg concentrations, and wet Hg deposition fluxes at LABS between 2010 and 2013.

Year	Sample size	Rain depth (mm)	Sample Hg conc. Range (ng L^{-1})	VWM Hg conc. (ng L^{-1})	Wet deposition flux ($\mu\text{g m}^{-2} \text{ yr}^{-1}$)
2010	36	3173	4.1–24.9	9.6	30.5
2011	40	3271	2.1–28.2	7.7	25.2
2012	36	3736	2.4–35.1	13.1	48.9
2013	38	3503	0.8–32.4	7.0	24.4

LABS was about 1.5–6.0 times the values measured at 15 sites in the eastern U.S. (Zhang *et al.*, 2016a). Compared to tropical sites, the mean annual wet Hg deposition at LABS was about 1.7 times the values reported from Amapá state and Near Manaus, the low-elevation rural sites in northern Brazil. On the other hand, the wet Hg deposition flux at LABS was close to the value ($27.9 \mu\text{g m}^{-2} \text{ yr}^{-1}$) observed at a tropical site in the Luquillo Mountains, Puerto Rico (Shanley *et al.*, 2015), likely due to comparable amounts of rainfall (Table 2). These comparisons indicated

that rainfall amount was an important factor driving the wet Hg deposition flux in the tropical region. For sites of Taiwan, the wet Hg deposition flux at LABS ($32.3 \mu\text{g m}^{-2}$) was 3 times the value measured at the island of Pengjiayu ($10.2 \mu\text{g m}^{-2}$) because of the higher rainfall at LABS (3421 mm) as compared to at Pengjiayu (1438 mm), even though VWM Hg concentrations at both sites were close. LABS is a high-elevation mountain site, while Penjiayu is a low-elevation remote island site (Sheu and Lin, 2013), this comparison likely indicated the influence of geographical

Table 2. Summary of wet Hg deposition at various sites worldwide.

Location	Site characteristic	Rain depth (mm)	VWM conc. (ng L ⁻¹)	Wet deposition flux (μg m ⁻² yr ⁻¹)	Reference
Mt. Lulin, Taiwan	Mountain	3421	9.2	32.3	This study
Pengjiayu, Taiwan	Island	1438	8.9	10.2	Sheu and Lin, 2013
Mt. Waliguan, China	Mountain	290	6.9	2.0	Fu <i>et al.</i> , 2016
Mt. Leigong, China	Mountain	1533	4.0	6.1	Fu <i>et al.</i> , 2016
Mt. Ailao, China	Mountain	1931	3.7	7.2	Fu <i>et al.</i> , 2016
Mt. Damei, China	Mountain	1621	3.7	6.0	Fu <i>et al.</i> , 2016
Mt. Changbai, China	Mountain	751	7.4	5.6	Fu <i>et al.</i> , 2016
Bayinbuluk, China	High elevation	266	7.7	2.0	Fu <i>et al.</i> , 2016
Southeast Tibet Station, Tibetan Plateau	High elevation	978	4.0	3.9	Huang <i>et al.</i> , 2015
Longobucco, Italy	High elevation	473	6.6	3.1	Sprovieri <i>et al.</i> , 2017
Puerto Rico, U.S.	Rural	2855	9.8	27.9	Shanley <i>et al.</i> , 2015
Cape Point, South Africa	Rural	134–310	1.8–19.6	0.3–5.2	Sprovieri <i>et al.</i> , 2017
Hidalgo, Mexico	Rural	1042	8.2	9.4	Hansen and Gay, 2013
Amapá state, Brazil	Rural	1990	9.1	18.2	Fostier <i>et al.</i> , 2000
Near Manaus, Brazil	Rural	2068	9.8	20.3	Fadini and Jardim, 2001
Guiyang, China	Urban	1057	11.9	12.6	Fu <i>et al.</i> , 2016
Kathmandu, Nepal	Urban	1914	18.3	34.9	Tripathi <i>et al.</i> , 2019
Seoul, Korea	Urban	1235–1645	10.1–16.3	16.8–20.2	Seo <i>et al.</i> , 2012
Chongqing, China	Urban	921	30.7	–	Wang <i>et al.</i> , 2012
Lhasa, Tibetan Plateau	Urban	359	24.8	8.2	Huang <i>et al.</i> , 2013
10 sites around Japan	Various	882–2317	–	5.8–17.7	Sakata and Marumoto, 2005
Europe	Various	–	11.9–18.0	7.0–36.0	Fu <i>et al.</i> , 2015
15 sites in North America	Various	–	–	5.4–21.0	Zhang <i>et al.</i> , 2016a
10 sites in North America	Various	360–1470	–	6.0–12.6	Zhang <i>et al.</i> , 2012
MDN sites across North America	Various	–	9.5	3.0–25.0	Prestbo and Gay, 2009

characteristics on wet Hg deposition through its impact on rainfall distribution. Mountains occupy about half the area of Taiwan and usually with high annual rainfall, implying that Taiwan could receive a large amount of wet Hg deposition every year.

The wet Hg deposition flux at LABS was higher (4–16 times) than those reported from high-elevation and mountain sites in China, such as Mt. Waliguan, Mt. Leigong, Mt. Ailao, Mt. Damei, and Mt. Changbai (Table 2). Both rainfall amount and rainwater Hg concentration contributed to this difference (Table 2). Our result suggests a geographical trend with higher wet Hg deposition to the tropical mountain site (LABS) as compared to sites in the temperate regions, and in inland mountain and high-elevation areas as well. In addition to measurement data, a recent model study was conducted by Horowitz *et al.* (2017) to estimate the global wet Hg deposition fluxes using the updated GEOS-Chem model. Annual wet Hg deposition fluxes of 10–12 μg m⁻² were estimated for the regions around Taiwan (Horowitz *et al.*, 2017). These values were close to the observed value at Pengjiayu (10.2 μg m⁻² yr⁻¹). However, the high value at LABS (32.3 μg m⁻² yr⁻¹) was not captured by this model. Results of the current research tend to support the hypothesis that tropical region, especially the windward maritime slopes, may be hot spots of wet Hg deposition (Shanley *et al.*, 2015; Horowitz *et al.*, 2017).

Temporal Patterns of Wet Hg Deposition

The interannual variations in VWM Hg concentration and wet deposition flux at LABS are summarized in Table 1. The VWM Hg concentrations showed a significant interannual variability with the highest value in 2012 (13.1 ng L⁻¹) and the lowest value obtained in 2013 (7.0 ng L⁻¹). Interannual variability was also observed for wet Hg deposition fluxes, with the highest and lowest wet fluxes observed in 2012 (48.9 μg m⁻² yr⁻¹) and 2013 (24.4 μg m⁻² yr⁻¹), respectively. The highest annual rainfall amount (3736 mm) and VWM Hg concentration (13.1 ng L⁻¹) in 2012, resulting in the highest wet Hg deposition flux in 2012 as compared to other years (Table 1).

Seasonal variations in rainfall amounts, VWM Hg concentrations, and wet Hg deposition fluxes were observed at LABS (Fig. 4). Seasonal cumulated rainfalls (four-year mean) were 831, 1703, 601, and 286 mm, whereas seasonal VWM Hg concentrations were 9.4, 9.7, 9.7, and 7.4 ng L⁻¹ in spring (March–May), summer (June–August), fall (September–November), and winter (December–February), respectively (Figs. 4(A)–4(C)). The combination of higher rainfall and rainwater Hg concentration resulted in the highest wet deposition flux in summer (16.6 μg m⁻²), as compared to winter (1.9 μg m⁻²), fall (4.9 μg m⁻²), and spring (7.9 μg m⁻²). On average, summer contributed about 53% of the annual wet Hg deposition flux. The seasonal wet

Fig. 4. Seasonal distribution of (A) rainfall, (B) VWM Hg concentration, and (C) wet Hg deposition flux.

flux pattern at LABS was similar to those observed at 10 sites in Japan (Sakata and Marumoto, 2005), the NADP/MDN sites in the U.S. and Canada (Prestbo and Gay, 2009), Pengjiayu, Taiwan (Sheu and Lin, 2013), Huntington Wildlife Forest, US (Mao *et al.*, 2017b), and two tropical sites at Sisal, Mexico and Puerto Rico (Shanley *et al.*, 2015; Sprovieri *et al.*, 2017). At most of these sites, greater wet Hg deposition flux in summer was a result of both greater rainwater Hg concentration and higher rainfall, as was observed at LABS.

Between 2010 and 2013, the lowest VWM Hg concentration was found in winter ($p < 0.05$), whereas the values were nearly equal for spring, summer, and fall (Fig. 4(B)), as shown by the analysis of covariance (ANCOVA) using rainfall as a covariate. This seasonal pattern was different from the pattern, higher Hg concentrations in winter and lower in summer, reported by Fu *et al.* (2016) for some sites in China. Discrepancies between LABS and those Chinese sites could be due to the

impact of local anthropogenic Hg emissions. At these Chinese sites, seasonal patterns of VWM Hg concentration were partially driven by the elevated particulate-bound mercury (PBM) concentrations from local anthropogenic Hg emissions in winter, and the dilution effect of higher rainfall in summer (Fu *et al.*, 2016). However, there are no local anthropogenic Hg emission sources near the LABS (Sheu *et al.*, 2010). On the other hand, the pattern of lowest VWM Hg concentration in winter and higher in other seasons has been observed at Puerto Rico and Pengjiayu, where sites were not affected by direct anthropogenic Hg emissions. This has been attributed to the effect of rainfall associated with deep convection such as thunderstorm (Sheu and Lin, 2013; Shanley *et al.*, 2015). Higher summer rainwater Hg concentrations have been observed at many of the NADP/MDN sites that were also attributed to the effect of deep convection (Prestbo and Gay, 2009). Recent observation and model studies demonstrated that deep

convective rainfall can scavenge Hg(II) from the upper free troposphere, resulting in elevated rainwater Hg (Nair *et al.*, 2013; Holmes *et al.*, 2016). Results of these studies thus suggested the importance of rainfall types to the seasonal pattern of rainwater Hg concentration.

The oxidized forms of Hg, including gaseous oxidized mercury (GOM) and PBM, comprise the majority of rainwater Hg concentration due to its higher solubility (Manson and Sheu, 2002; Driscoll *et al.*, 2013). Anthropogenic Hg(II) emissions and photochemical oxidation of GEM, are the primary sources of Hg(II) in the atmosphere (Weiss-Penzias *et al.*, 2011; Weiss-Penzias *et al.*, 2016; Shah and Jaeglé, 2017). Transport of air pollutants, including Hg, from East and Southeast Asia to Taiwan has been well documented by numerous studies (e.g., Sheu *et al.*, 2010; Sheu and Lin, 2011; Sheu *et al.*, 2013; Ou-Yang *et al.*, 2014; Pani *et al.*, 2016; Pani *et al.*, 2017; Nguyen *et al.*, 2019; Sheu *et al.*, 2019). Export of atmospheric Hg from East and Southeast Asia to LABS occurs mainly in fall, winter, and spring due to the prevailing westerlies wind and northeast monsoon (Sheu *et al.*, 2010; Nguyen *et al.*, 2019). Seasonal distribution of BWTs in 2010–2013 (Figs. 5(A)–5(D)) indicated that air masses reached LABS mainly came from South China Sea and the West Pacific Ocean in summer, whereas air masses were mainly from the East Asia continent and Indochina Peninsula in fall, winter, and spring (Figs. 5(A)–5(D)). Although the Hg emissions from East and Southeast Asia primarily affected LABS in spring, fall, and winter, however, the rainwater VWM Hg concentration was the lowest in winter (Fig. 4(B)).

GOM and PBM are the two major contributors to the Hg in rainwater. However, these Hg forms have short lifetimes and hence tend to deposit near the emission sources instead of being transported long-distance (Driscoll *et al.*, 2013). A recent model study indicated a negative transport budget of GOM/PBM in mainland China, suggesting the direct GOM/PBM emissions deposit domestically rather than being exported to downwind regions (Wang *et al.*, 2018). Therefore, the contribution of direct anthropogenic GOM/PBM emitted from East and Southeast Asia to the wet Hg deposition at LABS might be limited. However, the anthropogenic GEM emitted from East and Southeast Asia can be transported to upper troposphere where it could be oxidized to produce Hg(II) then scavenged by cloud water and rainwater and hence contribute to the wet Hg deposition (Strode *et al.*, 2008; Sheu and Lin, 2013).

In summary, the seasonal pattern of VWM Hg concentration at LABS cannot be fully explained by the seasonal export pattern of air pollutants from East and Southeast Asia. This suggests that processes other than the export of Asian atmospheric Hg are also influencing rainwater Hg concentrations. As mentioned previously, rainfall type could be an important factor in determining the Hg concentration in rainwater (Holmes *et al.*, 2016). LABS is located in a region with pronounced seasonal monsoon activities, hence rainfall types vary seasonally (Lin *et al.*, 1999; Sheu *et al.*, 2010; Lin *et al.*, 2013). Therefore, the seasonal differences in rainwater Hg concentrations could be related to the seasonal differences in rainfall types, which are explored in the following section.

Fig. 5. Five-day backward trajectories of (A) spring, (B) summer, (C) fall, and (D) winter in 2010–2013.

Influence of Rainfall Types on Rainwater Hg Concentration

Effects of rainfall type on wet Hg deposition, including rainwater Hg concentration and wet flux, over various sampling sites in the U.S. have been discussed by recent publications (Holmes *et al.*, 2016; Kaulfus *et al.*, 2017). The current study focuses on exploring the influence of rainfall type on the concentration and seasonal pattern of rainwater Hg. To elucidate the impact of rainfall type on rainwater Hg concentration at LABS, seven rainfall types were categorized based on the approach proposed by Lin *et al.* (1999), including northeast flow (NE), frontal system in fall and winter (FA), circulation associated with high pressure in spring (HS), frontal system in spring (FS), low pressure in southern China or the South China Sea (LS), Pacific high pressure (PH), and outskirts of typhoon circulation (TP).

The VWM Hg concentration associated with each rainfall type was shown in Fig. 6. VWM Hg concentrations decreased in the following order: PH (13.5 ng L^{-1}) > HS (11.2 ng L^{-1}) > FA (10.7 ng L^{-1}) > LS (8.7 ng L^{-1}) $\approx$ FS (8.7 ng L^{-1}) > TP (8.6 ng L^{-1}) > NE (7.3 ng L^{-1}). Rain events associated with the PH weather system were about 2.3–6.2 ng L^{-1} greater in VWM Hg concentration as compared to other rainfall types. PH weather system often occurs in summer months in Taiwan. Because of intense surface heating under summer PH conditions, precipitation usually occurs locally due to strong convection, resulting in afternoon shower or thunderstorm (Lin *et al.*, 1999; Cheng *et al.*, 2014). Holmes *et al.* (2016) analyzed 800 individual rainfall events to investigate the contribution of thunderstorms (deep convection) to rainwater Hg concentration over various sites in the eastern U.S. during 2006–2011. They found that thunderstorms increased Hg concentration by 50% relative to other weak convective rainfall types. This value is close to our finding at LABS (47%), suggesting the important role of strong convection in driving rainwater Hg concentration at LABS. This enhancement in rainwater Hg has been attributed to the scavenging of upper-altitude GOM

Fig. 6. VWM Hg concentrations associated with various rainfall types in 2010–2013.

by deep convection (Selin and Jacob, 2008). Accordingly, the occurrence frequency of PH and other rainfall types could influence the seasonal pattern of rainwater Hg concentrations.

Seasonal relative frequency distribution in rainfall types at LABS between 2010 and 2013 is shown in Fig. 7 and a clear difference was observed. In winter, 77% of the rain events at LABS were categorized as NE rainfall type, which is the rainfall type with the lowest Hg concentration, leading to the lowest seasonal VWM Hg concentration. On the other hand, about 1/3 of the rain events in summer were under the categories of PH (Fig. 7), the rainfall type with highest Hg concentration (Fig. 6), resulting in the highest seasonal VWM Hg concentration. Therefore, it is evident that the seasonal distribution of rainfall types could influence the seasonal pattern of rainwater Hg concentrations. It is worth noting that mean rain depth per event was higher in summer ($175.6 \text{ mm event}^{-1}$) than in winter ($38.9 \text{ mm event}^{-1}$). The dilution effect should be more effective in summer

Fig. 7. Seasonal relative distribution frequencies of rainfall types in 2010–2013.

that would have led to lower rainwater Hg concentrations. However, because of the continuous supply of upper-altitude GOM to cloud water during PH-type rain events to counteract the dilution effect, rainwater Hg concentrations remained high in summer.

Elevated GOM concentrations have been observed frequently at mountain and high-elevation sites, such as LABS, Mount Bachelor Observatory and Storm Peak Laboratory, (Swartzendruber *et al.*, 2006; Fain *et al.*, 2009; Sheu *et al.*, 2010). Aircraft measurements and model simulation also suggested the formation of GOM in the upper atmosphere (Sillman *et al.*, 2007; Talbot *et al.*, 2007). Results of these studies thus indicated the existence of a GOM pool in the free troposphere, supporting the argument about the continuous supply of upper-altitude GOM to cloud water in PH-type rain events in summer. Although direct anthropogenic GOM emissions from the East Asian continent may not contribute to the measured rainwater Hg concentrations, anthropogenic GEM emissions could be transported to the upper troposphere where they can be oxidized to form GOM (Strode *et al.*, 2008), which will then be scavenged by cloud water and rainwater.

CONCLUSIONS

The purpose of this study was to characterize the wet Hg deposition to LABS, a tropical mountain site with high rainfall in central Taiwan. In 2010–2013, rainwater Hg concentrations ranged between 0.8 and 35.1 ng L⁻¹, with an overall VWM Hg concentration of 9.2 ng L⁻¹. A significant but weak correlation ($R^2 = 0.036$, $p = 0.022$) was found between weekly rainwater Hg concentration and rainfall, suggesting the scavenging of atmospheric oxidized Hg by rainwater in the early stages of rain events and the dilution effect caused by substantial rainfall. The VWM Hg concentration at LABS was higher than those reported from Chinese mountain sites, suggesting factors other than scavenging and dilution, such as meteorological conditions, rainfall type, atmospheric chemistry, and Hg emissions and transport, could also contribute to this difference.

Weekly wet Hg deposition fluxes ranged between 0.01 and 7.6 $\mu\text{g m}^{-2}$ with a mean value of 0.82 $\mu\text{g m}^{-2}$. A positive correlation ($R^2 = 0.67$, $p < 0.01$) was observed between weekly rainfall and wet deposition flux, whereas a weak correlation ($R^2 = 0.032$, $p = 0.033$) was observed between sample Hg concentration and wet deposition flux. This indicates that rainfall amount is a more important factor than rainwater Hg concentration in determining the wet Hg deposition flux. Annual wet Hg deposition ranged between 24.4 and 48.9 $\mu\text{g m}^{-2}$ with a mean value of 32.3 $\mu\text{g m}^{-2}$. This mean value was about 1.5–6.0 times the values reported from 15 sites in the U.S. and 4–16 times the values reported from mountain and high-elevation sites in China. Both rainwater Hg concentration and rainfall amount contributed to this difference, but rainfall amount played a more important role.

Seasonally, both the rain depth and rainwater VWM Hg concentration were the highest in summer, resulting in the highest wet Hg deposition flux in summer. On average,

summer contributed about 53% of the annual wet Hg deposition flux. The seasonal pattern of rainwater Hg concentration was different from the pattern of the East Asian air pollutant export, indicating other factors were also influencing the seasonal distribution of rainwater Hg concentrations, such as rainfall type. Seven rainfall types have been categorized and the PH type was found to have the highest VWM Hg concentration as compared to the other rainfall types. In summer, about 1/3 of the rain events were under the categories of PH and hence resulting in the highest seasonal VWM Hg concentration. Scavenging of free tropospheric GOM by deep convection during PH-type rain events was likely the cause for the elevated rainwater Hg levels.

Results of this study indicated that tropical areas in East and Southeast Asia, especially the windward maritime slopes, could be hot spots of wet Hg deposition. However, measurement data are very limited in this region. Recently, the Asia Pacific Mercury Monitoring Network (APMMN), a network of sites operating in Asia and the Western Pacific region to measure Hg in precipitation cooperatively, has been established to fill this data gap. Some sampling sites of the APMMN are in the tropical areas of Southeast and South Asia and thus can provide the much needed wet Hg deposition measurement data. Besides, dry deposition of atmospheric Hg could also play an important role in total Hg deposition budget, particularly in East and Southeast Asia where with high Hg emissions. Therefore, dry Hg deposition studies are also needed to complement the wet deposition for a better estimation of total deposition.

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