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Influence of meteorological factors on the atmospheric mercury measurement by a novel passive sampler

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HIGHLIGHTS

- Impact of meteorological factors on a passive mercury sampler was identified.
- Correction factor for the sampling rate calculation was obtained.
- The passive sampler was applied in Beijing and Tibet to test its performance.

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ABSTRACT

In recent years, an incentive for developing simple and cost-effective samplers that are capable of monitoring over an extended period and require nonattendance at remote locations was obvious. Compared to traditional active sampling approaches, passive samplers require no electric power and are more flexible in field deployment, thus they are more appropriate for screening applications and long-term sampling. However, the performance of passive samplers may be influenced by meteorological factors, therefore inducing bias for the result of passive sampling. In this study, the effects of temperature, relative humidity, and wind speed on the performance of a novel passive sampler for gaseous mercury were investigated. The meteorological factors were well controlled in an exposure chamber. The passive samplers were tested in different conditions: temperature ranging from −10 to 35 °C, relative humidity ranging from 25 to 90%, wind speed ranging from 0.5 to 5.0 m s^{-1}. The results showed that temperature and relative humidity had no significant influence on the performance of the passive sampler. However, wind speed was found to have significant impact on the sampling rate of the passive sampler. Wind correction should be considered when making comparisons among samplings with different average wind speeds. In the field application in Beijing and Tibet site, the passively measured data were well correlated with the active measurements.

1. Introduction

Mercury is a global atmospheric pollutant and attracts increasing attentions due to its inverse impacts on human health. Mercury in atmosphere is typically measured as three fractions: gaseous elemental mercury (GEM), particulate-bound mercury (PBM) and gaseous oxidized mercury (GOM) (Lyman et al., 2010). GEM is the prevalent form in the atmosphere, consisting of >90% of the total mercury (Ebinghaus et al., 2002; Huang et al., 2014). Due to its volatility and chemical stability, GEM can circulate in the atmosphere for 1–2 years, allowing its wide dispersion and long-distance transportation (Fang et al., 2009). In order to understand the sources, trends, and potential influence of mercury to the environment, it is important to evaluate the temporal and spatial patterns of atmospheric mercury (Lyman et al., 2010). Traditional automated instruments for measurement of gaseous mercury rely on electric power, a large financial investment and continuous

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operating costs, and require attendance of trained technical staff (May et al., 2011; Pirrone et al., 2013; Huang et al., 2012). Therefore, the active sampling technology has difficulties in assessing atmospheric mercury in remote sites. Additionally, concerns regarding the accurate and precise measurements of atmospheric mercury using active sampling instruments have also been raised. For example, for the Tekran 1130/1135/2537 system, the GOM measurements using KCl-coated denuders have been recently reported to underestimate GOM concentrations by 2–4 times (Huang et al., 2014). The measurement of total gaseous mercury by the Tekran system depends on how the instrument is deployed (Huang et al., 2014; Gustin et al., 2013).

In response to the limitations of active sampling, there has been a growing interest in the use of passive samplers (May et al., 2011). Passive samplers, with the advantage of low-cost, simple to use, and requiring no power, are developed as a cost-effective alternative to active samplers. In field applications, passive samplers would usually need a sampling time of more than one week. This allows them to record a time-average trend of atmospheric mercury. In addition, because of the flexibility in field applications, passive samplers can cover a large spatial area with deployment of large quantities across a broad geographic region simultaneously (May et al., 2011). Passive techniques are working well for the monitoring of a number of persistent organic pollutants (Pirrone et al., 2013). Nevertheless, one major disadvantage of passive samplers reported by many studies is that the sampling rate of passive samplers would be affected by meteorological factors and thus a systematic bias is induced. According to previous studies, temperature, humidity, and wind speed were discovered to have impacts on sampling rate, especially wind speed (Gustin et al., 2011; Plaisance et al., 2004; Tuduri et al., 2006; Pozo et al., 2004; Seethapathy et al., 2008). Literature reported that wind speed may influence the effective diffusion path length (Fan et al., 2006). For tube-type passive samplers, it was reported that a length:-diameter ratio of 2.5–3 is sufficient to overcome the effects of wind turbulence (Harper and Purnell, 1987). However, Plaisance et al. (2004) showed that the magnitude of wind effect is very high and the rule of length:diameter ratio of 2.5–3 is incorrect.

In order to identify the influence of meteorological factors on the performance of passive samplers, laboratory experiments with exposure chamber are used (Plaisance et al., 2004). Controlling the tested meteorological factor at the designed range, this approach can estimate the magnitude of the influencing factors and explore modifications of passive sampling technologies. In our previous study, a passive sampler for measurement of gaseous mercury in the atmosphere was developed (Zhang et al., 2012). In this study, the influence of temperature, humidity and wind speed on the performance of the passive sampler was investigated in an exposure chamber. The relationship between the meteorological factors and the sampling rate was studied, and the correction factor for the sampling rate calculation was obtained to improve the accuracy of the passive measurement. Application of the passive sampler was conducted in Beijing and Tibet to test its suitability and stability under various environmental conditions.

2. Materials and methods

2.1. The passive sampler

The passive sampler for gaseous elemental mercury consists of a diffusion tube, a rain shield and an adsorption carrier with a mesh screen (pore size of 75 μm). The mesh screen was used to further reduce the face air velocity effects and minimize the entrapment of large aerosol particles, however, diffusion of fine particles such as PM2.5 cannot be prevented. The cylindrical shelter is designed to reduce the turbulence of external air, and it is sealed with the rain shield on the top of the diffusion tube. It has 24 evenly distributed 5-mm diameter openings at its bottom for air exposure. The mesh screen of the adsorption carrier was designed to reduce the influence of deposition of the particulate matters in the air. The material carrier was fixed at the ceiling of the tube with pulverized sulfur-impregnated carbon (Calgon Carbon Corporation) in it with the size of 250–380 μm as adsorption material. The more specific configuration of the passive sampler is described in our previous paper (Zhang et al., 2012).

2.2. Exposure chamber experiment

In the laboratory experiments, an exposure chamber was used to investigate the impacts of meteorological factors (humidity, temperature, and wind speed) on the sampling rate of the passive sampler. The schematic of the exposure chamber is presented in Fig. 1. In the exposure chamber, the level of the meteorological parameters can be preciously controlled: temperature (−20 °C–50 °C, 0.1 °C), relative humidity (10%–90%, 1%), wind speed (0 m s^{-1}–5 m s^{-1}, 0.1 m s^{-1}). A series of laboratory experiments with the combination of the three meteorological parameters at different levels were carefully designed, and summarized in Table 1. The range of the parameters was set according to the common values of the natural environment. For example, the environmental relative humidity usually ranges from 30% to 90%. Thus, the impact of relative humidity the performance of the passive sampler was investigated in four batches ranging from 25% to 90%, and in each batch, the temperature was controlled at 25 °C and the wind speed was set at 0.5 m s^{-1}. Similarly, according to the common range of temperature in the environment, the impact of temperature was studied in five levels ranging from −10 °C to 35 °C in five experiment batches respectively. In each batch, the level of relative humidity and wind speed was controlled at 25% and 2.0 m s^{-1}, respectively. In previous studies, wind speed was suggested as a very possible meteorological factor affecting the sampling rate of diffusive passive samplers (Gustin et al., 2011; Zhang et al., 2012; Seethapathy et al., 2008; Fan et al., 2006). Therefore, in this study, the impact of wind speed was investigated ranging from 0.1 to 5.0 m s^{-1}. In each experiment for wind speed, the level of relative humidity and temperature was set at 35% and 25 °C respectively.

In each batch of experiment, five passive samplers were deployed in the chamber as replicates (the distance between samplers was approximately 8 cm). The mass of the adsorption material (sulfur-impregnated carbon) in each sampler was 0.946 ± 0.049 g (Mean ± SD). The deployment lasted 8 days for each batch. The mercury concentration within the exposure chamber

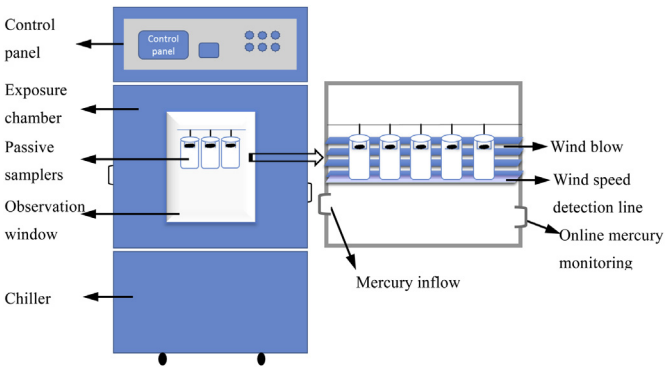

Fig. 1. Schematic of the exposure chamber.

Table 1
Design of the exposure chamber experiments with the different levels of the meteorological parameters.

Meteorological factor	No.	Humidity (%)	Temperature (°C)	Wind velocity (m s^{-1})
Humidity	1	25.0	25.0	0.5
	2	35.0	25.0	0.5
	3	55.0	25.0	0.5
	4	90.0	25.0	0.5
Temperature	1	25.0	−10.0	2.0
	2	25.0	0.0	2.0
	3	25.0	15.0	2.0
	4	25.0	25.0	2.0
	5	25.0	35.0	2.0
Wind velocity	1	35.0	25.0	0.5
	2	35.0	25.0	1.0
	3	35.0	25.0	2.0
	4	35.0	25.0	3.0
	5	35.0	25.0	4.0
	6	35.0	25.0	5.0

was maintained at a relatively stable level (5–10 ng m^{-3}) using a mercury permeation tube. A mercury analyzer (Lumex RA-915, Lumex Ltd., St. Petersburg, Russia) was used for online monitoring of the mercury concentration in the exposure chamber simultaneously with the passive sampling. It provides accurate measurements of gaseous mercury concentrations and exhibits a good compatibility with other traditional methods at concentrations higher than 2 ng m^{-3} (Southworth et al., 2004; Kim et al., 2006; Zhang et al., 2012; García-Sánchez et al., 2006). Fu et al. (2011) compared the performance of Lumex RA-915 and Tekran 2537A in measuring the atmospheric mercury concentrations in the city of Guiyang, China. They concluded that both of the instruments could accurately measure gaseous mercury concentration in ambient air.

2.3. Field application in Beijing and Tibet

In order to investigate the performance of the passive sampler in different field conditions, the passive samplers were deployed at an urban site in Beijing and a remote site in Namtso as field application. Namtso is a mountain lake on the border between Damxung County of Lhasa Prefecture and Baingoin County of Nagqu Prefecture in the Tibet Autonomous Region of China, approximately 112 km NNW of Lhasa. The passive samplers were deployed at the Namtso Monitoring Station of the Institute of Tibetan Plateau Research. The Beijing site represents an urban site with relatively high gaseous mercury concentration and low wind speed. The Namtso site represents a remote site with gaseous mercury at the atmospheric background level and high wind speed. At each site, the passive samplers were deployed for three periods, covering 30 days, 60 days and 90 days, respectively. At the Beijing site, additional field deployments for 10 and 20 days were also conducted. With each deployment, four passive samplers ($n = 4$) were deployed as replicate samples. Meteorological data, including wind speed, temperature, relative humidity and precipitation was recorded using HOBO weather stations (Onset Co., USA) installed at both sites. To evaluate the performance of the passive samplers, active measurements were performed simultaneously with the passive sampling, using Lumex RA-915 at the Beijing site and Tekran 2537A at the Namtso site. Prior to the field application, the performance of Lumex RA-915 and Tekran 2537 was compared for measuring mercury concentrations in urban air in Beijing. The two instruments were operated synchronously for two weeks, and the relative error of the observed mercury concentrations of the two instruments was less than 5%. The meteorological data and actively measured mercury concentration in each deployment was summarized in Table 2.

2.4. Laboratory analysis

All the samples were transported to the laboratory for analysis immediately after deployment. The analysis of mercury was conducted using a direct mercury analyzer DMA-80 (Milestone Inc., Italy). For each sample, approximately 0.3000 g of the adsorption material was analyzed in accordance with USEPA method 7473. Detailed information on the laboratory analysis procedure was described in our previous paper (Zhang et al., 2012). Briefly, the sample was analyzed using the integrated sequence of thermal decomposition (180 s at 650 °C), amalgamation and atomic absorption spectrophotometry. Absorbance was measured at 253.7 nm as a function of the mercury concentration.

2.5. Quality assurance/quality control

After deployment, the samplers were disassembled and washed with distilled water. Prior to deployment, the diffusive tubes and adsorption carriers were immersed in a 15% HNO_3 bath for at least 48 h, and then rinsed three times with distilled water and another three times with Milli-Q ultrapure water (Millipore, MA, USA). Method blanks for laboratory experiment and travel blanks for field application were prepared, transported, deployed and analyzed following the same procedure as the samples, and the blanks were left in sealed bottles for the same time interval of each sampling period. For the exposure chamber experiment, the blank value was 0.58 ± 0.09 ng g^{-1} (Mean ± SD). For the field application, the blank value was 0.71 ± 0.11 ng g^{-1} (Mean ± SD). The data of the laboratory experiment and field application were corrected using the corresponding blank value.

The method detection limit (MDL) of the passive sampling materials was calculated as three times the standard deviation of the method blanks. In this study, the MDL for the 8-day deployment in the exposure chamber was 0.18 ng m^{-3}, and for the 30-day deployment of the filed application was 0.05 ng m^{-3}. Replicate samplers ($n = 5$ in the laboratory experiment and $n = 3$ in the field application) were deployed. The relative standard deviation (RSD) of the replicate samplers was averaged at 11% in the laboratory experiment and 9% in the field application.

During the analysis of the adsorption material, the DMA80 analyzer was calibrated to optimize the response over the working range (0–20 ng mercury). Standard working solutions for mercury were freshly prepared by diluting the stock solutions with Milli-Q ultrapure water (Millipore, MA, USA). All of the samples were measured in duplicate to check for reproducibility, and the RSD of the duplicate measurements was less than 8%.

Table 2
Meteorological parameters and actively measured data for the field application.

Site	Event no.	Sampling period (d)	Active data (ng m^{-3})	Wind speed (m s^{-1})	Temperature (°C)	Humidity (%)
Beijing	1	30	2.85	1.16	18.04	56.73
	2	60	4.21	1.43	−3.33	56.09
	3	90	4.08	1.34	−0.46	58.79
	4	10	3.50	1.02	15.34	54.90
	5	20	5.00	1.53	19.70	48.57
Namtso	1	30	1.61	2.37	8.70	63.90
	2	60	1.52	2.42	7.76	58.87
	3	90	1.42	2.52	5.41	52.65

2.6. Data analysis

The data were processed using Microsoft Excel 2010, and the statistical analysis was performed using SPSS 11.0. The significance level was 0.05 unless specially noted. The passive measurements (ng g^{-1} day^{-1}) of the passive samplers were normalized against the corresponding actively measured concentrations in ambient air (ng m^{-3}) and the results were presented as sampling rate in m^3 g^{-1} day^{-1}. The sampling rate of the passive sampler was calculated as follows:

$$R_p = \frac{M_p}{m \times C_a \times T} \quad (1)$$

where M_p is the amount of mercury adsorbed in the sampler (ng); m is the mass of the adsorption material (g); C_a is the concentration of gaseous mercury (ng m^{-3}); T is the duration of deployment (day).

3. Results and discussion

3.1. Effect of temperature and humidity

The mass transfer of gaseous mercury for passive samplers is mostly controlled by molecular diffusion. Temperature and relative humidity could have potential impacts on the performance of the passive samplers (Strandberg et al., 2006). In the exposure chamber experiments, five batches were conducted for different temperature and four batches for different relative humidity. The passive sampling rate under the controlled conditions was illustrated in Fig. 2. One-way ANOVA analysis showed that the difference in the sampling rate at different temperature was not significant ($p = 0.116$, $n = 25$). For relative humidity, no significant difference was found in the five batches ($p = 0.071$, $n = 20$). Therefore, the effect of temperature and humidity on the sampling rate is minor. It should be noted that the tested temperature ranged from −10 °C to 35 °C and the relative humidity ranged from 25% to 90%. Although the effects of temperature and relative humidity beyond these ranges should be further examined, the ranges of temperature and humidity in this study were wide enough to represent normal natural conditions in most cases. In literature, Gustin et al. (2011) reported that no correlation of the sampling rate with temperature or relative humidity was found in the field deployment.

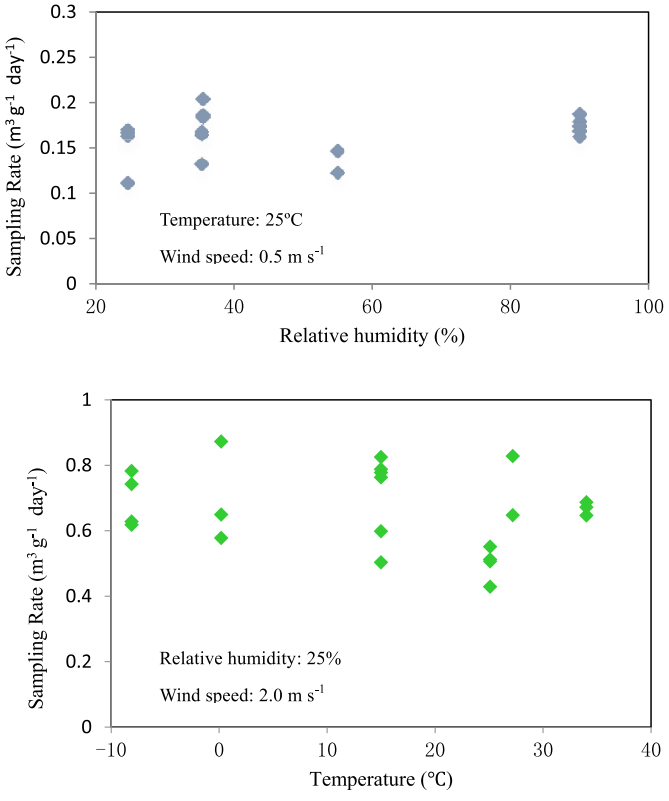

Fig. 2. Sampling rate at different relative humidity and temperature.

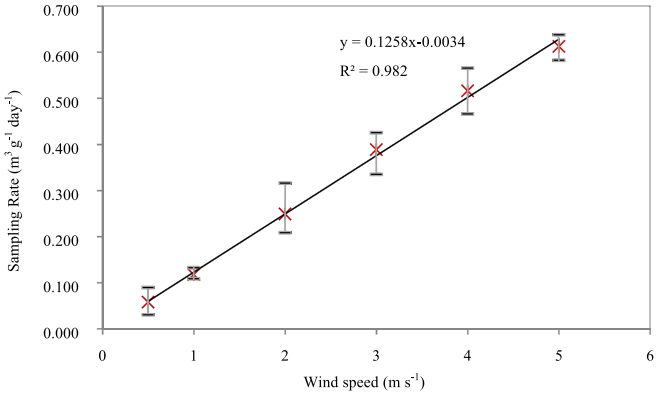

Fig. 3. Linear relationship between the sampling rate and wind speed (The "×" represents the mean value, the top and bottom line represents the maximum and minimum value, respectively).

3.2. Effect of wind speed

For passive air samplers, because the effective diffusion path length is affected by the air turbulence, wind speed is an important factor affecting the performance of the passive sampling. This is particularly evident for tube-type samplers (Seethapathy et al., 2008; Zhang et al., 2012; Tuduri et al., 2006). In previous studies, efforts were conducted to minimize the effect of wind. For example, Harper and Purnell (1987) reported that for a tube-type sampler, the effect of wind turbulence can be overcome if the ratio of the tube length versus diameter is between 2.5 and 3. However, Plaisance et al. (2004) showed that the rule of length:diameter ratio of 2.5–3 is incorrect and the magnitude of wind influence is very high. However, they pointed out that the turbulence on the static air layer within the tube tends to decrease with increasing wind speed (Plaisance et al., 2004). In this study, the length:diameter ratio of the passive sampler was 3.3. In order to investigate the effect of wind speed on the performance of the passive sampler, laboratory experiments were conducted. In the laboratory chamber experiment, the sampler was exposed to six groups of wind speed ranging from 0.5 to 5.0 m s^{-1}, which covers the normal conditions in the natural environment. For each group, five passive samplers were deployed as replicate samples. Correlation analysis showed a significant positive relationship between the sampling rate and the wind speed ($p < 0.001$).

In previous studies, the positive correlation between sampling rate and wind speed was also reported (Harner et al., 2003; Plaisance et al., 2004; Tuduri et al., 2006; Sderstrm and Bergqvist, 2004), including those applied to gaseous oxidized mercury (Lyman et al., 2010). The regression curve for the sampling rate and wind speed was widely discussed. For example, some researchers reported that the relationship between the sampling rate and wind speed followed a logarithmic equation (Perez Ballesta et al., 1993). While in other studies, a quadratic function or linear function provides a better fit (Plaisance et al., 2004; Lyman et al., 2010). In

this study, an ANOVA test of linearity was conducted and the result showed that the relationship between sampling rate and wind speed was linear ($p < 0.001$) and deviation from linearity was not significant ($p = 0.409$), indicating that there is a linear relationship between sampling rate and wind speed, and significant variation can be explained by a linear model. Therefore, linear regression was conducted for the sampling rate (in $m^3\ day^{-1}$) and wind speed (in $m\ s^{-1}$), as shown in Fig. 3. The linear function for sampling rate and wind speed was derived:

$$\text{sampling rate} = 0.1258 \times \text{wind speed} - 0.0034 \quad (2)$$

The regression coefficient of sampling rate versus wind speed was significantly different from zero ($p < 0.001$), and the y-intercept was not significantly different from zero ($p = 0.734$). Fig. 3 showed that the wind effect was not completely removed, and the performance of the passive sampler was correlated with wind speed.

3.3. Calibration of the passive sampler and field application

The passive samplers were deployed in Beijing and Namtso to test its field application under different environmental conditions. The sampling rate for the passive samplers deployed at each site was calculated according to Equation (1). The average value was $0.225 \pm 0.022\ m^3\ day^{-1}\ g^{-1}$. The uptake rate of the passive sampler was significantly correlated with the actively measured concentration ($p < 0.001$), and the relationship was displayed in Fig. 4.

It should be noted that the wind dependence of the sampling rate may produce a bias in the passive measurement. Therefore, the results of the passive sampler should be corrected for wind speed. Based on Equation (2), the concentrations of gaseous mercury can be calculated as:

$$C = \frac{U}{0.1258 \times W - 0.0034} \quad (3)$$

where C is the calculated concentration of gaseous mercury ($ng\ m^{-3}$); U is the uptake rate of mercury to the passive samplers ($ng\ g^{-1}\ day^{-1}$); W is the average wind speed during deployments ($m\ s^{-1}$). The equation is only valid when the range of average wind speed is from 0.5 to 5.0 $m\ s^{-1}$.

The concentrations of gaseous mercury were calculated from the passive measurement and compared with the active data measured during the same period (Fig. 5). The mercury concentrations derived from the passive measurement (with wind correction) were well associated with the actively measured mercury concentrations ($R^2 = 0.509$). The slope of the linear relationship was 0.991 (the 95% confidence interval is 0.895 and 1.086), and not significantly different from one ($p = 0.421$). In Fig. 5, the mercury concentrations were also calculated without wind correction. The slope of this relationship was 0.763 (the 95% confidence interval is 0.685 and 0.841), and was different from one ($p < 0.001$). This result showed that the effect of wind speed on sampling rate cannot be negligible in the field deployments. The average wind speed during the field deployments in Beijing and Namtso varied from 1.02 to 2.52 $m\ s^{-1}$, therefore the wind effect is significant. It should be noted that the wind effect would be more significant when comparing the passive sampling data between sites with larger variation in average wind speed. Under such condition, wind correction is important.

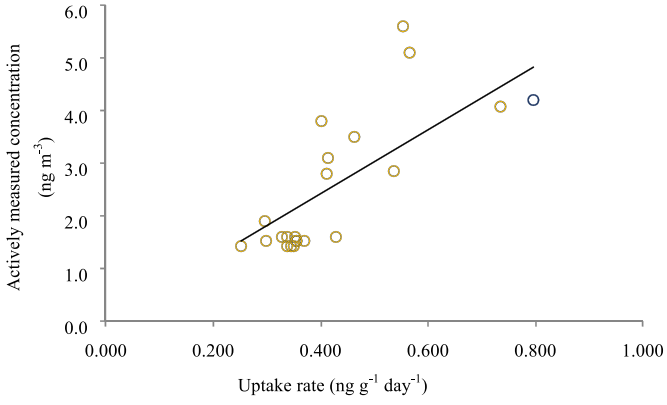

Fig. 4. Relationship between the actively measured mercury concentration and uptake rate of the passive sampler.

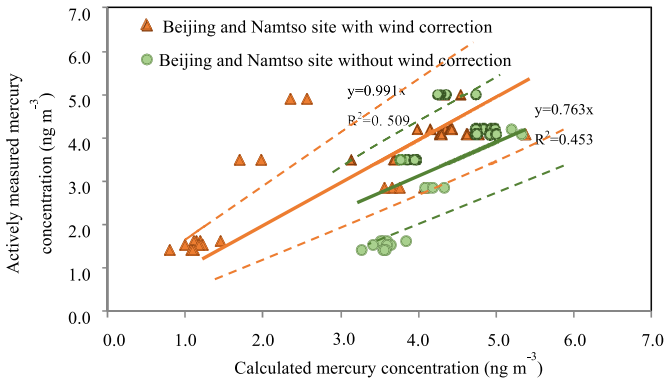

Fig. 5. Comparison of the mercury concentration calculated from the passive data and actively measured concentration (with and without wind speed correction, including the Beijing and Namsto site deployments). The dash line represents the 95% confidence interval.

4. Conclusion

In this study, the sampling rate of a diffusive passive sampler for atmospheric gaseous mercury was calculated and evaluated. The results of exposure chamber experiments demonstrated that common temperature or relative humidity did not have significant influence on the sampling rate. However, the effect of wind speed on sampling rate was significant. Based on the result of the chamber experiments, a linear model between the sampling rate and wind speed was developed. In the field deployments at the Beijing site and the Namtso site, the concentrations of gaseous mercury were calculated using the equation for wind correction derived from the exposure chamber experiments. The calculated concentrations were not significantly different from the actively measured values. Therefore, wind correction is important when comparing the passive sampling data between sites with large variation in wind speed.

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