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# Desert dust as a significant carrier of atmospheric mercury☆

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ABSTRACT

The atmospheric circulation plays a critical role in the global transport and deposition of atmospheric pollutants such as mercury (Hg). Desert dust emissions contribute to nearly 60–95% of the global dust budget and thus, desert dust may facilitate atmospheric Hg transport and deposition to the downwind regions worldwide. The role of desert dust in biogeochemical cycling of Hg, however, has not been well recognized by the Hg research community. In this study, we measured the concentration of particulate bound Hg ($Hg_P$) in total suspended particulate (TSP) collected from China's largest desert, Taklimakan Desert, between 2013 and 2017. The results show that $Hg_P$ concentrations over the Taklimakan Desert atmosphere are remarkably higher than those observed from background sites in China and are even comparable to those measured in most of the Chinese metropolitan cities. Moreover, $Hg_P$ concentrations in the Taklimakan Desert exhibit a distinct seasonal pattern peaking during dust storm outbreak periods in spring and summer (March to August). A preliminary estimation demonstrates that export of total Hg associated with atmospheric dust from the Taklimakan Desert could be 59.7 ± 60.3 (1SD) Mg $yr^{-1}$. The unexpectedly high $Hg_P$ concentrations during duststorms, together with consistent seasonal pattern of Hg revealed from the snow/ice, clearly demonstrate that Asian desert dust could act as a significant carrier of atmospheric Hg to the cryosphere of Western China and even can have further global reach.


## 1. Introduction

The global atmospheric dust load is largely contributed by emissions from the North African (e.g., the Sahara Desert, 50–70%) and Asian deserts (10–25%) (Tegen and Schepanski, 2009). Asian deserts produce 50% of the global dust production (up to 800 Tg $yr^{-1}$) that can be dispersed not only locally but also globally in the Northern Hemisphere (Tegen and Schepanski, 2009; Huang et al., 2014a). About one quater of the dust emitted from Asian deserts is finally deposited in the ocean (Shao et al., 2011), even in the North Pacific Ocean and the west coast of North America after more than ten thousand kilometers long-range transport (Tegen and Schepanski, 2009). The major sources for Asian dust lie in deserts of western (e.g., Taklimakan Desert) and northern (e.g., Gobi Desert in Mongolia) parts of China, and Central Asia (Chen et al., 2017), where high winds generally mobilize significant amount of desert dust into the atmosphere (Zhang et al., 1997; Huang et al., 2014a). The impacts of desert dust are numerous and wide-ranging, including effects on economic development, atmospheric chemistry, climate processes, and biogeochemical cycling of various metals, etc. (Shao et al., 2011; Mahowald et al., 2014). For example, the impacts of desert dust hazard have repercussions for human

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society as a result of economic losses (Seinfeld et al., 2004; Middleton, 2017). Moreover, as the dominant contributor of atmospheric aerosols globally (Tanaka and Chiba, 2006; Middleton, 2017), desert dust may regulate the transportation and deposition of associated atmospheric pollutants (e.g., trace metals, persistent organic pollutants and microbes) over terrestrial ecosystems (Garrison et al., 2006; Federici et al., 2018) and micronutrients to marine ecosystems worldwide (Goudie and Middleton, 2006; Mahowald et al., 2018).

Mercury (Hg), a highly toxic trace metal, is a globally recognized pollutant and have a direct adverse effect on human health (AMAP/UNEP, 2018). In addition, longer residence time (~1 year) in the atmosphere (Schroeder and Munthe, 1998) allows a high dregree of bioaccumulation in food chain and various ecosystems (Larson, 2014; Selin, 2014). In the atmosphere, Hg exists in three operationally defined forms: gaseous elemental Hg (GEM or Hg(0) (>95%), gaseous oxidized Hg (GOM) and particulate bound Hg ($Hg_P$, Hg associated with atmospheric particulate) (GOM + $Hg_P$ <5%). Global Hg emissions to the atmosphere from anthropogenic and natural sources are estimated to be 2100 Mg $yr^{-1}$ and 5900 Mg $yr^{-1}$ (AMAP/UNEP, 2018), respectively. The Hg emissions are mainly in the form of Hg(0) but can undergo atmospheric oxidation in the presence of hydroxyl radical and halogen radicals to form divalent Hg(II) species. GOM, comprised of Hg(II) compounds, is readily adsorbed onto dust surface and can be transported along with desert dust or dissolved in cloud droplets, and therefore is easily deposited through wet/dry deposition (Lin and Pehkonen, 1999). Hence, desert dust, one of the largest contributor of global dust budget (Tanaka and Chiba, 2006; Middleton, 2017), may facilitate the transport and deposition of atmospheric Hg affecting its biogeochemistry downwind of the source region.

Western China has one of the most highly concentrated cryosphere at low- and mid-latitudes (Kang et al., 2010; Guo et al., 2015) (Fig. 1b). Thus, glacier snow/ice serves as excellent natural archives for documenting the modern and past changes of atmospheric Hg deposition (Loewen et al., 2007, 2008; Kang et al., 2016, 2019). Earlier studies from the snow/ice Hg records (Fig. S1 and Text S1) have suggested that the spatial distribution and temporal variations of Hg in glaciers over Western China were largely governed by atmospheric dust (Loewen et al., 2007; Huang et al., 2012a,b; 2014b; Zhang et al., 2012). Several studies have been conducted in an effort to understand the sources, speciation, atmospheric transport and deposition mechanisms of snow/ice Hg (Zhang et al., 2012; Huang et al., 2012a,b, 2014b; Sun et al., 2017). However, due to the lack of $Hg_P$ measurements from Asian deserts, it was still not clear whether Hg in glacier snow/ice of Western China is derived from long-range transport of Hg along with desert dust or from gas-particle Hg partitioning near the deposition site of glaciers. In addition, even measurement of atmospheric Hg (i.e., $Hg_P$) remain very scarce in Western China (Fu et al., 2008, 2012; Huang et al., 2016; Zhang et al., 2015), and there is no information available in terms of $Hg_P$ concentration and temporal variation over the desert atmosphere.

An earlier study on the Hg adsorption on atmospheric dust (i.e., $Hg_P$) have suggested that desert dust may play an important role in transport and deposition of Hg to the cryosphere of Western China and beyond (Paudyal et al., 2017). It is notable that Asian deserts, such as the Taklimakan Desert, are closely situated upwind of the Tibetan Plateau (Fig. 1) and thus, desert dust could serve as a significant carrier of anthropogenic Hg pollution via long-range transport. Hence, there is an urgent need of researches on the fate and spatio-temporal distribution of atmospheric Hg over these vast desert areas in Asia. In this study, measurement of atmospheric $Hg_P$ in total suspended particulate (hereafter, TSP) samples, for the first time, were conducted in China's largest desert (Taklimakan Desert) during five consecutive years (2013–2017). The main objective of our study is to provide new insights for better understanding the role of desert dust as a carrier for subsequent study of biogeochemical Hg cycling. The knowledge obtained from this study can serve as a precarious step for assessing the impacts of the desert dust on the biogeochemical Hg cycling at regional and global scales.

## 2. Materials and methods

### 2.1. Study area and sample collection

Western China mainly consists of the Tibetan Plateau (>4000 m a.s.l.) in the south and Xinjiang Uyghur Autonomous Region (hereafter, Xinjiang) in the northwest (Fig. 1). The large scale atmospheric circulation patterns over Western China (Fig. 1) are mainly dominated by westerlies (Yanai and Wu, 2006). The westerly jet stream (i.e., westerlies), known as a prevailing planetary wind system in the upper troposphere (Yanai and Wu, 2006), influences Western China in the whole year. The Taklimakan Desert, located in the Tarim Basin of Xinjiang with an area of 330,000 $km^2$, is China's largest desert and the world's second-largest shifting sand desert. The Taklimakan Desert is one of the key dust sources in the Northern Hemisphere (Zhang et al., 1997; Huang et al., 2014a). Although dusty weather (e.g., duststorm, floating dust and blowing dust) occurs in most of the year over the Taklimakan Desert, frequent dust outbreaks is a common phenomenon during spring and summer (from March to August) (Wang et al., 2004; Zhang et al., 2018). The Taklimakan Desert is characterized by strong windy days during duststorm season (Wang et al., 2004).

TSP samples were collected from five observational stations in the Taklimakan Desert, including the Kashgar (KS), Taxkorgan (TX), Minfeng (MF), Ruoqiang (RQ), and Tazhong (TZ) stations (Fig. 1, Table 1 and S1). The TSP samples from KS (n = 93, in 2013), MF (n = 149, in 2013) and TZ (n = 90, in 2014) stations were collected from 2013 to 2014, while the samples from RQ (n = 20) and TX (n = 9) stations were sampled in April 2017. The sample was collected on a quartz-fiber filter (length: 180 mm, width: 230 mm) (Whatman® GF/F, 2.2 μm, Maidstone, UK) for $Hg_P$ analysis using a middle-flow impact aerosol sampler (RP 1400a: Thermo-Fisher Scientific, Madison, WI, USA). Quartz-fiber filters are widely used for atmospheric $Hg_P$ sampling (Tsai et al., 2003; Huang et al., 2016; Guo et al., 2017), due to its easy manipulation, good mechanical strength, ability to withstand high temperature, and low Hg blank and flow resistance (Lu and Schroeder, 1999; Munthe et al., 2001; Lynam and Keeler, 2002). Each sampling was conducted for 22 h at a constant flow rate (100 L $min^{-1}$). The standard air volume was converted into standard conditions (25 °C, 101.3 kPa) according to the ambient meteorological conditions. Field blank filters (one blank filter each month) were placed in the sampler without air being drawn through it. The filters were baked (550 °C, > 6 h) to remove any Hg on the filters and were weighed by a microbalance (sensitivity: ± 0.01 mg) before and after sampling. Prior to weighing, all filters were equilibrated at constant temperature (25 °C) and relatively humidity (30%) conditions for > 24 h. After sampling, the filters were packed in plastic bags and stored in a cooler (4 °C) prior to analysis. In order to understand the background concentrations of Hg in the desert surface sediments, a total of 10 surface dust samples (depth: 0–2 cm) were collected from our five observational stations (2 samples from each station).

### 2.2. Analytical procedures and QA/QC

One-tenth of each sampled filter (2.27 $cm^2$) was punched for $Hg_P$ analysis, using a Leeman Hydra-$II_C$ Direct Hg Analyzer (Leeman

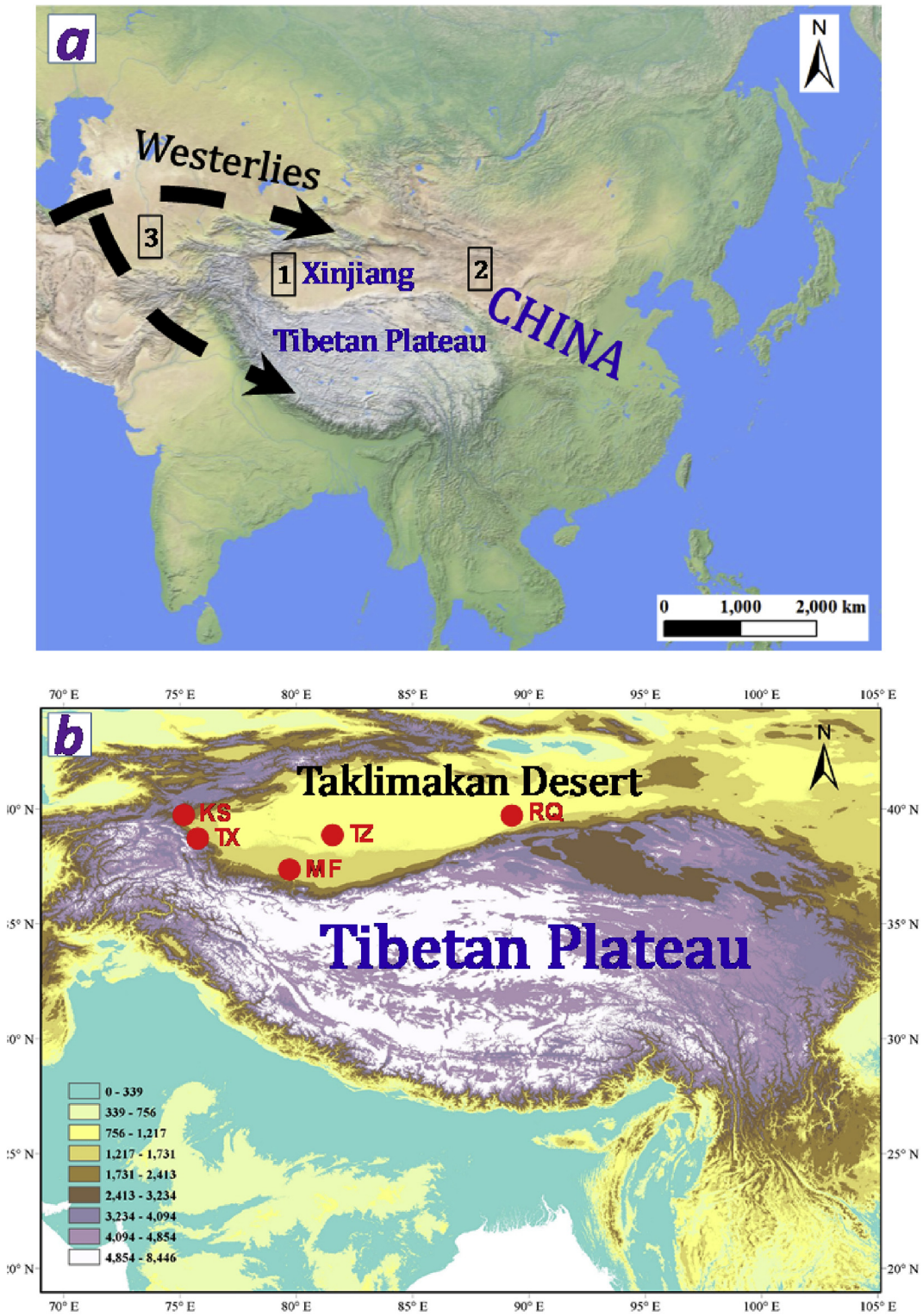


**Fig. 1.** Map showing the general pattern for atmospheric circulation system over Western China (***a***). The sampling locations (***b***) including the Kashgar (KS) station (75°45′ E; 39°29′ N, 1385.6 m a.s.l), Taxkorgan (TX) station (75°14′ E; 37°47′ N, 3093.7 m a.s.l), Minfeng (MF) station (82°72′ E; 37°04′ N, 1409.7 m a.s.l), Ruoqiang (RQ) station (88°10′ E; 39°02′ N, 889.3 m a.s.l), and Tazhong (TZ) station (83°39′E; 38°58′N, 1099.3 m a.s.l) in the Taklimakan Desert (1). Geographic locations of the other vast deserts surrounding the western China (***a***): (2) represents desert regions in East Asia including Gobi Desert, Mu Us Desert, Turpan Desert, Gurbantünggüt Desert, Junggar Basin, and Horqin Desert, China; (3) represents desert regions in Central Asia including Karakum Desert, Turkmenistan, Kyzylkum Desert, Uzbekistan, and Betpaqdala Desert, Southern Kazakhstan.

Lab Hydra, Teledyne Leeman Laboratories, Hudson, NH, USA) following a previous method (US EPA Method 7473). $Hg_P$ concentrations are reported in volume-based concentration (pg $m^{-3}$) and mass-based concentration (μg $kg^{-1}$), respectively. Here the mass-based concentration (μg $kg^{-1}$) is defined by the total Hg mass contained in the TSP sample ($Hg_P$/TSP). Quality assurance (QA) and quality control (QC) include method blanks, field blanks, and standard reference materials. The method detection limit (MDL) for filter samples, calculated using 3 times the standard deviation of 10 replicates measurements of a blank quartz-fiber filter, ranged from 0.06 to 0.52 pg $m^{-3}$ with an average of 0.31 pg $m^{-3}$ at the flow rate employed in our study. Standard reference materials, GSS-9 (soil) and Tort-3 (lobster) were measured with every 20 samples, resulting Hg recoveries of 93%–106% for GSS-9 (n = 18) and 98%–107% for Tort-3 (n = 18). The volume-based $Hg_P$ concentrations in field blanks (avg. = 0.48 pg $m^{-3}$, n = 20) were always comparable to the average MDL (avg. = 0.31 pg $m^{-3}$), indicating limited Hg contamination during sampling, transport, treatment and storage. Hg concentration in the dust samples was determined in duplicate in the same way as the filter samples. The standard reference material of GSS-9 was measured in every 5 samples, which yielded Hg recoveries of 99 ± 4% (n = 2).

## 3. Results and discussion

### 3.1. Overall concentrations

As shown in Table 1, the annual average volume-based $Hg_P$ concentrations from five stations are in the order of KS (516.7 ± 296.0 pg $m^{-3}$) > RQ (243.6 ± 101.2 pg $m^{-3}$) > TX (198.4 ± 93.6 pg $m^{-3}$) > MF (165.7 ± 175.1 pg $m^{-3}$) > TZ (86.1 ± 44.8 pg $m^{-3}$). It should be noted while comparing the annual average $Hg_P$ values of KS, MF and TZ that TX and RQ samples were collected only during duststorm season in April of 2017 (Table S1). In general, the volume-based $Hg_P$ concentrations reported in this study are significantly higher than those observed in background stations but within the range over urban areas of China (Table S1). For example, the averages of volume-based $Hg_P$ concentrations in the Taklimakan Desert (Table 1) were almost an order of magnitude higher than that observed in the background sites (e.g., Mt. Waliguan, 19.4 pg $m^{-3}$). The averages can even account for about 20% of TGM concentration value (1.33 ng $m^{-3}$) obtained from the inland Tibetan Plateau region (Yin et al., 2018). The high $Hg_P$ concentrations could be largely attributed to the adsorption of atmospheric Hg on dust surface (i.e., gas-particle partitioning) or resuspension of desert surface sediments that are rich in Hg. The first case seems the main cause of the high $Hg_P$ concentrations in this study, because desert surface sediments investigated in the present study showed extremely low Hg concentrations (avg. = 1.9 ± 0.4 ng $g^{-1}$, n = 10, Table S2). Although no information is available at this time on the Hg concentration and speciation in the atmosphere over the Taklimakan Desert, we believe that the Hg adsorbed on atmospheric dust particulates mainly come from long-range, regional and local sources. First, the upwind sources of anthropogenic Hg emission are principally Europe, the Middle East and Central Asia (Streets et al., 2019). Under control of the westerlies (Fig. 1), there could be a significant contribution of anthropogenic Hg pollution from the trans-Eurasian transport. Second, previous studies demonstrated that the Taklimakan Desert atmosphere has been influencing by anthropogenic emissions from surrounding oasis cities (Liu et al., 2010), indicating that anthropogenic activities in the Tarim Basin may contribute a substantial amount of Hg to the atmosphere through their adsorption on dust surfaces. Third, there is an evidence (Liu, 2013) that natural gas within the Tarim Basin has significantly elevated Hg concentrations, and numerous local gas flaring could easily result in the observed increase in Hg for desert atmospheric dust. Hg emission from the above-mentioned sources could greatly facilitate adsorption of Hg pollution on dust particulates in the atmosphere of Taklimakan Desert. Therefore, the unexpectedly high $Hg_P$ concentrations were for the first time found in the desert atmosphere, implying that desert dust could act as an important carrier of Hg pollution to the downwind region.

### 3.2. Seasonal patterns

As shown in Table 1, there was a significant seasonal variation of TSP over the desert atmosphere (excluding TX and RQ), which was great in the duststorm season with the highest TSP concentrations during duststorm outbreak period. Similarly, the volume-based $Hg_P$ concentrations at KS, MF and TZ stations peaked during duststorm outbreak period (Fig. 2). When we divide into two seasons (i.e., duststorm season and non-duststorm season) to consider this pattern, we found that the volume-based $Hg_P$ from KS, MF and TZ stations (Fig. 3a, c, 3e) were significantly positively correlated with dust particulates (TSP) during duststorm season. In the Taklimakan Desert, duststorms mostly occur during spring and summer seasons (March–August) (Wang et al., 2004) when the synoptic

**Table 1**
Averages of volume-based $Hg_P$ concentrations (pg $m^{-3}$) and mass-based $Hg_P$ concentration (μg $kg^{-1}$) and the estimates of annual amount of Hg associated with dust from the Taklimakan Desert. Dust emission flux was taken from Chen et al. (2017).

| Site | Average TSP Concentration (μg $m^{-3}$) | | Average Volume-based $Hg_P$ Concentration (pg $m^{-3}$) | | Average Mass-based $Hg_P$ Concentration (μg $kg^{-1}$) | | Dust Emission Flux (Tg $yr^{-1}$) | | Amount of Hg Associated with Dust (Mg $yr^{-1}$) | |
|---|---|---|---|---|---|---|---|---|---|---|
| | Duststorm season | Non-duststorm season | Duststorm season | Non-duststorm season | Duststorm season | Non-duststorm season | Duststorm season | Non-duststorm season | Duststorm season | Non-duststorm season |
| Kashgar (KS) | 966.2 ± 732.8 (n = 48) | 744.6 ± 394.1 (n = 45) | 540.0 ± 334.0 (n = 48) | 491.9 ± 250.7 (n = 45) | 776.8 ± 481.0 (n = 48) | 894.6 ± 797.2 (n = 45) | – | – | – | – |
| Minfeng (MF) | 2025.2 ± 1806.6 (n = 99) | 832.1 ± 455.9 (n = 50) | 206.8 ± 202.4 (n = 99) | 87.7 ± 44.4 (n = 50) | 142.9 ± 156.9 (n = 99) | 126.2 ± 70.8 (n = 50) | – | – | – | – |
| Tazhong (TZ) | 2150.1 ± 1805.6 (n = 60) | 491.9 ± 398.0 (n = 30) | 110.3 ± 43.5 (n = 60) | 61.7 ± 31.1 (n = 30) | 93.7 ± 79.1 (n = 60) | 171.7 ± 87.6 (n = 30) | – | – | – | – |
| Ruoqiang (RQ) | 2534.9 ± 657.6 (n = 20) | – | 211.6 ± 54.8 (n = 20) | – | 207.8 ± 83.5 (n = 20) | – | – | – | – | – |
| Taxkorgan (TX) | 2431.6 ± 586.3 (n = 9) | – | 223.2 ± 68.3 (n = 9) | – | 201.1 ± 78.4 (n = 9) | – | – | – | – | – |
| Taklimakan Desert | 1795.7 ± 1614.9 | 697.8 ± 432.1 | 257.1 ± 267.1 | 224.4 ± 252.8 | 275.5 ± 379.8 | 415.6 ± 602.2 | 129.3 | 58.0 | 35.6 ± 49.1 | 24.1 ± 34.9 |

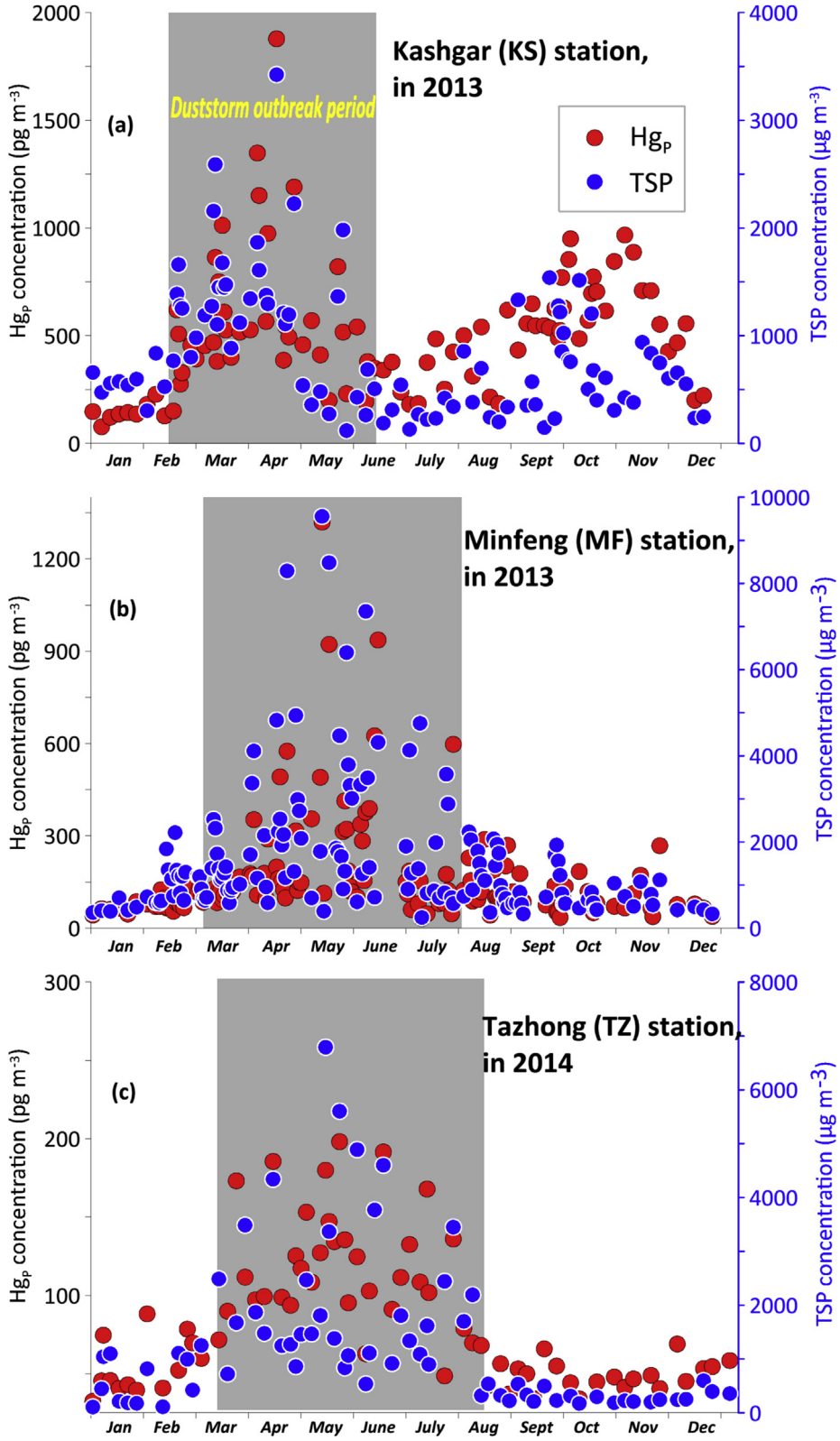


**Fig. 2.** Daily variations of volume-based $Hg_P$ and TSP concentrations in the desert atmosphere of KS (a), MF (b) and TZ (c) stations, respectively. The gray bars represent the period of duststorm outbreak.

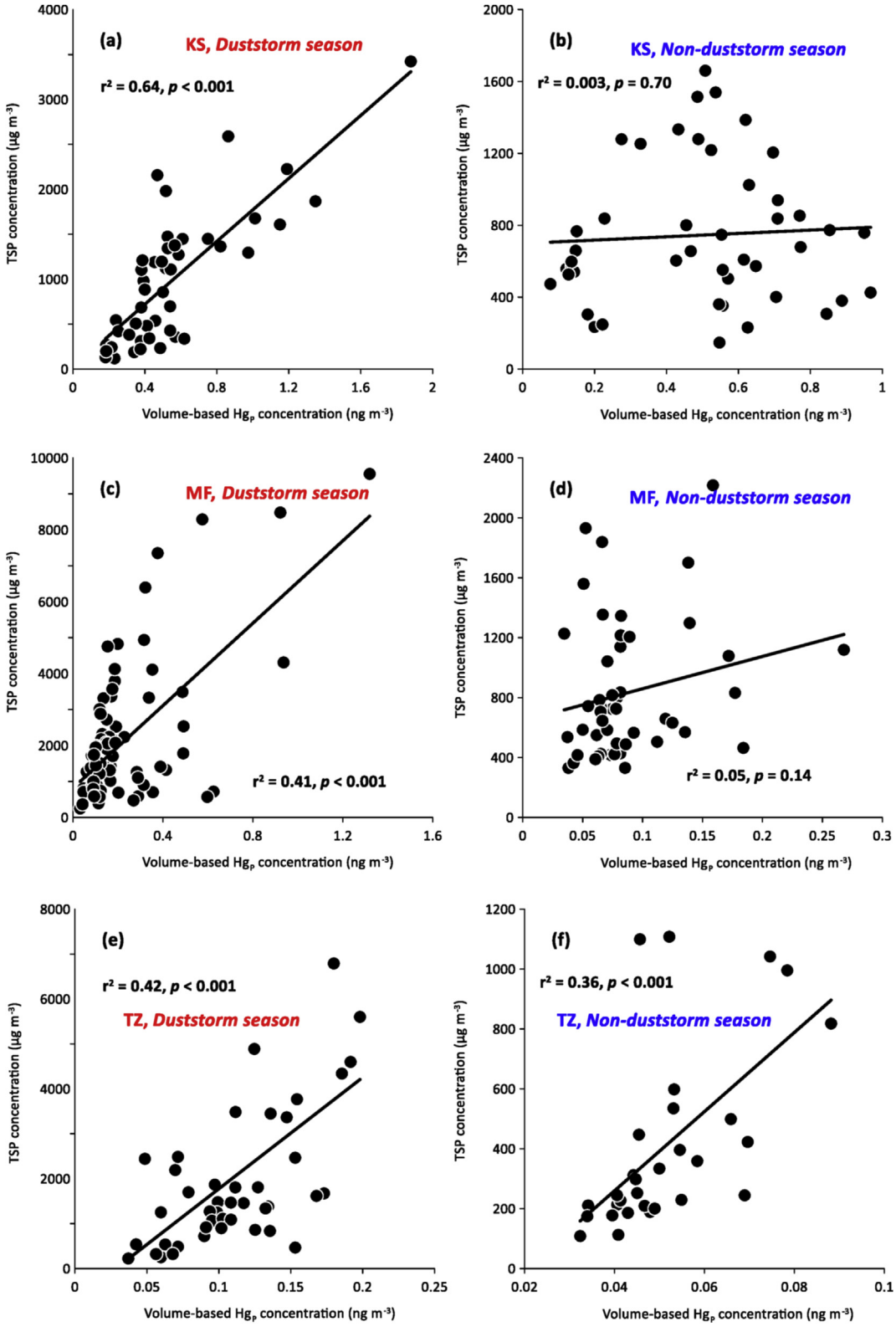


**Fig. 3.** Relationships between volume-based $Hg_P$ concentration and TSP concentration during duststorm season (in red) and non-duststorm season (in blue) at KS (a, b), MF (c, d), and TZ (e, f) stations. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

systems over East Asia are highly unstable and the climate is the driest of the year (Sun et al., 2001). Earlier studies have suggested that the majority of dust emissions from deserts occurred in the duststorm season (Xuan et al., 2000). The concentrations of atmospheric suspended particulates and tracers of dust (e.g., Ca and carbonate carbon) during duststorm season were found to be several times higher than that during non-duststorm season over the Taklimakan Desert (Zhang et al., 2017). Similarly, the high $Hg_P$

concentrations were simultaneously observed at our study sites during duststorm season (Fig. 2), indicating that desert dust favors the adsorption of atmospheric Hg on surface. The intensive dust emissions could provide a significant amount of particulate carriers and favorable conditions for gas-particle Hg partitioning. Therefore, the extensive dust particulates during duststorm season has led to a dramatic increase of $Hg_P$ in the desert atmosphere (Fig. 2).

During non-duststorm season, however, volume-based $Hg_P$ levels were found to be not significantly (Fig. 3b and d) or weakly (Fig. 3f) correlated to TSP concentrations. This finding indicates that some other factors such as regional/local anthropogenic sources and meteorological conditions (e.g., wind speed) are more influential than the dust particulates. For example, $Hg_P$ at KS station showed a peak during non-duststorm season (i.e. October and early November) in addition to there is another peak occurring during dust storm season (Fig. 2a). Since the KS station is located in close proximity ($\approx$20 km) to the Kashgar city, this $Hg_P$ peak during non-duststorm season could be greatly influenced by regional/local anthropogenic sources such as coal combustion for domestic heating in winter (Wang et al., 2015). Moreover, the wind speed is generally lower during non-duststorm season than that during duststorm seasons (Wang et al., 2004). In the non-duststorm season, the weaker wind and low ambient temperature can't greatly loosen the binding of dust from the desert land, and thus dust emission driven by land surface processes is relatively weaker. This seasonal pattern was also supported by the fact that the mass-based $Hg_P$ concentration was not correlated with TSP concentration during non-duststorm season (Fig. S2), further indicating either the presence of multiple sources affected and/or that the magnitude of gas-particle partitioning was different depending on the site and time. As a result, variability in some other factors (e.g., meteorological conditions) could add to the seasonal variability in $Hg_P$ and dust particulate, which leads to the different seasonal patterns at our study sites (Fig. 3).

### 3.3. Estimation of Hg amount associated with desert dust

Though occasional and irregular signals as discussed above could increase seasonal variability in the $Hg_P$ and dust particulate, a significant positive correlation was observed between volume-based $Hg_P$ and TSP for year-round samples from all five study sites (Fig. 4a). Similar to the volume-based $Hg_P$, moreover, the mass-based $Hg_P$ showed a generally similar trend of seasonal variation with TSP (Fig. S2). A significant positive correlation was consequently determined between volume-based and mass-based $Hg_P$ concentrations (Fig. 4b). Therefore, analysis of relationships among volume-based $Hg_P$, mass-based $Hg_P$ and TSP (Fig. 4) confirms that desert dust could be an important carrier in the atmospheric Hg transport and deposition.

The mass-based $Hg_P$ concentration could represent the capacities of dust particulates to contain Hg (Huang et al., 2016; Guo et al., 2017), which allows for a preliminary estimation of Hg amount associated with dust from the Taklimakan Desert. The Taklimakan Desert has the highest intensity of dust emission in East Asia (Chen et al., 2017). The total dust emission flux of the Taklimakan Desert is approximated to be 187.3 Tg $yr^{-1}$ (Table 1), and the dust emission during duststorm season alone accounts for ~ 69% of the total dust emission (Chen et al., 2017). Given the high intensity of dust emission and $Hg_P$ concentrations in our study region, we estimate the potential contribution of $Hg_P$ amount to the atmosphere along with the injection of desert dust by using the following equation:

$$F = C_{Hg} \times D \quad (1)$$

where F is the annual amount of Hg associated with desert dust (Mg $yr^{-1}$), $C_{Hg}$ is the mass-based $Hg_P$ concentration (μg $kg^{-1}$), and D is the dust emission flux (Tg $yr^{-1}$). The averages of $C_{Hg}$, as determined from this study, were taken as 275.5 ± 379.8 μg $kg^{-1}$ in duststorm season by the arithmetic means of KS, MF, TZ, TX, and RQ, and 415.6 ± 602.2 μg $kg^{-1}$ in non-duststorm season by the arithmetic means of KS, MF, and TZ, respectively (Table 1). The dust emission flux is estimated to be 129.3 Tg $yr^{-1}$ in the duststorm season and 58.0 Tg $yr^{-1}$ in the non-duststorm season (Chen et al., 2017). Therefore, the estimated amount of Hg associated with dust from the Taklimakan Desert is 59.7 ± 60.3 (1SD) Mg $yr^{-1}$, with amounts of 35.6 ± 49.1 (1SD) Mg of $Hg_P$ in duststorm season and 24.1 ± 34.9 (1SD) Mg of $Hg_P$ in non-duststorm season. Our estimates show a general consistency with duststorm activities and seasonal patterns of atmospheric $Hg_P$ levels (Fig. 2). The total $Hg_P$ amount (59.7 Mg $yr^{-1}$) could even account for 11% of Hg emissions from the desert/metalliferous/non-vegetated zones worldwide

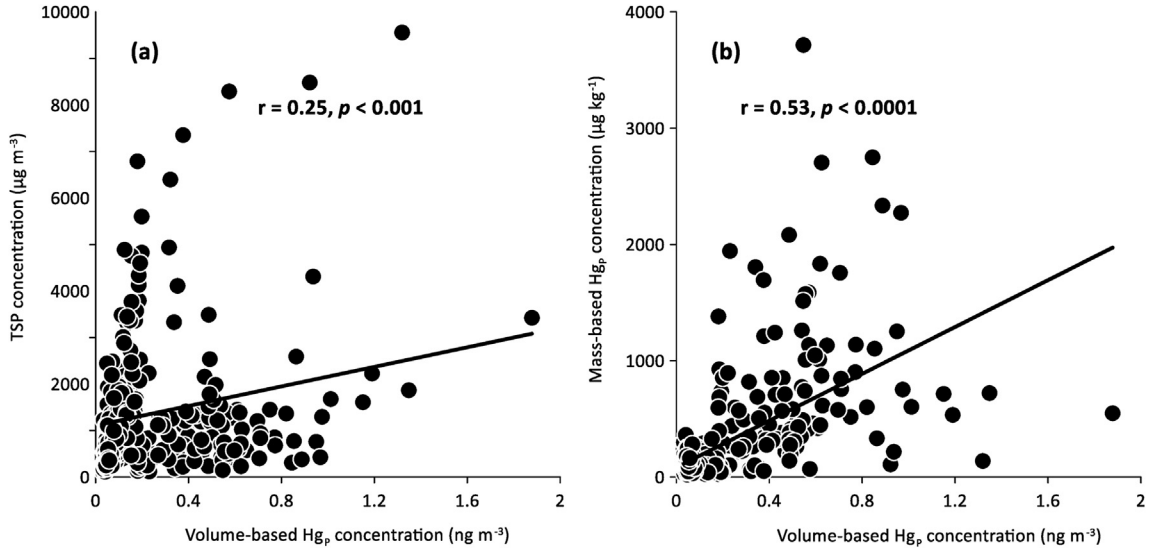


**Fig. 4.** Relationships between volume-based $Hg_P$ concentration and TSP concentration (a), and volume-based $Hg_P$ concentration and mass-based $Hg_P$ concentration for year-round samples of all five stations (b).

(546 Mg $yr^{-1}$) (Pirrone et al., 2010), suggesting that $Hg_P$ in the atmosphere of Taklimakan Desert is an important contributer to the global atmospheric Hg pool.

In particular, approximately 23% of dust emission from Asian deserts could be suspended in the atmosphere or subject to long-range transport, though 77% of total emission is redeposited onto the underlying surface by dry and wet deposition processes (Han et al., 2004; Chen et al., 2017). This indicates that 13.7 Mg $yr^{-1}$ of $Hg_P$ in the Taklimakan Desert atmosphere is prone to long-range transport, though our estimates contain substantial uncertainties such as the adsorption and partitioning of GEM and GOM in different size-ranges of particulates. In addiditon, though most of the Hg sorbed to the dust will likely be deposited within the desert itself (77%), the low concentrations in desert surface sediments (Table S2) suggest that this deposition is likely only temporary and the Hg will evaporate again. It is noteworthy that deserts are widely distributed in Asia such as the Gobi Desert which is situated in the northern parts of China with a comparable amount of dust emission (179.7 Tg $yr^{-1}$) to the Taklimakan Desert (Chen et al., 2017). It has been suggested that about 800 Tg of dust is injected annually into the atmosphere from Asia (Huang et al., 2014a), indicating the export of Hg associated with Asian desert dust could be an important Hg source to the downwind region.

### 3.4. Environmental implications

Our study reveals unexpectedly high concentrations of $Hg_P$ in the Taklimakan Desert dust and thus indicates that Asian deserts could act as important Hg sources to the downwind regions via long-range transport. This is further supported by a similar seasonal pattern revealed between the $Hg_P$ measurements and snow/ice Hg records (Loewen et al., 2007; Huang et al., 2012b), which was hypothesized earlier that duststorm activities could play an important role in atmospheric Hg deposition to the cryosphere of Western China. Hg in the snow/ice over Western China is primarily associated with dust particulate (Zhang et al., 2012; Huang et al., 2012 a,b; 2014b). Our study has essentially confirmed that the desert dust provides for a dominant mechanism for the gaseous Hg to be deposited to the cryosphere.

Furthermore, the snow/ice Hg records and speciation measurements of Hg in precipitation of Western China indicated that the dominant Hg species was mainly in the form of $Hg_P$ (Huang et al., 2012c, 2013), highlighting the importance of $Hg_P$ scavenging to atmospheric Hg deposition by wet/dry Hg deposition over Western China (Huang et al., 2012c, 2013). It is concluded, therefore, that the contributions of land surface processes from Asian deserts may significantly affect the environmental burdens of Hg on a regional scale. Our study highlights that Asian desert dust could act as a significant carrier of atmospheric Hg to the cryosphere of western China and beyond. Therefore, Asian deserts could have considerable impacts on biogeochemical Hg cycling on a regional scale.

More importantly, considering that duststorm activities in Asia are known to have global reach, delivering dust particulates to the Pacific Ocean (Huang et al., 2008; Eguchi et al., 2009), North America (VanCuren and Cahill, 2002), Greenland (Bory et al., 2002), Atlantic Ocean (Yumimoto et al., 2009), and on occasion even further (Uno et al., 2009), more studies addressing the impacts of long-range transport of atmospheric $Hg_P$ from Asian deserts are needed to have a better understanding of biogeochemical Hg cycling at a global scale.

## 4. Conclusions

Overall, the present study, for the first time, revealed high concentrations of $Hg_P$ in the desert atmosphere of the Taklimakan Desert. Adsorption of atmospheric Hg on desert dust could facilitate transport and deposition of Hg. A very distinct seasonal pattern for $Hg_P$ and resemblance between desert dust and snow/ice records indicate that the Taklimakan Desert as well as other Asian deserts are a significant carrier of atmospheric Hg to the cryosphere of Western China and beyond. Our results may further indicate that the vast coverage of deserts worldwide and their dust transports could be an important but unrecognized source of $Hg_P$ to the global atmosphere. Further studies are necessary to provide a better understanding of atmospheric $Hg_P$ from Asian deserts as well as global deserts. This paper provides new insights into the vital role of desert dust on Hg evolution to fully understand the biogeochemical Hg cycling at regional and global scales.

## Author contribution

Jie Huang: Conceptualization, Writing - original draft, Project administration. Shichang Kang: Supervision, Conceptualization, Project administration, Funding acquisition. Runsheng Yin: Conceptualization, Writing- Reviewing and Editing, Visualization. Kirpa Ram: Writing- Reviewing and Editing, Investigation. Xinchun Liu: Data curation, Resources. Hui Lu: Data curation, Investigation, Resources. Junming Guo: Software, Formal analysis, Methodology, Validation. Siyu Chen: Visualization, Methodology. Lekhendra Tripathee: Methodology, Visualization, Investigation.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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