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First Observation of Mercury Species on an Important Water Vapor Channel in the Southeast Tibetan Plateau

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Abstract

The Tibetan Plateau is generally considered to be a significantly clean area owing to its high altitude; however, the transport of atmospheric pollutants from the Indian subcontinent to the Tibetan Plateau has infected the Tibetan environments. Nyingchi is located at the end of an important water vapor channel. In this study, continuous monitoring of gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM), and particle-bound mercury (PBM) was conducted in Nyingchi from March 30 to September 3, 2019, to study the influence of the Indian summer monsoon (ISM) on the origin, transport and behavior of mercury. The atmospheric Hg concentrations during the preceding Indian summer monsoon (PISM) period (1.20±0.35 ng m^{-3}, 13.5±7.3 pg m^{-3}, and 11.4±4.8 pg m^{-3} for GEM, GOM, and PBM, respectively) were relatively higher than those during the ISM period (0.95±0.21 ng m^{-3}, 12.7±14.3 pg m^{-3} and 8.8±6.0 pg m^{-3}). The average annual total gaseous mercury concentration in the Nyingchi region was obtained using a passive sampler as 1.12±0.28 ng m^{-3}. The GEM concentration showed that the sampling area was very clean. The GEM has several patterns of daily variation during different periods. Stable high GEM

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

concentrations occur at night during PISM, which may be related to the nocturnal boundary layer. High values occurring in the late afternoon during the ISM may be related to long-range transport. The results of the trajectory model demonstrate that the sources of pollutants at Nyingchi are different under the control of different airflow fields. During westerly circulation, pollutants mainly originate from northeast India or Nepal. During the ISM period, the pollutants mainly originate from northeast India, or the Bay of Bengal, and the Indian Ocean. The strong precipitation and vegetation effects on Hg during the ISM resulted in low Hg concentrations transmitted to Nyingchi during this period. Further, principal component analysis showed that long-distance transport, local emissions, meteorological factors, and snowmelt factors are the main factors affecting the local Hg concentration in Nyingchi.

1. Introduction

Mercury (Hg) is classified as a hazardous pollutant because it is bio-accumulative and toxic. Generally, atmospheric Hg can be categorized into three major types: gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM), and particle-bound mercury (PBM) (Selin, 2009). The stable chemical properties of GEM coupled with its long atmospheric lifetime (approximately 0.3 to 1 year) makes GEM an important global pollutant (Selin, 2009;Travnikov et al., 2017). In contrast, GOM and PBM are easily removed from the atmosphere through wet and dry deposition because of their chemical activity, and have significant impacts on the local environment (Lindberg and Stratton, 1998;Seigneur et al., 2006). Both GOM and PBM have complex fundamental physiochemical properties and may have complicated relationships with other regional pollutants (Gustin et al., 2015). Understanding, identifying, and characterizing Hg sources and their global and regional transport mechanisms is crucial for global atmospheric Hg control and health effect research (UNEP, 2018). Since 2013, the Minamata Convention was established to control the global mercury pollution (UNEP, 2013a). Monitoring atmospheric Hg is an important prerequisite for implementing the convention. Currently, several Hg monitoring networks and studies have been established to better understand atmospheric Hg cycling. The Atmospheric Mercury Network (AMNet; Gay et al., 2013), Global Mercury Observation System (GMOS), and the Canadian Atmospheric Mercury Network (CAMNet) are the main monitoring networks operating in North America and Europe; majority of them only monitor GEM concentrations (Gay et al., 2013;Sprovieri et al., 2013;Sprovieri et al., 2016;Kellerhals et al., 2003). Researchers worldwide have also contributed to monitoring the data from different regions (Gustin et al., 2015;Jiang and Wang, 2019;Stylo et al., 2016). However, there exists some gaps in understanding the sources and transport of atmospheric Hg in some remote areas, especially in harsh environmental areas where performing monitoring is difficult.

Considering that GEM can be transported globally over long distances and that the transport distances of GOM and PBM vary greatly in different environments, atmospheric Hg concentration monitoring may not directly reflect the intensity of regional atmospheric Hg emissions. Our previous study of the Qomolangma National Nature Preserve (QNNP) (Lin et al., 2019) demonstrated that the Hg emitted from India can cross the Himalayas to reach the Tibetan Plateau. Further research on the transboundary transport of Hg should be conducted to better understand the transport mechanisms. This is particularly true in Asia, where the environmental pollution is generally severe. China and India are

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

reported to be the world's largest consumers of coal (BP Statistical Review of World Energy, 2018). Considering that coal is the largest source of Hg in the atmosphere (approximately 86% of Hg comes from fuel combustion (Chen et al., 2016)), both China and India have great Hg emission potential. Further research on pollutant transport in Asia should be conducted to support policy development and responsibility allocation.

The Tibetan Plateau, with an average elevation of more than 4,000 m above sea level, is a natural barrier between inland China and the Indian subcontinent (Qiu, 2008;Lin et al., 2019). In the southern part of the Tibetan Plateau, the Himalayas, with an average altitude of 6,000 m, can serve as a solid barrier to pollutant transport. However, this barrier cannot completely block the transboundary transportation of pollutants according to previous studies. The transboundary and long-distance transport of pollutants across the Himalayas has attracted considerable attention (Wang et al., 2018;Zhang et al., 2015a;Yang et al., 2018;Li et al., 2016;Feng et al., 2019;Zhu et al., 2019). Several studies have shown that the transboundary intrusion of atmospheric pollutants through the Himalayas on the Tibetan Plateau is crucial for many pollutants (Yang et al., 2018;Li et al., 2016;Zhang et al., 2015b;Pokhrel et al., 2016;Lin et al., 2019). Zhang et al. (2017) studied short-lived reactive aromatic hydrocarbons and indicated that the cut-off low system that have lower altitude in the Himalayas is a major pathway for long-distance transport of aromatic hydrocarbons in the Tibetan Plateau. Persistent organic pollutants have been reported to be transported to the interior of the Tibetan Plateau by traveling along valleys or across ridges (Gong et al., 2019a). The transport of aerosols and organic pollutants along the most important water vapor channel, the Yarlung Zangbu/Brahmaputra Grand Canyon (hereafter referred to as the YZB Grand Canyon), has been observed (Wang et al., 2015;Sheng et al., 2013). Our previous study in the QNNP, on the southern border of the Tibetan Plateau, proved that atmospheric Hg from the Indian subcontinent can be transported across high-altitude mountains directly to the Tibetan Plateau under the action of the Indian monsoon and local glacier winds (Lin et al., 2019). However, to the best of our knowledge, the monitoring of the passage of atmospheric Hg in the water vapor channel—the YZB Grand Canyon, into the Tibetan Plateau has not been conducted. Through the water vapor and airflow channel, air masses carrying large amounts of water vapor as well as pollutants may enter Tibet, resulting in heavy precipitation during the monsoon season. Huang et al. (2015) reported that the total Hg wet deposition in Nyingchi, located in the YZB Grand Canyon, was lower than that in other Tibetan Plateau regions, and the concentration was lower in the monsoon season than in the non-monsoon season. As an important transport channel for summer monsoon moisture into China (Xu et al., 2020;Feng and Zhou, 2012;Yang et al., 2013), the amount of water vapor transported into Tibet through this channel is considerable, and the transport of pollutants needs further investigation.

In this study, we set up high-precision Hg species monitoring in Nyingchi, southeastern Tibetan Plateau, covering both PISM and ISM periods. Hg passive sampling was also applied to cover the monitoring of the entire year. To the best of our knowledge, this is the first monitoring study of atmospheric Hg species in the most important water vapor channel of the Tibetan Plateau. To better identify the sources of Hg pollution and potential pollution areas, we combined real-time Hg monitoring data with backward trajectory analysis, cluster analysis, and potential source contribution function (PSCF) analysis. We also collected other pollutant concentrations and rainfall data near the monitoring station

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

during the same period to better analyze the source and transport characteristics of Hg. By combining the real-time monitoring data and model simulations, we attempted to better characterize the process of Hg entering Tibet through the water vapor channel, which could allow researchers to further analyze the transport of Hg from the Indian subcontinent into Tibet and provide scientific support for managerial decision making.

2. Materials and methods

2.1 Atmospheric Hg monitoring site

Atmospheric Hg monitoring was performed at the South-East Tibetan Plateau Station for Integrated Observation and Research of Alpine Environment (SET station, Figure 1) in Nyingchi, Tibet, China. The SET station is located in the southeastern part of the Tibetan Plateau (29°45′59N, 94°44′16E, 3263 m a.s.l.), in a water vapor transportation channel, from the Ganges River Plain to the Tibetan Plateau. Nyingchi is mainly under the control of westerly winds (from September to April) and ISM (from May to August), exhibiting sharp seasonal variations. The average annual air temperature is 5.6 °C. The Tibetan Plateau is generally a moisture sink in summer (Feng and Zhou, 2012;Xu et al., 2020), with climatological moisture originating from the Indian Ocean and the Bay of Bengal intruding into the center of the Tibetan Plateau along the water vapor channels. The average annual precipitation is approximately 700-1000 mm at the SET station. During the westerly control period, the air masses are mainly from mid-latitude inland areas with less water vapor, while during the ISM period, a large amount of water vapor from the Indian Ocean enters Tibet. The precipitation begins at the foot of the YZB Grand Canyon and is sustained along with the canyon into Tibet (Gong et al., 2019b), and the precipitation in the downstream Motuo County is more than twice that of the Nyingchi area (Ping and Bo, 2018). The unique geomorphological conditions and the effect of the strong monsoon have resulted in a unique high-altitude distribution pattern of various biomes and vegetation in the area. The SET station is 75 km from Bayi Town, where the capital of Nyingchi Prefecture is located, and 480 km from Lhasa, which is the capital city of the Tibet Autonomous Region. Owing to the high altitude and harsh living environment, the permanent population in Tibet is extremely small and only a few local pollutant emission sources have been observed (UNEP, 2013b;UNEP, 2018).

2.2 GEM, GOM and PBM active monitoring

Real-time continuous measurements of GEM, GOM, and PBM concentrations were carried out using Tekran Model 2537B, 1130, and 1135 instruments (Tekran Inc., Toronto, Canada) at the SET station from April 1, 2019, to September 4, 2019, which could show the diurnal and daily changes in atmospheric Hg concentration in detail. During the operation of the Tekran instruments, the sampling inlet was set at ~1.5 m above the instrument platform (shown in Figure S1). Considering the high altitude at which the instrument was installed, as well as to mitigate the impacts of low atmospheric pressures on the pump's operation, a low air sampling rate of 7 L min^{-1} for the pump model and 0.75 L min^{-1} (at standard pressure and temperature) for model 2537B were applied, based on the previous studies (Swartzendruber et al., 2009;Zhang et al., 2015a;Zhang et al., 2016;Lin et al., 2019). Air was sucked from the atmosphere in the Tekran instrument, and the Hg was divided into GOM, PBM, and GEM inside the instrument for analysis. Every 60 min, GOM was enriched on a KCL-coated annular denuder, PBM was enriched on a quartz fiber filter (QFF), and GEM was directly enriched on the gold tube of the Tekran 2537B. The collected

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

PBM and GOM were desorbed in succession to Hg(0) at temperatures of 800 °C and 500 °C in the following hour, respectively. Hg(0) was then measured by cold vapor atomic fluorescence spectroscopy (CVAFS) in the Tekran 2537B instrument. To ensure high data quality, the Tekran 2537B analyzer was set to use the internal Hg source for automatic calibration every 23 h. The instrument was calibrated using an external Hg source at the beginning and end of the monitoring period. The Tekran ambient Hg analyzer has been described in detail in previous studies (Landis et al., 2002;Rutter et al., 2008;de Foy et al., 2016;Lin et al., 2019). The monitoring data were also modified using the method from Slemr et al. (2016) as previous studies suggested that there may be a low bias for low sampling loads (Slemr et al., 2016;Ambrose, 2017).

2.3 Passive sampling of GEM concentration

Passive samplers were set up at the same station during and after the active monitoring period to better reflect the long-term pattern of local GEM concentration changes from April 2019 to March 2020. Sulfur-impregnated carbon (Calgon Carbon Corporation) was used as the sorbent for GEM (Guo et al., 2014;Zhang et al., 2012;Tong et al., 2016;Lin et al., 2017). Passive samplers were deployed in triplicate near the Tekran instrument at a height of ~2 m above the ground, and generally the passive samplers were replaced three times per month. After sampling, all samplers were sealed in a three-layer zip-lock bag and transported to the laboratory, where they were then measured with the DMA-80 (Milestone Inc., Itália). The passive sampling method has been successfully applied to the Tibetan Plateau (Guo et al., 2014;Tong et al., 2016) and North China (Zhang et al., 2012) in past studies. The use and quality control of the Hg passive sampler have been described in detail in our previous studies (Zhang et al., 2012;Guo et al., 2014;Lin et al., 2017).

2.4 Meteorological data and other pollutants data

During the monitoring period, the local temperature (with a precision of 0.1 °C), relative humidity (with a precision of 1%), wind speed (with a precision of 0.1 m s^{-1}), wind direction (with a precision of 1°), air pressure (with a precision of 0.1 hPa), solar radiation (with a precision of 1 W m^{-2}), and UV index (with a precision of 0.1 MEDs) were recorded at a 5-minute resolution by the Vantage Pro2 weather station (Davis Instruments, USA).

Hourly measurement data of $PM_{2.5}$, PM_{10}, SO_2, NO_2, O_3, and CO concentrations and AQI index were obtained from a nearby monitoring station in Nyingchi, which was hosted by the China Ministry of Ecology and Environment and published by the China National Environmental Monitoring Center. The measurements were conducted following the technical regulations for the selection of ambient air quality monitoring stations (National Environmental Protection Standards HJ 664-2013) (Yin et al., 2019).

2.5 Backward trajectory simulation

To better understand the source of atmospheric Hg, the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model was applied to calculate the backward trajectory many atmospheric particles (Stein et al., 2015;Chai et al., 2017;Chai et al., 2016;Hurst and Davis, 2017;Lin et al., 2019). HYSPLIT was developed by the US National Oceanic and Atmospheric Administration (NOAA) and is a known tool for explaining atmospherically transported, dispersed, and deposited of particles. The HYSPLIT model (https://www.arl.noaa.gov/hysplit/hysplit/) is a hybrid method that combines the Lagrangian and Euler approaches. The Lagrangian method calculates the movement of

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

matter under the action of advection and diffusion, and the Euler method uses a fixed three-dimensional grid to calculate the pollutant concentration. The backward trajectory simulation used Global Data Assimilation System (GDAS) data with 1°x1° latitude and longitude horizontal spatial resolution and 23 vertical levels at 6 h intervals. The trajectory arrival height was set to 1000 m a.g.l. Every backward trajectory was stimulated for 72 h at 6 h intervals, which can cover China, Nepal, India, Pakistan, and the majority of western Asia. Cluster analysis was performed after the trajectory calculation. Cluster analysis can summarize the concentrations of GEM and the main trajectories from all trajectories.

The PSCF model was used to calculate the conditional probability by calculating the endpoints of the trajectory segment ending in each cell, and the PSCF links the residence time in upwind areas with measured high concentrations (Ashbaugh et al., 1985;Hopke and Association, 2016). The value of PSCF represents the possibility that emissions from the source area will affect the air quality of the sampling site. In this study, we used PSCF to identify the possible source region of atmospheric GEM in Nyingchi. The PSCF values were also scaled by an arbitrary weighting function to reduce uncertainty owing to the low PSCF value (Polissar et al., 1999).

2.6 Principal components analyses

Principal component analysis (PCA) is a data reduction method that can group some measured variables into a few factors that can represent the behavior of the whole dataset (Jackson, 2005). PCA has been employed in many previous Hg studies to analyze the relationships between Hg and multiple pollutants and meteorological variables (Brooks et al., 2010;Cheng et al., 2012;Liu et al., 2007;Zhou et al., 2019). All variables were normalized by standard deviation prior to running the PCA. To ensure that PCA is a suitable method for the data set, the Kaiser-Meyer-Olkin measure of sampling adequacy (MSA > 0.5) and Bartlett's test of sphericity ($p < 0.05$) tests were performed during the data analysis process. Total variance and scree plots after rotation were used in the PCA analysis to determine the factor numbers. Components with variance ≥1.0 were retained. Variables with high factor loadings (generally > 0.5) were used to interpret the potential Hg source.

3. Results and discussion

3.1 Species Hg concentrations in Nyingchi

During the whole monitoring period, the GEM, GOM and PBM concentrations at SET station were 1.01±0.27 ng m^{-3}, 12.8±13.3 pg m^{-3}, and 9.3±5.9 pg m^{-3}(mean±SD), respectively. Figure 2 shows the GEM, GOM, and PBM concentrations and daily rainfall over the sampling period. Table S1 summarizes the statistical metrics of species Hg, meteorological factors, and other pollutants. To further discuss the patterns of Hg concentrations, the entire monitoring period was divided into the PISM period (before May 1) and the ISM period. The ISM period was further subdivided into three periods (ISM1 – ISM3) according to changes in precipitation. The atmospheric Hg concentrations during the PISM period (1.20±0.35 ng m^{-3}, 13.5±7.3 pg m^{-3}, and 11.4±4.8 pg m^{-3}, for GEM, GOM and PBM respectively) were higher than those during the ISM period (0.95±0.21 ng m^{-3}, 12.7±14.3 pg m^{-3}, and 8.8±6.0 pg m^{-3}, for GEM, GOM and PBM respectively). During the PISM period, the locally monitored GEM concentrations decreased continuously as the Indian monsoon developed and intensified (Figure 2), which may indicate a change in the local Hg source as the decisive wind field changes from westerly to Indian monsoon. GEM concentrations remained relatively stable during ISM1 and ISM2 (0.92±0.23 to 0.92±0.18 ng m^{-3}),

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

which may indicate that the source of GEM was relatively stable during this period. However, at the end of the monsoon (ISM3), the GEM concentration started to increase gradually to 1.04±0.21 ng m^{-3}. There was no significant correlation between GEM concentration and precipitation during the ISM period, which may be due to the stable chemical properties of GEM because the air mass sources are relatively stable during the ISM period (Horowitz et al., 2017), while GOM and PBM concentrations are strongly influenced by precipitation (Figure 2). With the increase in rainfall from 113.75 mm during ISM1 period to 373.28 mm during ISM2 period, the concentrations of GOM and PBM decreased sharply from 15.4±7.9 pg m^{-3} and 18.2±29.2 pg m^{-3} to 7.9±3.4 pg m^{-3} and 13.5±5.5 pg m^{-3}, respectively. The considerable precipitation increase may be responsible for the rapidly reduced GOM and PBM concentrations, as they are easily deposited in the atmosphere with precipitation (Lindberg and Stratton, 1998;Seigneur et al., 2006). In a previous study, Huang et al. (2015) found that even with heavy rain during the monsoon period, the total Hg concentration in precipitation in the SET region was small but still considerable, suggesting that there may be a stable source of Hg in the SET region during the ISM period.

Figure 3 shows the results of the GEM concentrations obtained through passive samplers throughout the year. The average GEM concentration is 1.12±0.28 ng m^{-3}, which is slightly higher than the average GEM concentration during the Tekran monitoring period. The average GEM concentration is lower (1.02±0.09 ng m^{-3}) during the ISM control period (from May to August) and higher during the westerly circulation control period (1.16±0.32 ng m^{-3}); however, the GEM concentration during westerly circulation control period has large fluctuations. Since there are almost no local industries and less human activity in Nyingchi, this difference may indicate a higher input of pollutants introduced by westerly circulation. For the variation throughout the year, the GEM concentration in May and June is the lowest with an average concentration of only 0.97±0.18 ng m^{-3}, while November and December have the highest GEM concentrations (1.24±0.37 ng m^{-3}).

Table 1 summarizes the GEM, GOM, and PBM concentrations from research papers of high-altitude regions around the world. Compared to other high-altitude sites, the Hg concentrations in the SET region were relatively low and did not reach the average Hg concentration level in the Northern Hemisphere (~ 1.5-1.7 ng m^{-3}). Compared to previous studies of high elevation (> 2000 m a.s.l) regions, only Concordia Station in Antarctica had lower GEM concentrations than those observed at the SET station. Ev-K2, Nam Co, Qomolangma, and Shangri-La, the nearest monitoring stations to the SET station and at higher altitudes, had higher GEM concentrations than those at the SET station. In particular, the GEM concentration at Shangri-La was more than two-fold of that at the SET station. The differences in the GEM concentrations among them may be mainly due to their different climatic conditions and different monsoon control zones, which result in different pollutant source regions and air mass transport trajectories. The Shangri-La station may be influenced by anthropogenic emissions within and outside China, and therefore, has higher GEM concentrations. For Ev-K2 and Qomolangma stations, which are under the influence of the ISM, they may be directly exposed to air masses with high concentrations of pollutants transported from India and Nepal. Although there are violent deposition processes during the climbing process to both Ev-K2 and Qomolangma stations, some Hg may survive reach the stations (Lin et al., 2019). The GOM concentrations at the SET station were approximately at the average level among

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

the monitored sites. PBM concentrations were relatively low at the SET station, which may be due to the high rainfall in the YZB Grand Canyon, easily washing away particulate Hg by rainwater.

The lower GEM concentrations during the ISM period may indicate that the pollutant sources of the SET region changed with the weakening of the westerly circulation and the strengthening of the Indian monsoon. In previous studies (Lin et al., 2019;Gong et al., 2019a;Wang et al., 2015), it is generally believed that pollutants from the heavily polluted Indian subcontinent may be transported to the Tibetan Plateau under the action of ISM, resulting in increased local pollutant concentrations on the plateau. This was verified at the Qomolangma, Nam Co, and Mt. Ailao stations, where GEM concentrations were higher during the ISM period than the PISM period (Lin et al., 2019;Yin et al., 2018;Zhang et al., 2016). However, in our study, the SET station observed lower species Hg concentrations during the ISM than the PISM period. For GEM, the decrease in concentration may be due to the absorption effect from the dense vegetation during the monsoon period (Fu et al., 2016), while air masses from the Indian Ocean bring large amounts of halogens, which may react with and deplete GEM. For GOM and PBM, increased concentrations were observed during the ISM1 period, whereas their concentrations decreased sharply during the ISM2 and ISM3 periods. The decreases in GOM and PBM concentrations may be mainly due to the rapid increase in local precipitation during the Indian monsoon period, which starts after the monsoon enters China from northwestern India. A large amount of water vapor from the Indian monsoon climbs more than 3,000 m within ~100 km in the YZB Grand Canyon, producing considerable precipitation. Therefore, GOM and PBM may settle during transportation and are unable to reach the Nyingchi area.

3.2 Diurnal Variation

Figure 4 shows the diurnal variation of species Hg and the concentrations of other pollutants during the entire monitoring period. In general, the species Hg concentrations varied significantly during the PISM period, and the diurnal variation was relatively small after entering the ISM period. During the PISM period, the GEM concentrations were relatively low during the daytime (average 1.07 ng m^{-3} from 11:00 to 18:00), gradually accumulated after sunset, and finally reached a relatively stable high value (average 1.26 ng m^{-3}) at night. During the ISM period, the GEM concentration variation pattern was not as pronounced as during the PISM period, with the lowest GEM concentration of the day usually occurring around sunrise (0.83, 0.80, 0.88 ng m^{-3} for ISM1-3, respectively). During ISM1, the GEM concentration reached a high value around 9:00 a.m., fluctuated less during the daytime, reached a maximum value in the evening, and gradually dissipated in the early morning. During ISM2, the maximum value was reached at approximately 16:00, was more stable in the evening, and gradually dissipated in the early morning. During ISM3, the maximum value was reached at approximately 20:00 and dissipated in the early morning. The average of the daily maximum values were 1.04, 1.00, 1.16 ng m^{-3} for ISM1-3 periods, respectively. After midnight, GEM concentrations gradually decreased. In general, the daily variation of GEM in previous research were about 0.2-0.9 ng m^{-3} globally, and were lower at the SET site (0.21, 0.20, 0.28 for ISM1-3 periods, respectively). For GOM and PBM, the diurnal variations showed U-shaped variation patterns during the PISM period. During this period, the concentrations of GOM and PBM reached low values between 10:00 and 14:00, then gradually accumulated and peaked around midnight. After midnight, the concentration gradually decreased to its

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

lowest point. During the ISM1 period, GOM and PBM concentrations were higher in the afternoon and evening, and showed a decreasing trend after midnight. During ISM2-3, GOM and PBM did not show clear daily variation patterns. Except for the ISM2 period, there was little difference between GOM and PBM concentrations during the other periods, which may be due to similar sources and behavioral patterns in the environment. In contrast, during the ISM2 period, more precipitation (Figure 2) led to a sharp decrease in PBM concentrations, and it is speculated that GOM may have additional sources during this period.

Compared with other Hg monitoring in previous studies, some diurnal variation trends of Hg at the SET site were unique. In previous studies (Sprovieri et al., 2016;Yin et al., 2018;Zhang et al., 2015a;Zhang et al., 2016;Fu et al., 2012;Fu et al., 2010;Lan et al., 2012), a common pattern of highest concentration around noon and lowest concentration before sunrise was mostly observed. The decrease in GEM concentration at night may be due to the chemical dissipation of GEM within the stable nocturnal boundary layer, or deposition in the effect of dew (Mao et al., 2008;Kim, 2010). After sunrise, partial GEM re-emission occurs in the sunlight, along with the mixing effect of the residual boundary layer downward, which may lead to an increase in GEM concentration (Mao and Talbot, 2012;Selin et al., 2007;Weiss-Penzias et al., 2009;Talbot et al., 2005). The height of the boundary layer increases after noon during the daytime, which produces dilution of GEM at the surface and may be the reason for the decrease in GEM concentration in the afternoon. The GEM diurnal variation pattern at the SET is particularly special during the PISM period, while a similar variation pattern was also observed at the Qomolangma site in our previous research (Lin et al., 2019), which is another high-altitude site with a sparse population and rare industry. This similar pattern suggests that they have a similar mechanism of GEM diurnal variation. Considering that neither site has an obvious local source of GEM, the variation in GEM concentrations may only be subject to these mechanisms. Similar to the study of Qomolangma, the variation in the boundary layer height may be one of the reasons for the diurnal variation of GEM concentration in the SET region. The stable and low height nocturnal boundary layer at night causes the GEM concentration to gradually concentrate, and the boundary layer gradually increases to a higher altitude after sunrise. The gradual increase in GEM concentration during the daytime may be due to the reduction of GOM from nearby local snowy mountains (Holmes et al., 2010) or the field GEM source brought in by airflow (Lin et al., 2019). During the ISM period, the nighttime GEM dissipation may be due to the fact that this area enters a rapid leaf-growing season (Fu et al., 2016) after entering the ISM period, that the air masses from the Indian Ocean bring a large amount of halogens, and that depletion of GEM occurs under the boundary layer at night.

3.3 Source identification for atmospheric Hg in Nyingchi

To further investigate the contributions of different sources to the SET site, air mass back trajectory simulation and trajectory cluster analyses were performed. Figure 5 shows the cluster analysis results for the PISM and ISM1-3 periods. Based on the results of the total spatial variation index, 3-5 clusters were grouped for each period. Each clustered trajectory contained detailed information about the trajectory from the source region to the SET site, the trajectory frequency during the period, and the concentrations of the pollutants carried by the air mass when the trajectory arrives.

During the PISM period (Figure 5a, Table S2), the trajectories mainly originated from or passed

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

through Nepal, northeastern India, and central Tibet, and moved along the southern border of the Himalayas Mountains. During this period, Nyingchi was mainly controlled by westerly circulation. The two clusters with the highest concentrations (traj1 and traj2, with GEM concentrations at 1.21 and 1.20 ng m^{-3}) originated from or passed through central Tibet, accounting for 35.32% of all trajectories in this period. Although the GEM concentrations of the two clusters were relatively high during this period, they were still lower than the background GEM concentration in the Northern Hemisphere (~ 1.5-1.7 ng m^{-3}), indicating that the air mass transported to SET station is relative clean. Traj0, from the southern border of the Himalayas, was relatively high in proportion (with a frequency of 61.28%), mainly controlled by the southern branch of the westerly circulation, and has a relatively low concentration (1.05 m^{-3}). This cluster made a turn in the Bay of Bengal and began to ascend toward the Tibetan Plateau, according to UNEP reports (UNEP, 2018;UNEP, 2013b), Hg emission intensities along the trajectory paths were weak. In addition, the air mass arriving at the SET site during this period came from the interior of the continents and contained less water vapor, which may lead to less precipitation and less Hg deposition during the transboundary transport.

During the ISM period (Figure 5b-d), the trajectory of arrivals at the SET site changed significantly with the onset and rise of the Indian monsoon. The trajectory path gradually shifted from the southern margin of the Himalayas to the Bay of Bengal. As the source of the air mass changes and the monsoon enters the plateau, it is possible that the concentration of pollutants decreases because of the change in the source region. Because the air mass originated from the Indian Ocean, the concentration of water vapor should increase significantly. During the ISM1 period (Figure 5b, Table S2), both the rising monsoon and the tail of the westerly circulation control the region, causing the transported air masses to exhibit complex trajectories and combined effects. The cluster with the highest concentration (traj2, 0.96 ng m^{-3}, and 29.0%) mainly came from or passed through central India. The trajectory with the largest proportion (traj0, 61.45%) had a relatively short path, mainly from northwest India and northern Bay of Bengal, and showed relatively lower concentrations (0.88 m^{-3}). Based on the existing atmospheric Hg emission inventories (Simone et al., 2016;UNEP, 2018;UNEP, 2013b), the Hg emission intensities of northwest India and the Bay of Bengal are very low, which may be the reason for the low GEM concentrations in this cluster. It is noteworthy that although the concentrations were low throughout the period, the trajectories of the different sources showed large concentration differences. Figure S2 shows all the trajectories during the ISM1 period and the GEM concentrations they represent. GEM concentrations are still high in trajectories originating from or passing through northern India and Nepal, which may be related to the high intensities of Hg emissions from anthropogenic sources in those regions (De Simone et al., 2015) and wildfire events (Lin et al., 2019;Finley et al., 2009). Trajectories from central Tibet also have high concentrations, which may be caused by local emissions such as yak dung burning (Lin et al., 2019;Huang et al., 2016), and the local sources do not pass through the areas with high deposition.

During the ISM2 period (Figure 5c, Table S2), a typical period of Indian monsoon control, almost all trajectories came from or passed through the southern part of the SET site and were influenced by the monsoon. Only about 5.4% of the trajectories originated from central Tibet with very low GEM concentration (traj0 with 0.81 ng m^{-3} and 3.17%, traj4 with 1.00 ng m^{-3} and 2.24%). Other trajectories

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

were almost identical, but with different track lengths, and showed low GEM concentrations (traj1 with 0.90 ng m^{-3} and 70.34%, traj2 0.94 ng m^{-3} and 19.40%). During this period, trajectories that originated in the Indian Ocean brought a large amount of water vapor and caused considerable precipitation during transportation. The Hg carried in the air masses may have been heavily deposited during transport. At the same time, the areas through which the trajectory passed were sparsely populated and underdeveloped and were unable replenish Hg to the air masses. The range of atmospheric mercury concentrations during the ISM2 phase was extremely small (Figure 2), which may indicate that under the strong control of the Indian monsoon, the main source region, transport path, and mechanism of transportation during this period remain stable. Compared with the ISM1 period, although many wildfires existed during this period, the large amount of precipitation may have prevented the dispersion of these pollutants from the wildfire sites.

During the ISM3 period (Figure 5d, Table S2), the Indian monsoon remained controlling this region, but its intensity weakened, and the precipitation in the Nyingchi area was greatly reduced. Most of the trajectories still came from northwest India (traj0 and traj3, 46.2% and 45.11%, respectively), and the trajectories remained short. But the GEM concentration at SET increased compared with the ISM1-2 period (average at 0.92 ng m^{-3} in ISM1 and ISM2 to 1.04 ng m^{-3} in ISM3 period). At the end of the ISM3 period, the GEM concentration showed an upward trend (Figure 2), which may be due to the weakening of monsoon control. A shortened trajectory at the end of the monsoon period was also observed in another study at a nearby site (QNNP) (Lin et al., 2019), which may indicate the withdrawal of the monsoon.

To visualize the potential Hg source areas during the monitoring period, PSCF simulation was also applied in this study in combination with the backward trajectories and detailed Hg monitoring at the SET station. Figure 6 shows the potential source regions of Hg during the PISM and ISM periods. The potential source areas of Hg did not change significantly throughout the study period and were mainly concentrated in the YZB Grand Canyon and the southern plains of the Canyon, south of Nyingchi. Due to the complex terrain, even the strong Indian monsoon can linger in the canyon for a long time because of the elevation rise and tortuous path. It is worth noting that heavy precipitation began when the monsoon began to climb and continued until the end of climbing. Therefore, before arriving at the SET station, Hg underwent multiple deposition processes. This may be the reason for the extremely low Hg concentration at the SET station during the monsoon period.

3.4 Hg concentration controlling factor indicated by PCA results in Nyingchi

Overall, 4-5 factors were resolved for each period from the PISM to ISM3 periods. Some factors are unique to each period, and certain factors are found throughout the monitoring period. Table 2 lists the four underlying PCA factors for important Hg-related components. They were assigned as long-distance transport, local emissions, meteorological factor, and snow melt factor.

The long-distance transmission factor (F1) found in the PISM and ISM3 periods mainly contain GEM, wind speed, CO (positive loading), temperature, and SO_2 (negative). GEM could be considered an indicator of long-distance transportation due to its long lifetime in the atmosphere, especially when GOM and PBM are not significant in this factor. This factor indicates that the long-distance transportation of GEM may mainly occurs in the pre-monsoon and the end of the monsoon period, which is similar to the trajectory analysis in Section 3.3. The negative correlation between GEM and temperature may

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

indicate that the long-distance transport of GEM during the PISM period occurs mainly during periods of lower temperatures. Compared with the diurnal variation of GEM during the ISM period (Figure 4), it is possible that the increasing GEM concentration in the evening in the PISM period is mainly due to the long-distance transportation of GEM.

Factor 2 involved GOM and PBM (high positive loading) in each period, mainly with positive O_3, PM_{10}, $PM_{2.5}$, and negative temperature. GOM concentrations were positively correlated with PBM concentrations, which implies that these two species probably originated from the same sources. The high positive loadings of PBM, GOM, and some particle pollutants may indicate that the main source of PBM and GOM is local emissions. The long-distance transport of particle pollutants from the Indian subcontinent may have heavy wet deposition when the air mass climbs into the Tibetan Plateau and cannot reach Nyingchi successfully. Thus, the local monitored particle pollutants, as well as easy-deposition pollutants, may mainly originate from regional emissions. One possible source is from yak dung; in the Tibetan Plateau, yak dung is a widely used household biofuel (Xiao et al., 2015) and the burning of yak dung may release Hg and other particulate matter.

Meteorology factor (F3) was found during the ISM period with positive temperature, wind speed, solar radiation, and negative humidity and rain, which are likely associated with meteorological conditions. This factor shows that meteorological conditions may profoundly affect the overall local pollutant distributions during the ISM period, which suggests that the air mass carried by the ISM not only cannot increase the long-distance transportation of pollutants to the Nyingchi area, but may also reduce the local contribution of pollution. For existing pollutants, the strong positive loading of solar radiation may indicate that pollutant reactions under strong radiation are relatively active in this high-altitude region. The strong negative humidity and rain may indicate that rain has played a strong role in the cleaning process, especially during the ISM1 and ISM2 periods, when precipitation is relatively strong.

Factor 4 had a strong positive correlation with GEM, ROM, and solar radiation, and negative loading with humidity during the ISM1 period. This suggests that as solar radiation increases in the afternoon (Figure 3), more GEM and GOM are emitted to the air, which may reflect the snow/ice melt process (Huang et al., 2010;Dommergue et al., 2003). Previous studies have shown that the snowmelt process can increase atmospheric GEM concentration (Huang et al., 2010;Dommergue et al., 2003). GEM may come from the evaporation of snow melting or be driven by the photoreduction of snow Hg^{II} (Song et al., 2018). Previous simulations indicated that the oxidation of GEM may occur at the snow/ice interface in the action of solar radiation, leading to extra GOM release in the ISM1 period. The peak concentrations of GEM and GOM both appeared in the afternoon during the ISM1 period, when the solar radiation was the highest and humidity was the lowest. The increase in GEM and GOM concentrations may be related to solar radiation, according to the PCA results.

3.5 Implications

The Tibetan Plateau is a direct invasion target of the ISM. Blocked by the high altitude of the Himalayas, the Indian monsoon could bypass the high mountains and enter Tibet via the YZB Grand Canyon. When the summer monsoon enters Tibet, it is generally believed that pollutants from India and the Indian Ocean, as well as large amounts of water vapor, may be carried along with the air masses (Lin

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

et al., 2019;Yang et al., 2013;Wang et al., 2018). Located in the water vapor channel where the Indian monsoon enters, Nyingchi is believed to receive a large amount of foreign air mass (Yang et al., 2013). Considering that Nyingchi has little local emission because of the sparse population and lack of industry, the pollutants present in the area should mostly have been transported by monsoons over long distances. However, our monitoring results show that during the ISM period, the Hg concentrations in the Nyingchi are extremely low (0.95±0.27 ng m^{-3}); lower than the background Hg concentration in the Northern Hemisphere and the Hg concentrations observed at surrounding monitoring sites in the literature (Table 1).

The low concentration during the ISM period may be related to the regional deposition process and complex regional terrain. When monsoon winds carry large amounts of Indian Ocean moisture and enter the YZB Grand Canyon, strong wet deposition occurs during transport due to an increase in elevation and a decrease in temperature. The process of rainwater scouring from wet deposition may result in significant deposition of pollutants from carried air masses (Lindberg and Stratton, 1998;Seigneur et al., 2006). Meanwhile, the air flow in the canyon is slow owing to the complex terrain. The slow migration of the air mass further strengthens the deposition process. In addition, during the ISM period, the dense forest in the canyon may deplete some of the mercury during transport (Fu et al., 2016). Therefore, pollutants from the Indian subcontinent struggle to go deep into the Tibetan Plateau during the ISM period. The deposited pollutants may flow into the downstream area via rivers to Southeast Asia and South Asia, and these transportation mechanisms needs to be further studied. However, long-distance transboundary transport remains an important mechanism of Hg distribution in this area during the period of westerly circulation. As discussed in Section 3.1, the Hg concentration in Nyingchi during PISM period (1.20±0.35 ng m^{-3}) was much higher than that during the ISM period (0.95±0.27 ng m^{-3}). The high Hg concentration during the PISM period may indicate that a large amount of external source Hg entered the Nyingchi area during the non-ISM period, which needs to be further studied.

The results of our previous study on Qomolangma were different from those in Nyingchi. At Qomolangma(Lin et al., 2019), the ISM was the main period of transboundary transport of Hg, and the PISM period accounted for a smaller amount of transport. The difference in Hg transportation patterns between the two sites may mainly originate from the difference in the geographical location, with different actions of the westerly circulation and the Indian summer wind. Qomolangma is located on the northern side of the Himalayas, a typical terrain on the southern side of the Tibetan Plateau. The Nyingchi area is another typical pathway for air masses to enter the Tibetan Plateau. Together, they describe two typical transboundary transport patterns of Hg occurring on the Tibetan Plateau at different periods of the year.

4. Conclusions

Comprehensive species Hg monitoring was carried out in Nyingchi, a high-altitude site in the southeast of the Tibetan Plateau. Nyingchi is located on the main pathway for water vapor carried by the monsoon to enter Tibet during the ISM period, which could characterize the spread of pollutants from the Indian subcontinent. The atmospheric Hg concentrations during the PISM period (1.20±0.35 ng m^{-3}, 13.5±7.3 pg m^{-3}, and 11.4±4.8 pg m^{-3}, for GEM, GOM, and PBM respectively) were relatively higher than those during the ISM period (0.95±0.21 ng m^{-3}, 12.7±14.3 pg m^{-3}, and 8.8±6.0 pg m^{-3}, for GEM,

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

GOM, and PBM respectively). The Hg concentration in Nyingchi is particularly low compared with other high-altitude stations around the world. GEM concentration shows a distinct and unique diurnal variation, with a gradual increase in GEM concentration during the day and a maximum concentration at night. This diurnal variation may be due to the re-emission of GEM by snowmelt and the trapping effect of pollutants by the planetary boundary layer at night.

According to the trajectory model, atmospheric Hg in the Nyingchi area originated mainly from or passed through Nepal and central Tibet during the PISM period. During the ISM period, Hg may mainly originate from northeast India and the Bay of Bengal. During the ISM period, the Hg concentrations were particularly low because of the strong deposition process during transportation. Through comprehensive PCA analysis using local meteorological conditions and multiple pollutants, long-distance transport, local emissions, meteorological factor, and snowmelt factor have been identified to affect local Hg concentrations. In the Nyingchi area, Hg mainly comes from westerly circulation during the non-ISM period. During the ISM period, pollutants from the Indian subcontinent may travel to South Asia and Southeast Asia along the major rivers in this area.

Acknowledgments

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Data availability. All the data presented in this paper can be made available for scientific purposes upon request to the corresponding authors.

Author contributions. HL,XW, YT, QZ and XY designed the research and performed field measurements. HL YT and CY performed the data analysis and model simulations. HL led the paper writing. LC,SK,LL,JS and BF contributed to the scientific discussion and the paper preparation.

Competing interests. The authors declare that they have no conflict of interest.

Figure captions

Figure 1. Location of the South-East Tibetan Plateau Station for Integrated Observation and Research of Alpine Environment (SET station or Nyingchi station, the red star). SET station is located in a water vapor channel from the Ganges River Plain to the Tibetan Plateau. The red dot is Lhasa, the capital city of the Tibet Autonomous Region, which is the most densely populated city in Tibet; the other red dot is the nearest town to the monitoring site, Bayi Town.

Figure 2. Time serious of GEM, GOM, and PBM concentrations and the daily rainfall over the

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

sampling period. The GEM concentration resolution is 5 min, and the GOM, PBM, and rain resolutions are 2 hours. According to the characters of monsoon development and precipitation, the monitoring periods are divided into four segments, namely PISM (before May), ISM1 (1 May- 2 June), ISM2 (3 June – 8 August), and ISM3 (after 9 August).

Figure 3. GEM concentrations obtained through passive samplers throughout the year. The black squares represent the atmospheric Hg concentrations obtained by passive sampling, and the upper and lower error lines are the standard errors of the passive samples monitored during the same time period. The red dots represent the GEM concentrations obtained through the Tekran instrument. The green horizontal line indicates the average of the atmospheric mercury concentrations during this period.

Figure 4. Diurnal variation of species Hg, concentrations of some other pollutants and meteorological information from PISM to ISM1-3 periods. The short horizontal line represents the concentration error range for each time period.

Figure 5. Clusters of the back trajectory analysis from SET site during PISM to ISM3 periods. The thickness of the line represents the ratio of the cluster in the time period, and the color of the line represents the GEM concentration carried by the cluster, the background is the globally Hg emission inventory developed by UNEP(UNEP, 2013a).

Figure 6. Potential source regions and pathways for the results of PSCF analysis. The PSCF values represent the probability that a grid cell is a source of Hg. (From a-d are PISM, ISM1-3 periods respectively)

Table captions

Table 1. Comparison of atmospheric Hg concentrations at high elevation (> 2000 m a.s.l) stations

Table 2. PCA factor loadings (Varimax Rotated Factor Matrix) for Hg in Nyingchi, Tibet, China

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

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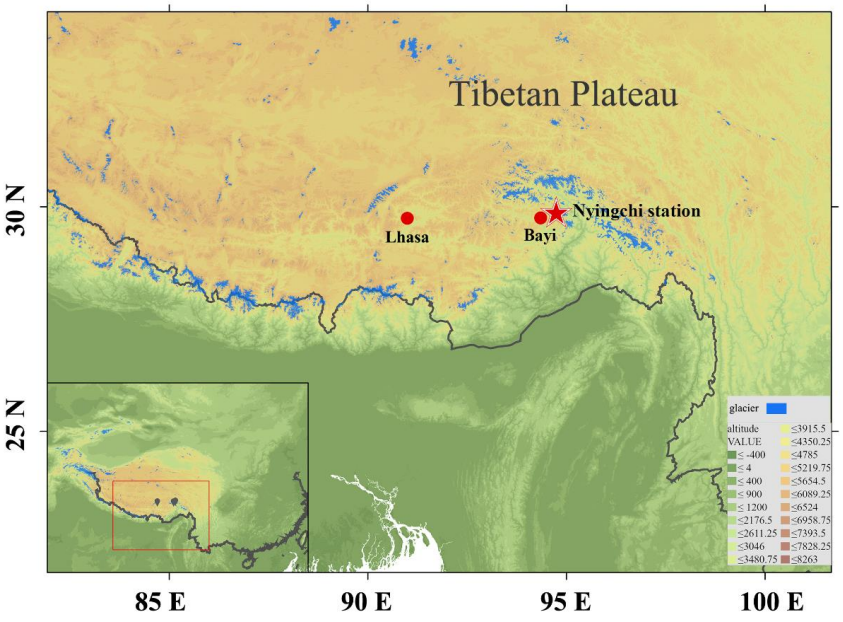
Figure 1



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Preprint. Discussion started: 27 September 2021

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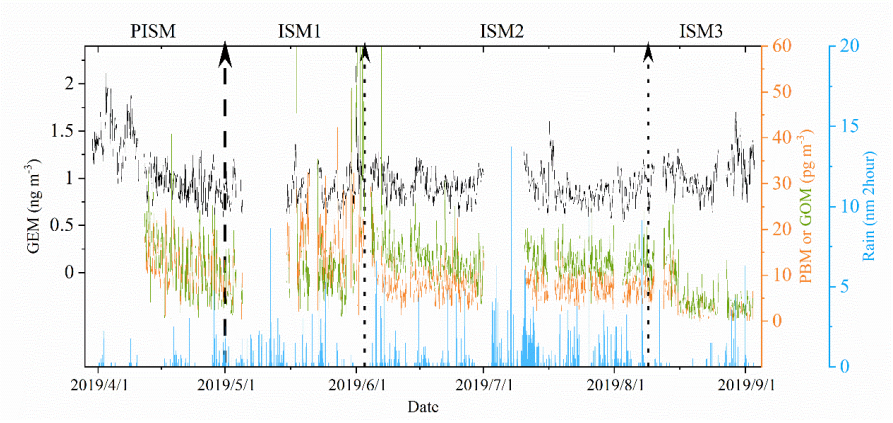
Figure 2



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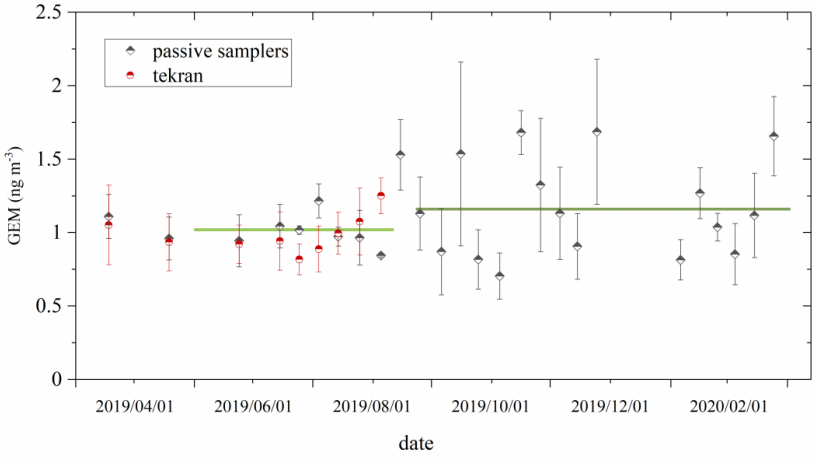
Preprint. Discussion started: 27 September 2021

Figure 3



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Preprint. Discussion started: 27 September 2021

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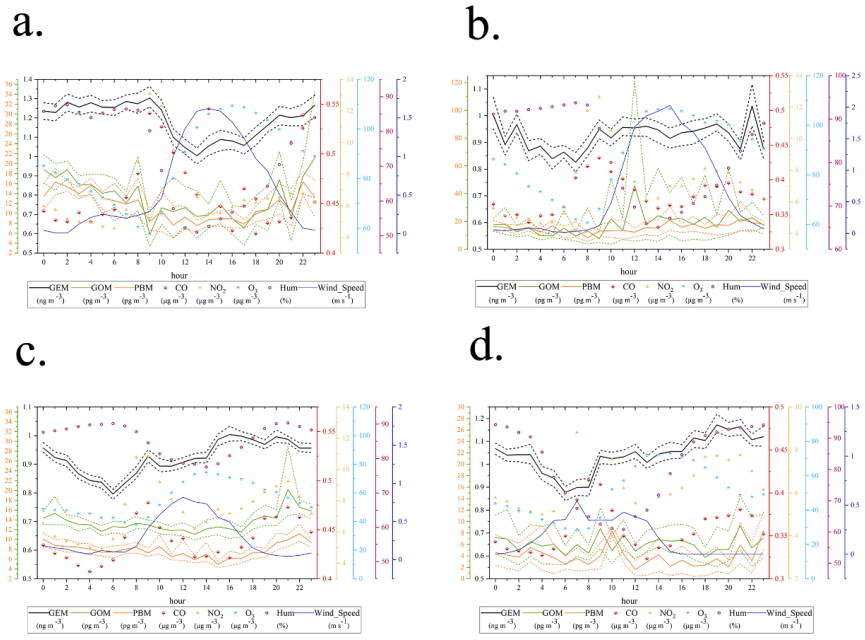
Figure 4



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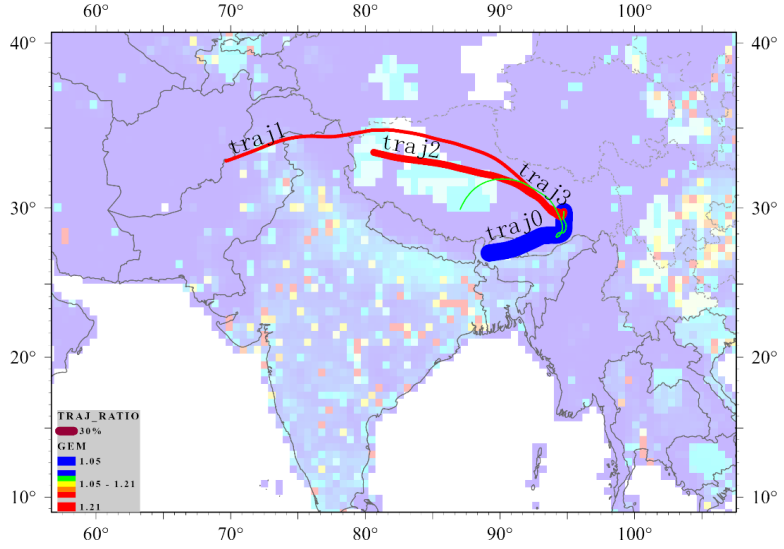
Preprint. Discussion started: 27 September 2021

Figure 5

a.



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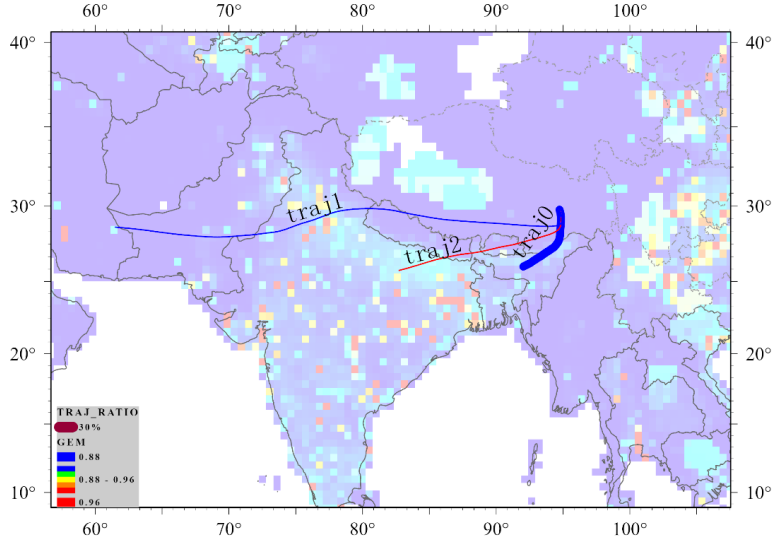

c.

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Preprint. Discussion started: 27 September 2021

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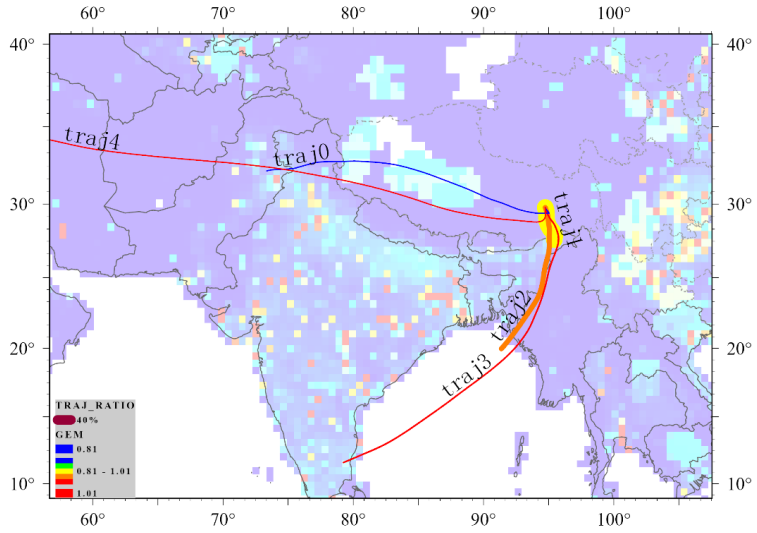
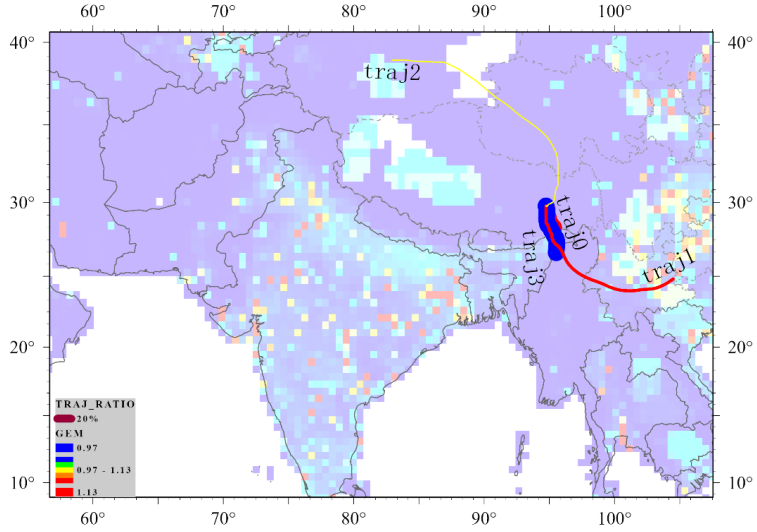


d.



https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

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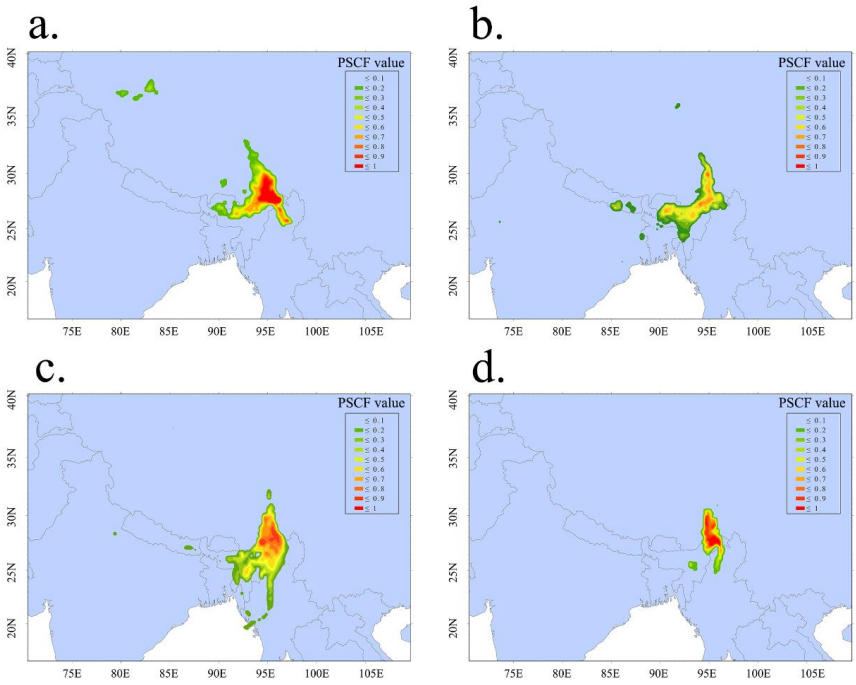
Figure 6



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Preprint. Discussion started: 27 September 2021

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Table 1. Comparison of atmospheric Hg concentrations at high elevation (> 2000m a.s.l) stations

Site	Country	Lat & Lon	Elevation	Type	Time Period	GEM or TGM	GOM	PBM	Reference
						mean ±SD, ng/m3	mean ±SD, pg/m3	mean ±SD, pg/m3	
Concordia Station	Antarctica	-79.1/123.35	3220	Remote	2013-2014	0.80 ±0.25	-	-	Sprovieri et al., 2016
SET	**China**	**29.77/94.74**	**3263**	**Remote**	**2019**	**1.01 ±0.27**	**12.8 ±13.3**	**9.3 ±5.9**	**This study**
Ev-K2	Nepal	27.96/86.81	5050	Remote	2012-2014	1.19 ±0.30	-	-	Sprovieri et al., 2016
Nam Co	China	30.78/90.99	4730	Remote	2012-2014	1.33 ±0.24			Yin et al., 2018
Qomolangma	China	28.37/86.95	4276	Remote	2016	1.42 ±0.37	21.4 ±13.4	25.6 ±19.1	Lin et al.,2016
Kodaicanal	India	10.23/77.47	2333	Rural	2013-2014	1.54 ±0.23	-	-	Sprovieri et al., 2016
Col Margherita	Italy	46.37/11.79	2545	Rural	2014	1.69 ±0.29	-	-	Sprovieri et al., 2016
Lulin	China	23.51/120.92	2862	Remote	2006-2007	1.73 ±0.61	12.1 ±20.0	2.3 ±3.9	Sheu et al., 2010
Mt. Walinguan	China	36.29/100.90	3816	Remote	2007-2008	1.98 ±0.98	7.4 ±4.8	19.4 ±18.0	Fu et al., 2012c
Mt.Ailao	China	24.53/101.02	2450	Remote	2011-2012	2.09 ±0.63	2.2 ±2.3	31.3 ±28.0	Zhang et al., 2016
Shangri-La	China	28.02/99.73	3580	Rural	2009-2010	2.55 ±0.73	8.2 ±7.9	38.8 ±31.3	Zhang et al., 2015
Mt. Leigong	China	26.39/108.20	2178	Remote	2008-2009	2.8 ±1.51	-	-	Fu et al., 2010b

https://doi.org/10.5194/acp-2021-357
Preprint. Discussion started: 27 September 2021

Table 2. PCA Factor Loadings (Varimax Rotated Factor Matrix) for Hg in Nyingchi, Tibet, China

tentative identification		GEM	PBM	GOM	Temp	Hum	Wind_ Speed	Rain	Solar _Rad	CO	NO_2	O_3	PM_{10}	$PM_{2.5}$	SO_2	VE
long-distance transport	PISM	**0.92**		0.10	**-0.79**		**0.64**	-0.15		0.28	0.43			-0.27	**-0.73**	19.86
	ISM3	**0.78**			-0.22	0.26	0.18	0.49	-0.13	**0.76**		0.29	-0.11		**-0.83**	17.05
local emission	PISM	0.13	**0.91**	**0.92**			-0.20			0.22	-0.47	-0.13	0.12		-0.44	15.96
	ISM1	0.26	**0.56**	0.19				-0.12	-0.16		-0.12		**0.69**	**0.86**	0.32	12.97
	ISM1	0.17	**0.60**	0.16	-0.40		0.19	-0.14	-0.11			**0.91**	0.22	-0.19		11.11
	ISM2	0.50	**0.89**	**0.77**	**-0.51**	**-0.52**	**0.71**	-0.12	0.10	0.14		**0.71**	**0.71**	**0.72**		30.26
	ISM3	0.25	**0.96**	**0.95**	-0.13	-0.19	-0.12	-0.34	0.27			-0.15		0.16	0.32	16.46
meteorology	ISM1		-0.18	0.11	**0.82**	**-0.77**	**0.62**	**-0.80**	**0.68**		**-0.59**		0.13		0.31	23.46
	ISM2	**-0.51**		-0.23	**0.66**	**-0.80**	0.21	**-0.79**	**0.85**		-0.10	0.25	0.43	0.18	-0.13	22.02
	ISM3	-0.19	0.17		**0.85**	**-0.89**		-0.30	**0.88**	-0.44		0.46	0.31	0.21		21.49
melt	ISM1	**0.78**	0.13	**0.85**		**-0.57**			**0.50**		0.33	0.24		0.21	0.25	15.94

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Preprint. Discussion started: 27 September 2021

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