



Measurements and Distribution of Atmospheric Particulate-Bound Mercury: A Review

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Abstract

Atmospheric particulate-bound mercury (PBM) plays an important role in the geochemical cycling of mercury (Hg). This study reviewed research progress of the PBM, including the possible emission and deposition pathways, measurement methods and the global distribution. The primary PBM sources are anthropogenic sources, but natural sources could be also a considerable contributor, for instance, chemical transport and dust in the arid and desert area. Different filter methods, such as quartz fibre filters, have been applied to the PBM measurement, and PBM can also be real-time monitored automatically. Generally, the average PBM concentrations were higher in the Northern Hemisphere than in the Southern Hemisphere. However, the PBM level of Antarctica is quite high. PBM concentrations were higher in the urban areas than in the remote areas, and there was a high PBM level in the developing countries. Moreover, high PBM concentrations were observed in the range 20°–60° of northern latitude.

Keywords Mercury · Hg · Particulate-bound mercury · Anthropogenic sources

Mercury (Hg), as a persistent, toxic, bioaccumulative heavy metal and a global pollutant, is about 1–5 ng m⁻³ in the atmosphere (Morel et al. 1998; Sprovieri et al. 2016). More than 90% of Hg exists in the form of gaseous element mercury (GEM). Less than 10% of Hg usually exists in the form of gaseous oxidized mercury (GOM) and particulate-bound mercury (PBM) (Rothenberg et al. 2010; Fu et al. 2012; Sprovieri et al. 2016). Despite the short atmospheric lifetime, PBM is still important for the distribution and transport of atmospheric Hg due to special chemical and physical

properties (Murphy et al. 1998; Kim et al. 2012). It is found that PBM is mainly associated with aerosols in the atmosphere, and resulted from various physical and/or photochemical processes in the atmosphere (Ariya et al. 2015). Hg is emitted into the atmosphere from various natural and anthropogenic sources, either as GEM or as GOM and PBM. PBM concentrations depend on the direct emission from local and regional sources and the situ formation in the atmosphere (Pirrone et al. 2010; Ariya et al. 2015).

PBM may be deposited at medium or long distances, the deposition depend largely on the size of particles or aerosols, and the main meteorological conditions (atmospheric stability, relative humidity and wind speed) (Xiu et al. 2009; Zhu et al. 2014). To the total PBM deposition, however, fine and coarse PBM together only accounted for a small fraction of the total Hg dry deposition (Wang et al. 2006; Fang et al. 2010, 2012b). But the relative contributions from Hg species were mainly affected by their partitioning in air concentrations, emissions from surface, dominant land covers, and meteorological conditions (Lindberg and Stratton 1998; Fang et al. 2012a).

So far, many previous works have developed analytical methods for detecting PBM (Lu and Schroeder 1999; Munthe et al. 2001). However, in the atmosphere, PBM concentrations are extremely low, which are at the

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picogram level. Thus PBM measurement is quite difficult and demands sufficient sample preconcentration and high sensitivity of instrument. However, with the continuous development of chemical/analytical techniques, some sensitive instruments have made it possible to reliably determine PBM using less than 1 ng of samples, allowing sampling in a shorter time and/or at a lower flow rate. For example, the Tekran Model 1135 Particulate Mercury Unit provides a direct indication of PBM amount in the atmosphere (Sprovieri et al. 2016).

Possible Emission and Deposition Pathways of PBM

PBM may originate from direct human activities and some types of natural sources or processes (Pirrone et al. 2010). Anthropogenic emission is very important, especially, the coal-fired utilities and other combustion sources, including the fossil fuels, biomass burning, residential coal combustion, municipal waste, ferrous and nonferrous metals smelting processes, caustic soda production, fluorescent lamp production and miscellaneous industries to PBM emissions (Carpi 1997; Li et al. 2007; Obrist et al. 2008; Wang et al. 2015; Zhang et al. 2015b). Besides, natural processes directly emit PBM to the air or indirectly result in the formation of PBM with atmospheric chemical/physical transport, including outgassing of the earth's mantle crustal or material, evasion from surficial soils, water bodies, vegetation surfaces, wild fires, volcanoes, and geothermal sources (Pirrone et al. 2010; Wang et al. 2016). In addition, re-emission of previously deposited Hg is also important, but only a limited number of published measurements estimated Hg re-emission fluxes (Pirrone et al. 2010; Ariya et al. 2015).

The total amount of PBM deposited is difficult to quantify. Total wet deposition of Hg is widely measured using rain and snow samplers although individual deposition of GOM and PBM cannot be measured in this way (Fu et al. 2016). However, PBM is more important for atmospheric Hg deposition due to its higher dry deposition velocities and scavenging coefficients than GOM (Sakata and Asakura 2007; Zhang et al. 2016). However, there is no widely accepted measurement technology for dry deposition of PBM, and dry deposition measurements of PBM often have large uncertainties because of extremely low PBM concentration in the air, the spatial heterogeneity of ground surface, the small vertical gradient of PBM concentration, the influence of rapid chemical reaction and local source (Ariya et al. 2015). Therefore, modeling work is usually used to estimate dry deposition of Hg using a resistance approach (Zhang et al. 2016).

Methods for PBM Measurements

In the last three decades, some manual and automated methods have been developed to measure PBM (Xiao et al. 1991; Munthe et al. 2001; Sprovieri et al. 2016). A variety of different filter methods have been applied to collect PBM samples, such as Teflon or quartz fibre filters, cellulose-acetate filters (Lu and Schroeder 1999; Qie et al. 2018). In some early measurements, wet chemical method usually was used to digest the filter samples, which were pre-treated using a solution of nitric acid and hydrogen peroxide in a Teflon vessel, then Hg was determined by cold vapour atomic absorbance spectrometry (CVAAS) or cold vapour atomic fluorescence spectrometry (CVAFS) after pre-concentration on gold trap followed by EPA (U.S. Environmental Protection Agency) 1631 method (Munthe et al. 2001). However, the wet chemical method is more complex. In the later development, a denuder-based system was used for sampling PBM and GOM. PBM (defined as Hg associated with particles $< 2.5 \mu\text{m}$) was collected on 47 mm diameter quartz fibre filter housed in a Teflon filter holder downstream of the denuder. Each quartz fibre filter was used for one sampling and analysis cycle to prevent from possible contamination. Both PBM and GOM samples were collected every 2 h with a sampling flow rate of 10 L min^{-1} . The Hg collected on the filter can be released thermally at 900°C , followed by gold trap amalgamation and CVAFS detection (Zhang et al. 2015a).

Base on the manual denuder-based system, an automated measurement analyzer of PBM, Tekran 2537-1130-1135 mercury speciation system (Fig. S1), was developed, and make it possible to continuously determine both urban and background concentrations of PBM. Fine fraction ($< 2.5 \mu\text{m}$) PBM samples were collected on 21 mm diameter quartz fibre filters in the Tekran 1135 unit, with a flow rate of 10 L min^{-1} during a sampling period of 1 h. Following the collection period, PBM was thermally decomposed from the 1135 Tekran unit and measured by the Tekran 2537A/B/X analyzer within an 1 h desorption cycle. Operation, maintenance, calibration and data quality assurance (QA) procedure are followed by the Global Mercury Observation System (GMOS) Standard Operation Procedure (SOP) and Data Quality Management (Sprovieri et al. 2016).

Global and Regional Distribution of PBM

A summary of PBM dataset based on previous studies is presented in Table S1. The data covered most of all the data measured by filters in the previous studies. Because

we just evaluate the total distribution of PBM, the size distributions of PBM is not discussed in this study. To study the distribution and potential sources of PBM, PBM concentrations have been measured in some developed areas such as Northern America and Europe Union (EU) from 1990s of last century. However, a small number of monitoring sites have been setup to measure PBM in Asia, nearly all the PBM data came from China, Japan and South Korea. There are several reports of PBM in Turkey, Israel, India and Nepal. There are no PBM monitoring data in the Southern Hemisphere except for Antarctica. There are more PBM monitoring points in remote areas than in urban areas. Most of PBM data were from the developed countries, the PBM data is quite lacking and there is a blank in the developing countries except for China.

Globally, the mean PBM concentration is 110.6 ± 98.9 pg m⁻³ in this study (Table S1), and PBM concentrations in the Northern Hemisphere (113.8 ± 102.1 pg m⁻³) showed a much higher level than in the Southern Hemisphere (53.5 ± 47.4 pg m⁻³). In general, PBM concentrations are closely related to its original areas and transport pathways. Because most of industry activities are in the Northern Hemisphere. Anthropogenic PBM emissions in the Northern Hemisphere are higher than that in the Southern Hemisphere. China and India are the most important mercury emitters in the Northern Hemisphere, and the rapid development of industrial activities has released large amounts of mercury into the air (Pirrone et al. 2010; Fu et al. 2015). Biomass combustion in South and Southeast Asia is also an important source of atmospheric Hg (Wang et al. 2015). In addition, developed countries and areas such as EU and the United States, Japan and South Korea also emitted large amounts of mercury into the environment during previous industrial developments. The Hg deposited in the environment is released into the atmosphere again and converted into PBM in the atmosphere, which may also be a source of PBM.

However, the PBM level of Antarctica is higher than that in the Arctic (Table S1, Fig. 1), the highest mean concentration of PBM was 166 ± 147 pg m⁻³ measured in the Antarctica pole, which is higher than the global mean PBM concentration (Arimoto et al. 2004). The Arctic, unlike Antarctica, is surrounded by industrialized continents that contribute gaseous and particulate pollution (arctic haze). Therefore Arctic pollution is caused by both natural and anthropogenic pollutants from the Eurasian continent and North America, particularly in late winter and spring (Pirrone et al. 2010; Zhang et al. 2015b). However, in the Antarctica, the reason of high PBM might be resulted from the in-situ physicochemical productions, such as the result of gas phase oxidation of GEM and scavenging by aerosol particles (Lin and Pehkonen 1999; Sprovieri et al. 2002; Pfaffhuber et al. 2012; Ariya et al. 2015).

The PBM level of developing countries was higher than that in the developed countries. Generally, the PBM level in the United States of America (USA), Canada, EU and Japan, was lower than that in China, India and Mexico (Chand et al. 2008) (Table S1, Fig. 2). Due to coal combustion, waste incineration, metal smelting, refining and manufacturing as well as heavy burning of coal in residential and small industrial settings without particulate matter (PM) controls, PBM emissions are high in the developing countries (Wang et al. 2010; Zikang Cui et al. 2019). In addition, the large amount of biomass combustion in some developing countries and regions is also the reason for the high PBM in the atmosphere (De Simone et al. 2017). Some studies have shown that biomass combustion from South Asia, Southeast Asia and South America contributes significantly to local high PBM concentrations (Wang et al. 2015).

The PBM level in urban areas, particularly in China and South Asia, was higher than that in the remote areas (Table S1, Figs. 2, 3). Mean PBM concentrations at urban sites ranged from 2.3 to 1180 ng m⁻³ (mean 225 ± 174.5 ng m⁻³), significantly higher than the PBM concentrations measured at remote sites (mean 25.3 ± 29.7 ng m⁻³). In the urban areas, PBM emissions to the atmosphere were from anthropogenic sources including fossil-fuel power plants and industrial facilities, particularly from coal, oil and biomass combustion as well as from solid waste incineration (Pirrone et al. 2010). Although PBM residence time in atmospheric environment is relatively short, the release of PBM can rapidly increase PBM concentrations near the emission source, resulting in the high PBM level in urban areas. However, a small amount of PBM from anthropogenic sources can be transported to remote background areas through long-distance transportation. Additionally, natural emissions, such as volcanoes, forest fires and physicochemical transformation in the atmosphere, are also the sources of PBM (Wang et al. 2016). This could be the main factors affecting PBM distribution in remote background areas.

In this study, we found that the spatial distribution of PBM is linked to latitude, the higher PBM concentrations were in the range 20°–60° of northern latitude (Fig. 4). Within this latitude range, there are the major population and industries, such as China, USA, EU and India, large amounts of PBM from anthropogenic sources was emitted into the atmosphere, which resulted in a high PBM concentrations in the regions (Pirrone et al. 2010; Fu et al. 2015). Another reason is that most of the world's land is in the Northern Hemisphere, and most of the mercury emissions from land-based natural sources are concentrated in this dimension, such as forest fires and other biomass burning. Previous studies have found that biomass combustion from Southeast Asia and South Asia can increase PBM concentrations in the region. In addition, some studies have found

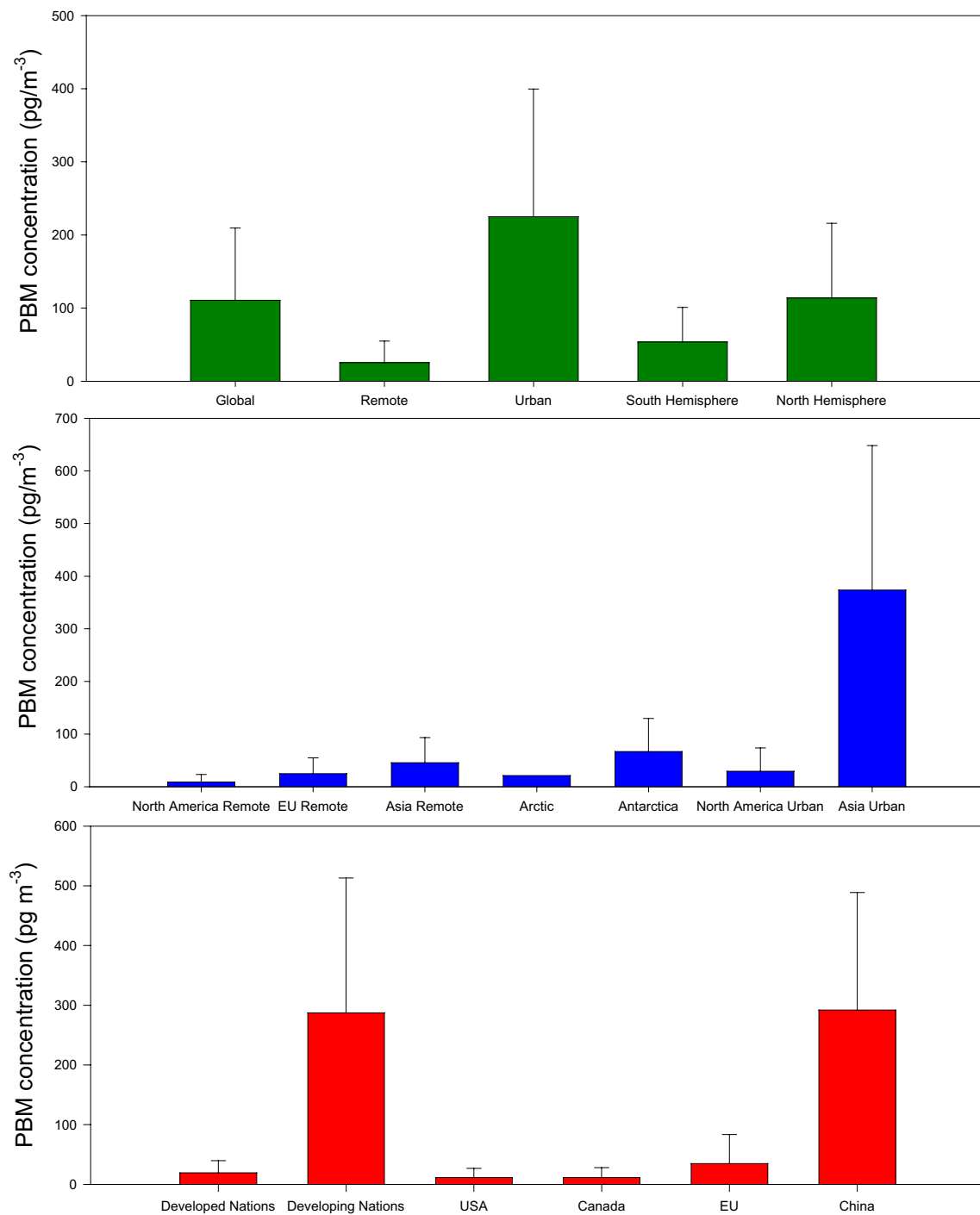


Fig. 1 The mean PBM concentration in the world and different areas

that dust and haze weather is also an important reason for the increasing of PBM in the atmosphere in these areas (Wang et al. 2015; Hong et al. 2016).

PBM has multiple sources and undergoes complex transport and transformation processes in the atmosphere (Ariya et al. 2015). Further systematic study is thus needed to better quantify the PBM from different sources and get a more

accurate PBM emission inventory, which is beneficial for controlling atmospheric Hg emissions. Additionally, isotope analysis technology is very important for the study of atmospheric PBM. Recent studies have focused on the isotope composition and fractionation of PBM in atmosphere (Fu et al. 2019; Huang et al. 2019). According to these studies, it is better to study the chemical changes and cycles

Fig. 2 PBM concentrations in the remote areas. The data of the PBM emission (mg km^{-2}) were from AMAP/UNEP geospatially distributed mercury emissions dataset 2010v1 (AMAP/UNEP 2013)

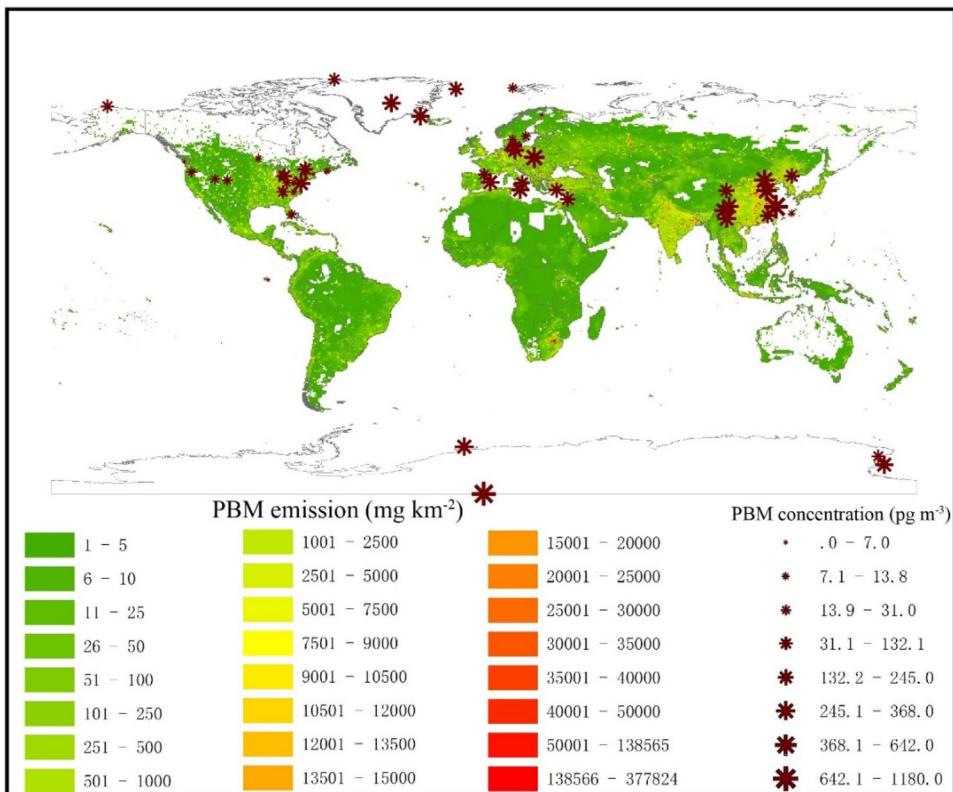


Fig. 3 PBM concentrations in the urban areas. Data of the PBM emission (mg km^{-2}) were from AMAP/UNEP geospatially distributed mercury emissions dataset 2010v1 (AMAP/UNEP 2013)

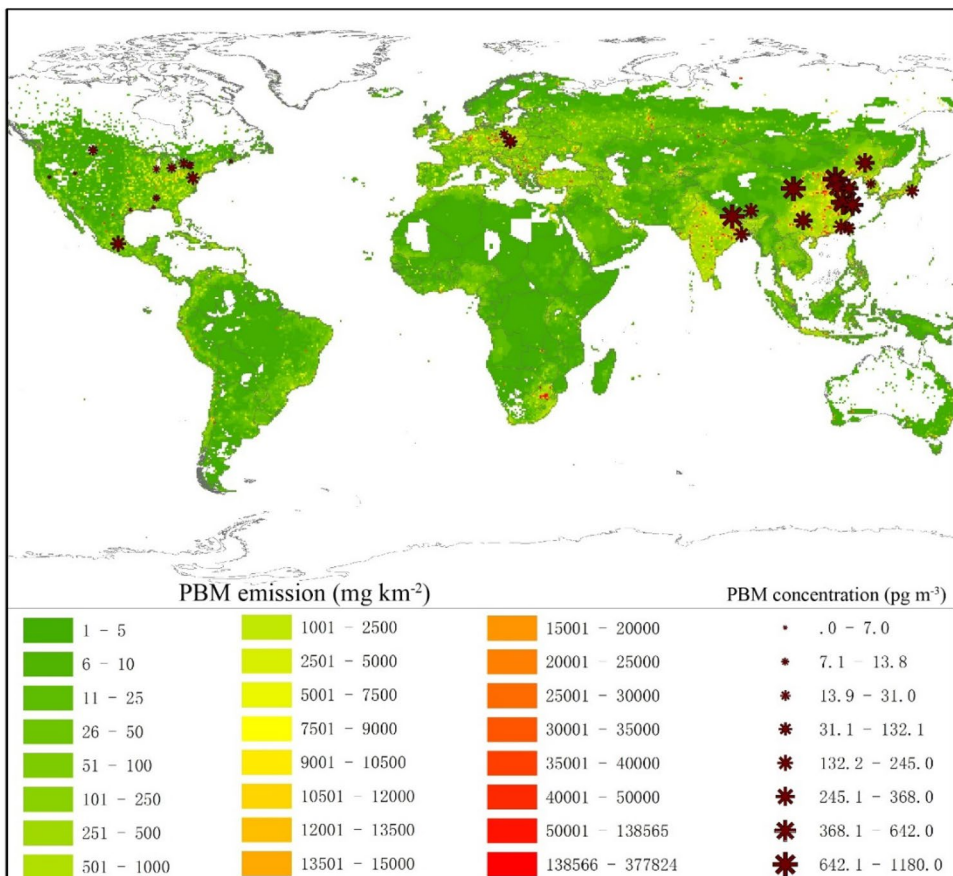
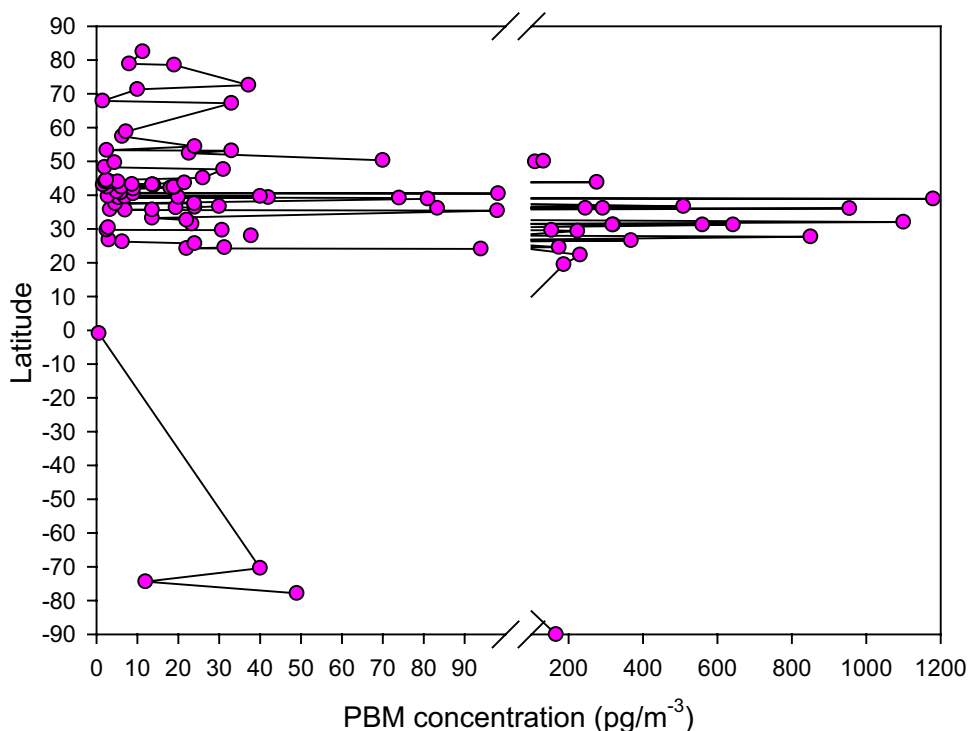


Fig. 4 PBM distribution with the latitude

of atmospheric Hg, and recognize and explain the different potential source characteristics of atmospheric Hg, so as to promote to understand the global biogeochemical cycling of Hg.

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