



Mercury Deposition, Climate Change and Anthropogenic Activities: A Review

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As a toxic and harmful global pollutant, mercury enters the environment through natural sources, and human activities. Based on large numbers of previous studies, this paper summarized the characteristics of mercury deposition and the impacts of climate change and human activities on mercury deposition from a global perspective. The results indicated that global mercury deposition changed synchronously, with more accumulation during the glacial period and less accumulation during the interglacial period. Mercury deposition fluctuated greatly during the Early Holocene but was stable and low during the Mid-Holocene. During the Late Holocene, mercury deposition reached the highest value. An increase in precipitation promotes a rise in forest litterfall Hg deposition. Nevertheless, there is a paucity of research on the mechanisms of mercury deposition affected by long-term humidity changes. Mercury accumulation was relatively low before the Industrial Revolution ca. 1840, while after industrialization, intensive industrial activities produced large amounts of anthropogenic mercury emissions and the accumulation increased rapidly. Since the 1970s, the center of global mercury production has gradually shifted from Europe and North America to Asia. On the scale of hundreds of thousands of years, mercury accumulation was greater in cold periods and less in warm periods, reflecting exogenous dust inputs. On millennial timescales, the correspondence between mercury deposition and temperature is less significant, as the former is more closely related to volcanic eruption and human activities. However, there remains significant uncertainties such as non-uniform distribution of research sites, lack of mercury deposition reconstruction with a long timescale and sub-century resolution, and the unclear relationship between precipitation change and mercury accumulation.

Keywords: mercury deposition, site distribution, climate change, anthropogenic activities, industrialization

INTRODUCTION

As a toxic and harmful global pollutant, mercury enters the environment through natural sources (volcanic eruptions, oceans, soil, and forests, etc.), and human activities (fossil fuel combustion; gold, silver, and mercury mining; non-ferrous metal smelting; etc.; Tang et al., 2012; Guédron et al., 2018; Obrist et al., 2018; Pratte et al., 2018). Mercury in the atmosphere includes particulate mercury and gaseous mercury. Particulate mercury includes Hg(0) and HgCl₂, among others, which are adsorbed on the surface of particles or combined with particles; gaseous mercury mainly

exists in the form of Hg(O) (Wang, 2011). Due to its high volatility, low chemical activity, and low solubility, mercury is characterized by a long atmospheric life (0.5–2 years), and long-distance transport (Roos-Barraclough et al., 2002). Furthermore, there are a few other volatile mercury compounds, including HgBr₂, HgCl₂, and Hg(OH)₂, that are easily soluble in water and reduced to Hg(0), referred to as reactive gaseous mercury (RGM; Wang, 2011). Mercury under long-distance transportation can enter natural archives, including peatlands, lacustrine deposits, ice cores, coastal salt marsh sediments, and marginal pelagic sediments, through dry and wet deposition (Schuster et al., 2002; Tang et al., 2012; Donovan et al., 2013; Krabbenhoft and Sunderland, 2013; Guédron et al., 2018; Kim et al., 2019). These characteristics of mercury accumulation in natural archives can, to some extent, reflect the process of atmospheric mercury deposition during historical periods, and the deposition rate may be subject to factors such as climate change, geological conditions, volcanic eruptions, and human activities in the natural environment (Biester et al., 2007; Guédron et al., 2019).

In recent years, there have been many studies reconstructing atmospheric mercury deposition during historical periods using the natural archives. These studies focused mainly on the Northern Hemisphere, whereas studies in the Southern Hemisphere were limited to Chile (Biester et al., 2002, 2003; Hermanns and Biester, 2013a,b; Daga et al., 2016; Guédron et al., 2019), Brazil (Peirez-Rodríguez et al., 2015; De Lacerda et al., 2017), and Peru of South America (Cooke et al., 2009; Beal et al., 2014). In the Northern Hemisphere, European countries, represented by Switzerland (Roos-Barraclough et al., 2002; Roos-Barraclough and Shotyck, 2003; Zacccone et al., 2008; Thevenon et al., 2011), Spain (Martínez-Cortizas et al., 1999, 2012; Gallego et al., 2013; Serrano et al., 2013; Corella et al., 2017), Denmark (Shotyck et al., 2003), Greenland (Shotyck et al., 2003; Zheng, 2015; Pérez-Rodríguez et al., 2017), Scotland (Farmer et al., 2009; Küttner et al., 2014), Norway (Steinnes and Sjøbakk, 2005; Drevnick et al., 2012), and Sweden (Bindler et al., 2004), were studied mostly in terms of mercury deposition, followed by North America, represented by Canada (Givelet et al., 2004; Sunderland et al., 2008; Zdanowicz et al., 2015, 2016; Wiklund et al., 2017; Korosi et al., 2018; Štok et al., 2019), and the United States (Benoit et al., 1998; Schuster et al., 2002; Arnason and Fletcher, 2003; Conaway et al., 2004; Gray et al., 2005; Roos-Barraclough et al., 2006; Donovan et al., 2013; Megaritis et al., 2014; Kurz et al., 2019). In contrast, there have been fewer studies on mercury deposition in China, with the research areas mainly distributed in the Shennongjia Dajiu Lake Basin (Li et al., 2016, 2017), the Greater Khingan Range (Bao et al., 2016), the Lesser Khingan Range (Xu et al., 2010; Tang et al., 2012), the Tibetan Plateau (Wang et al., 2010; Yang et al., 2010; Huang et al., 2013), and the South China Sea (Xu et al., 2010; Liu et al., 2012). In turn, there have numerous time series analyses that document changes in mercury deposition on 10³ to 10¹ timescales that evaluate the impact of climate change and human activities on mercury deposition. However, these studies were almost always limited to a certain place or a certain area and explained only the mercury accumulation in a certain area. We are unaware of previous reviews of mercury deposition on a longer timescale

from a global perspective. The authors hoped to find similarities and differences between different regions by analyzing and summarizing historical mercury deposition data at a global scale, and explored the relationship between mercury deposition and climate change and human activities.

This paper identified and analyzed relevant studies on atmospheric mercury deposition regarding climate change and human activities from a global perspective, discussing the prospects for future research. The global mercury deposition history reflected the global climate change and regional human activities during historical periods. Our work is conducive to the systematic understanding of mercury deposition and its influencing factors. On this basis, new methods and perspectives can be developed to conduct more in depth research.

DATA SOURCES

The data used in this study were mainly based on peer-reviewed papers and the data for Dajiu Lake Basin from our research group. The authors searched for documents in Elsevier SDOL, Nature, Science, Google Scholar, and the CNKI database, among others, using keywords such as “mercury deposition/mercury accumulation,” “climate change,” and “anthropogenic activities/human activities.” Following the principles of a uniform spatial distribution and an extensive timescale, 36 sets of mercury deposition data from 60 papers were selected, covering the history of atmospheric mercury deposition on six continents (Asia, Europe, Africa, North America, South America, and Antarctica) recorded by natural archives (fen peat, lacustrine deposits, ice cores, and marine deposits, etc.) from 670 ka BP to now, as shown in **Table 1**.

According to the global studies on atmospheric mercury deposition, the authors found that the site distribution of global mercury deposition studies was characterized by an overall pattern of “more in the north and fewer in the south”; that is, the research areas were mostly distributed in the Northern Hemisphere, such as Europe (including Belgium, the Czech Republic, Denmark, Greenland, Ireland, Norway, Scotland, Spain, Sweden, and Switzerland), North America (including the United States and Canada), and China (including the Northeast Hani, Shennongjia Dajiu Lake Basin, Greater Khingan Range, Lesser Khingan Range, Sichuan Hongyuan, Huguangyan Maar Lake, Chao Lake, Tibetan Plateau, Xinjiang Mount Tianshan, East China Sea, Yellow Sea, and South China Sea), while in the Southern Hemisphere, the areas were distributed only in South America (including Chile, Peru, Brazil, and Bolivia), Africa (including the Berg River of South Africa and Lake Tanganyika in East Africa), and some parts of Antarctica, as shown in **Figure 1**.

RESULTS AND DISCUSSION

Regional Comparison of Global Mercury Deposition

At the global scale, atmospheric mercury deposition is closely related to climate variability (such as temperature and humidity changes) and human activities (such as mining, metal smelting,

TABLE 1 | Information of global mercury deposition research spots.

| Number | Study site | Region | Location | Altitude/m | Materials dated | Depth/m | Dating methods | Age range | Selected references |
|--------|---------------------------|--------------------------------------|-----------------------------|------------|---|---------|---|------------------|-----------------------------------|
| a1 | The Upper Fremont Glacier | Wyoming, United States | 44°20'02" N 111°36'49" W | 4100 | Ice core | 160 | Isotope and chemical dating | 1720–1993 AD | Schuster et al., 2002 |
| a2 | San Francisco Bay | United States | 37°40'00" N 122°25'00" W | / | Coastal salt marsh | 1.6 | ¹³⁷ Cs, ²¹⁰ Pb | 1850–2000 AD | Donovan et al., 2013 |
| a3 | Southern Baffin Island | Arctic Canada | 63°57'53" N 68°15'42" W | 1825 | Firn | 22.7 | δ ¹⁸ O | 1970–2010 AD | Zdanowicz et al., 2015 |
| a4 | The Bay of Fundy | Canada | 45°12'36" N 66°54'12" W | / | Peat bogs, Lake sediments, Coastal salt marsh | / | ¹³⁷ Cs, ²¹⁰ Pb | 1800–2000 AD | Sunderland et al., 2008 |
| a5 | Lake Ontario | Canada | 49°48'00" N 93°48'00" W | / | Lake sediments | / | ¹³⁷ Cs, ²¹⁰ Pb | 1850–2000 AD | Wiklund et al., 2017 |
| a6 | Ellesmere Island | The Canadian High Arctic Archipelago | 78°43'00" N 74°27'00" W | 323 | Lake sediments | 12.1 | ¹³⁷ Cs, ²¹⁰ Pb | 1850–2000AD | Korosi et al., 2018 |
| a7 | Caribou Bog | Central Maine, United States | 44°58'97" N 68°48'35" W | 80 | Peat bogs | 5 | ²¹⁰ Pb, ¹⁴ C | 8000 BC–2000 AD | Roos-Barraclough et al., 2006 |
| a8 | Lost Lake | Wyoming, United States | 43°46'55" N 110°06'00" W | 2889 | Lake sediments | 21 | ¹³⁷ Cs, ²¹⁰ Pb | 1350–2000 AD | Kurz et al., 2019 |
| a9 | Oyster Point | California, United States | 45°25'28" N 111°20'28" W | / | Coastal salt marsh | 3 | ¹⁴ C | 1700–2000 AD | Conaway et al., 2004 |
| a10 | Arlberg Bog | Minnesota, United States | 46°56'00" N 92°41'00" W | / | Peat bogs | 0.6 | ¹³⁷ Cs, ²¹⁰ Pb | 1700–2000 AD | Benoit et al., 1998 |
| a11 | Narraguinsep Reservoir | Colorado, United States | 40°31'26" N 106°58'43" W | 2035 | Lake sediments | 10 | ¹³⁷ Cs | 1950–2000 AD | Gray et al., 2005 |
| a12 | Patroon Reservoir | New York, United States | 42°41'05" N 73°47'21" W | / | Lake sediments | 3 | / | 1955–2000 AD | Arnason and Fletcher, 2003 |
| b1 | Etang de la Gruère | Swiss Jura Mountains | 47°15'34" N 07°03'51" E | 1005 | Peat bogs | 6.5 | ²¹⁰ Pb, ²⁴¹ Am, ¹⁴ C | 12500 BC–2000 AD | Roos-Barraclough et al., 2002 |
| b2 | Franches Montagnes | Swiss Jura Mountains | 47°14'23" N 07°02'57" E | 1020 | Peat bogs | 0.8 | ²¹⁰ Pb, ¹⁴ C | 1200 BC–2000 AD | Roos-Barraclough and Shotyk, 2003 |
| b3 | Lake Lucerne | Central Switzerland | 46°13'25" N 07°40'28" E | 2661 | Lake sediments | 1.2 | ¹⁴ C | 14315 BC–2000 AD | Thevenon et al., 2011 |
| b4 | Roñanzas Bog | Asturias, Spain | 43°20'13" N 04°51'01" W | / | Peat bogs | 2 | / | 6000 BC–2000 AD | Gallego et al., 2013 |
| b5 | Galicia | Northwest Spain | 43°32'00" N 07°34'00" W | / | Peat bogs | 2.5 | ¹⁴ C | 2635 BC–1995 AD | Martínez-Cortizas et al., 1999 |
| b6 | Lake Montcortès | Pyrenees, Spain | 42°19'00" N 00°19'00" E | 1031 | Lake sediments | 1.1 | ²¹⁰ Pb, ¹⁴ C | 1386–2010 AD | Corella et al., 2017 |
| b7 | Chao de Lamoso bog | Xistral Mountains, northwest Spain | 43°14'35" N 09°05'45" W | 1039 | Peat bogs | 1 | ²¹⁰ Pb | 1825–2000 AD | Martínez-Cortizas et al., 2012 |
| b8 | Portlligat Bay | Iberian Peninsula | 42°17'32" N 03°17'28" E | / | Coastal salt marsh | 5 | ¹⁴ C | 2315 BC–2000 AD | Serrano et al., 2013 |
| b9 | Storelung Mose | Denmark | 55°15'23" N 10°15'22" E | / | Peat bogs | 1 | ¹⁴ C | 2000 BC–2000 AD | Shotyk et al., 2003 |

(Continued)

TABLE 1 | Continued

| Number | Study site | Region | Location | Altitude/m | Materials dated | Depth/m | Dating methods | Age range | Selected references |
|--------|-------------------------|--|--------------------------------------|------------|----------------------------|---------|---|------------------|------------------------------|
| b10 | Sandhavn | Southern Greenland | 59°59'54" N 44°46'36" W | / | Peat bogs | 0.4 | ²¹⁰ Pb, ¹⁴ C | 1270–2000 AD | Pérez-Rodríguez et al., 2017 |
| b11 | Raeburn Flow | Scotland, United Kingdom | 55°02'04" N 03°06'27" W | / | Peat bogs | 3.6 | ²¹⁰ Pb, ¹⁴ C | 1385 BC–2005 AD | Küttner et al., 2014 |
| b12 | Six ombrotrophic bogs | Norway | 58°–69° N 04°–12° E | / | Peat bogs | 1 | ²¹⁰ Pb, ¹⁴ C | 2000 BC–2000 AD | Steinnes and Sjøbakk, 2005 |
| b13 | Svalbard | Norwegian Arctic | 80°03'05" N 17°37'26" E | / | Lake sediments | / | ²¹⁰ Pb | 1800–1995 AD | Drevnick et al., 2012 |
| b14 | Store Mosse | South-central Sweden | 57°15'00" N 13°55'00" E | / | Peat bogs | 0.4 | ¹³⁷ Cs, ²¹⁰ Pb | 1860–2000 AD | Bindler et al., 2004 |
| b15 | Marano and Grado Lagoon | Northern Adriatic Sea | 40°12'–45°53' N 12°13'–19°34' E | / | Coastal salt marsh | 1.1 | ¹³⁷ Cs, ²¹⁰ Pb | 1650–2000 AD | Covelli et al., 2012 |
| b16 | Misten peat bog | Eastern Belgium | 50°38'28" N 03°11'17" E | / | Peat bogs | 1 | ²¹⁰ Pb, ¹⁴ C | 431–2011 AD | Allan et al., 2013 |
| b17 | Brdy Hills | The Czech Republic | 49°42'42" N 13°52'30" E | / | Peat bogs | 0.38 | ²¹⁰ Pb | 1807–2003 AD | Ettler et al., 2008 |
| b18 | Multiple lakes | Across England | 50°–55° N 04° W–02° E | 3–244 | Lake sediments | 1.5 | ¹³⁷ Cs, ²¹⁰ Pb, ²⁴¹ Am | 1850–2010 AD | Yang H. D. et al., 2016 |
| c1 | Tanghongling | Heilongjiang Province, northeast China | 46°42'–48°40' N 129°05'–129°55' E | / | Peat bogs | 1.05 | ²¹⁰ Pb, ¹⁴ C | 4480 BC–2000 AD | Tang et al., 2012 |
| c2 | Daju Lake Basin | Hubei Province, China | 31°29'27" N 109°59'45" E | 1760 | Peat bogs | 2.97 | ¹⁴ C | 14141 BC–2004 AD | Li et al., 2017 |
| c3 | Motianling mountain | Western Great Hinggan Mountains, China | 46°39'–47°39' N 119°28'–121°23' E | 1200 | Peat bogs | 0.78 | ¹³⁷ Cs, ²¹⁰ Pb | 1820–2005 AD | Bao et al., 2016 |
| c4 | Tibetan Plateau | Southwest China | 28°41'–37°17' N 85°23'–100°16' E | 2813–4652 | Lake sediments | 0.4 | ¹³⁷ Cs, ²¹⁰ Pb, ²⁴¹ Am, ²²⁶ Ra | 1830–2010 AD | Yang et al., 2010 |
| c5 | Qinghai Lake | Northeast Tibetan Plateau, China | 36°24'00" N 100°09'00" E | / | Lake sediments | 0.205 | ¹³⁷ Cs, ²¹⁰ Pb, ²⁴¹ Am, ²²⁶ Ra | 1860–2000 AD | Wang et al., 2010 |
| c6 | Guangjin Islan | South China Sea | 16°27'07" N 111°42'05" E | / | Eggshells | 1.05 | ²¹⁰ Pb, ¹⁴ C | 1280–2000 AD | Xu et al., 2010 |
| c7 | Xisha Islands | South China Sea | 15°47'–17°08' N 110°10'–112°55' E | / | Coral sand | 0.95 | ²¹⁰ Pb, ¹⁴ C | 1300–2000 AD | Liu et al., 2012 |
| c8 | Lake Sayram | Xinjiang Province, China | 44°30'–44°42' N 81°05'–81°15' E | 2072 | Lake sediments | 0.3 | ²¹⁰ Pb | 1810–2010 AD | Zeng et al., 2014 |
| c9 | Hani peat bog | Jilin Province, China | 42°12'50" N 126°31'05" E | 882–900 | Peat bogs | 9 | ¹⁴ C | 11927 BC–2010 AD | Xiao, 2017 |
| c10 | Okinawa Trough | East China Sea | 27°29'50" N 126°41'16" E | / | Marginal pelagic sediments | 4.95 | ¹⁴ C | 17990 BC–2010 AD | Lim et al., 2017 |

(Continued)

TABLE 1 | Continued

| Number | Study site | Region | Location | Altitude/m | Materials dated | Depth/m | Dating methods | Age range | Selected references |
|--------|----------------------|-----------------------------|--------------------------------------|------------|----------------------------|---------|---|-------------------|-------------------------------|
| c11 | East China Sea | China | 26°–32° N 120°–126° E | / | Marginal pelagic sediments | 1.4 | ¹³⁷ Cs, ²¹⁰ Pb | 1913–2015 AD | Zhang R. et al., 2018 |
| c12 | Shanghai | China | 31°26'17" N 121°23'02" E | / | Lake sediments | 0.35 | ¹³⁷ Cs, ²¹⁰ Pb | 1750–2010 AD | Yang J. et al., 2016 |
| c13 | Chao Lake | Anhui Province, China | 31°25'–31°43' N 117°16'–117°51' E | / | Lake sediments | 0.3 | ²¹⁰ Pb | 1890–2009 AD | Zhang H. X. et al., 2018 |
| c14 | Hongyuan | Sichuan Province, China | 32°46'46" N 102°30'58" E | 3510 | Peat bogs | 0.25 | ²¹⁰ Pb | 1850–2006 AD | Shi et al., 2011 |
| c15 | Huguangyan Maar Lake | Guangdong Province, China | 21°09'00" N 110°17'00" E | / | Lake sediments | 0.94 | ¹³⁷ Cs, ¹⁴ C | 766–2005 AD | Han, 2018 |
| c16 | Huguangyan Maar Lake | Guangdong Province, China | 21°09'00" N 110°17'00" E | / | Lake sediments | 1.175 | ¹³⁷ Cs, ²¹⁰ Pb, ¹⁴ C | 1350–2004 AD | Zeng et al., 2017 |
| d1 | Lake Titicaca region | Bolivia | 15°50'–16°13' S 68°03'–68°17' W | 3760–4040 | Peat bogs | 1.63 | ¹⁴ C | 11430 BC–2014 AD | Guédron et al., 2018 |
| d2 | Lake Chungará | Chile | 18°15'07" S 69°09'47" W | 4520 | Lake sediments | 1.46 | ¹³⁷ Cs, ²¹⁰ Pb | 686 BC–2014 AD | Guédron et al., 2019 |
| d3 | Lake Hambre | Patagonia, Chile | 53°36'13" S 70°57'08" W | 80 | Lake sediments | 13.94 | ¹⁴ C | 14722 BC–2008 AD | Hermanns and Biester, 2013a |
| d4 | The Gran Campo bog | Magellanic Moorlands, Chile | 52°47'26" S 72°56'37" W | / | Peat bogs | 1.5 | ²¹⁰ Pb, ¹⁴ C | 466 BC–1995 AD | Biester et al., 2002 |
| d5 | Lake Futalaufquen | Patagonia, Chile | 42°49'00" S 71°43'00" W | 518 | Lake sediments | 0.79 | ¹³⁷ Cs | 400–2000 AD | Daga et al., 2016 |
| d6 | Pinheiro Mire | Minas Gerais, Brazil | 18°03'44" S 43°39'42" W | 1230–1270 | Peat bogs | 2.2 | ¹⁴ C | 54990 BC–2010 AD | Peirez-Rodriguez et al., 2015 |
| d7 | Yanacocha | Southeast Peru | 13°56'42" S 70°52'30" W | 4910 | Lake sediments | 3.33 | ²¹⁰ Pb, ¹⁴ C | 10290 BC–2010 AD | Beal et al., 2014 |
| d8 | Huancavelica | Central Peru | 14°51'27" S 75°24'38" W | / | Lake sediments | / | ²¹⁰ Pb, ¹⁴ C | 2800 BC–2000 AD | Cooke et al., 2009 |
| e1 | Berg River | South Africa | 32°47'26" S 18°12'05" E | / | Coastal salt marsh | 0.3 | ²¹⁰ Pb | 1900–2007 AD | Kading et al., 2009 |
| e2 | Lake Tanganyika | East Africa | 04°40'–06°34' S 29°37'–29°59' W | / | Lake sediments | 0.28 | ²¹⁰ Pb, ¹⁴ C | 1600–2000 AD | Conaway et al., 2012 |
| f1 | King George Island | Western Antarctica | 62°11'57" S 59°58'48" W | / | Seal hairs | 0.425 | ¹³⁷ Cs, ¹⁴ C | 18–2002 AD | Sun et al., 2006 |
| f2 | Adélie Basin | Southern Antarctica | 66°12'53" S 140°26'17" E | / | Diatom ooze sediments | 170 | Diatom fossils | 6600 BC–2000 AD | Zaferani et al., 2018 |
| f3 | Dome C | Antarctica | 75°06'00" S 123°21'00" E | 3233 | Ice core | 3062.4 | δ ¹⁸ O | 652000 BC–2000 AD | Jitaru et al., 2009 |
| f4 | Dome C | Antarctica | 77°39'00" S 124°10'00" E | 3240 | Ice core | 905 | δ ¹⁸ O | 31710 BC–1990 AD | Vandal et al., 1993 |

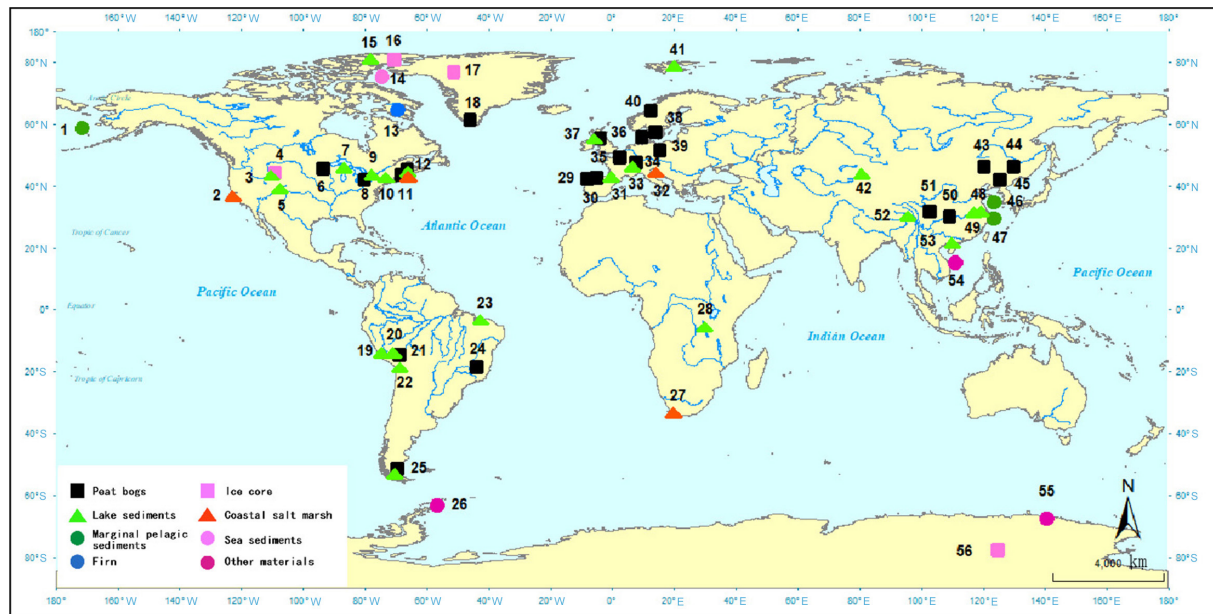


FIGURE 1 | Distribution of mercury deposition research spots. 1. Bering Sea (Kim et al., 2019); 2. San Francisco Bay (Donovan et al., 2013); 3. Wyoming (Kurz et al., 2019); 4. California (Conaway et al., 2004); 5. Colorado (Gray et al., 2005); 6. Minnesota (Benoit et al., 1998); 7. the Great Lakes (Grant et al., 2014); 8. Ontario (Givélet et al., 2004); 9. Lake Ontario (Wiklund et al., 2017); 10. New York (Arnason and Fletcher, 2003); 11. Maine (Roos-Barraclough et al., 2006); 12. the Bay of Fundy (Sunderland et al., 2008); 13. Baffin Island (Zdanowicz et al., 2015); 14. the Arctic Archipelago (Štok et al., 2019); 15. Ellesmere Island (Korosi et al., 2018); 16. Arctic Canada (Zdanowicz et al., 2016); 17. northwest Greenland (Zheng, 2015); 18. Narsaq Peninsula (Shotyk et al., 2003); 19. Huancavelica (Cooke et al., 2009); 20. Yanacocha (Beal et al., 2014); 21. Lake Titicaca region (Guédron et al., 2018); 22. Lake Chungará (Guédron et al., 2019); 23. Maranhão state (De Lacerda et al., 2017); 24. Minas Gerais (Peirez-Rodríguez et al., 2015); 25. Patagonia (Biester et al., 2002, 2003; Hermanns and Biester, 2013a; Daga et al., 2016); 26. King George Island (Sun et al., 2006); 27. Berg River (Kading et al., 2009); 28. Lake Tanganyika (Conaway et al., 2012); 29. Xistral Mountains (Martínez-Cortizas et al., 2012); 30. Asturias (Gallego et al., 2013); 31. Pyrenees (Corella et al., 2017); 32. Adriatic Sea (Covelli et al., 2012); 33. Lake Lucerne (Thevenon et al., 2011); 34. Swiss Jura Mountains (Roos-Barraclough and Shotyk, 2003); 35. Belgium (Allan et al., 2013); 36. Denmark (Shotyk et al., 2003); 37. Scotland (Farmer et al., 2009; Küttner et al., 2014); 38. Sweden (Bindler et al., 2004); 39. the Czech Republic (Ettler et al., 2008); 40. Norway (Steinnes and Sjobakk, 2005); 41. Svalbard (Drevnick et al., 2012); 42. Mount Tianshan (Zeng et al., 2014); 43. Greater Khingan Range (Bao et al., 2016); 44. Lesser Khingan Range (Xu, 2010; Tang et al., 2012); 45. Hani peat bog (Xiao, 2017); 46. Yellow Sea (Kim et al., 2019); 47. East China Sea (Lim et al., 2017; Zhang H. X. et al., 2018; Zhang R. et al., 2018); 48. Shanghai (Yang H. D. et al., 2016); 49. Chao Lake (Zhang H. X. et al., 2018; Zhang R. et al., 2018); 50. Dajiu Lake Basin (Li et al., 2016, 2017); 51. Hongyuan (Shi et al., 2011); 52. Tibetan Plateau (Wang et al., 2010; Yang et al., 2010); 53. Huguangyan Maar Lake (Zeng et al., 2017; Han, 2018); 54. Xisha Islands (Xu et al., 2010; Liu et al., 2012); 55. Adélie Basin (Zaferani et al., 2018); and 56. Dome C (Vandal et al., 1993; Jitaru et al., 2009).

and other industrial activities). Due to the interaction between these factors and regional differences in human activities, the mercury deposition changes in different regions showed both certain commonalities and relatively large differences through time. Based on previous studies, the history of global mercury deposition was divided into the following four stages:

670 to 11.7 ka BP, Before the Holocene

The Dome C ice core in Antarctica provided a mercury deposition time series for past 670 ka (Figure 2). This record revealed mercury value peaks during glacial periods, while in the warm interglacial periods, the Hg concentration was relatively low (Jitaru et al., 2009). Similar trends occurred in the ice core records of Dome C for the past 34 ka, with greater temporal resolution (Figure 3A): The mercury concentration was relatively low 34 ka BP, and then it rose, maintaining a high value during 28 to 18 ka BP, corresponding to the Last Glacial Maximum (LGM, ~26 to 16 ka BP); there was a reduction in 17 to 13 ka BP, representing the transition from the Last Glacial Period to the Holocene. According to peat records in southeastern

Brazil (Figure 3A), the mercury concentration fluctuation 57 to 28 ka BP was relatively smooth, and the change in mercury concentration was mainly affected by atmospheric wet deposition related to precipitation and mineral input in the catchment area (Peirez-Rodríguez et al., 2015). The mercury concentration suddenly decreased to its lowest value 27 ka BP and then increased sharply and remained high from 26 to 17 ka BP, coinciding with the LGM (Figure 3A; Peirez-Rodríguez et al., 2015). Similar glacial to interstadial contrast in mercury levels was reported for lake sediment records in northeastern Brazil (De Lacerda et al., 2017). Records from this lake showed decreased Hg concentrations at ca. 17 to 13 ka BP, with subsequent recovery to the level prior to 17 ka BP, and until the sudden increase ca. 12 ka BP, corresponding to the Younger Dryas (YD) cooling event (De Lacerda et al., 2017). The mercury accumulations in the Dajiu Lake Basin, the Swiss Jura Mountains, and Peru also showed a significant increasing trend before and after the YD event (Figure 3B). Through the comparison of the above data, we concluded that glacial and interglacial timescales ca. the past 600 ka global mercury deposition appears to fluctuate in parallel

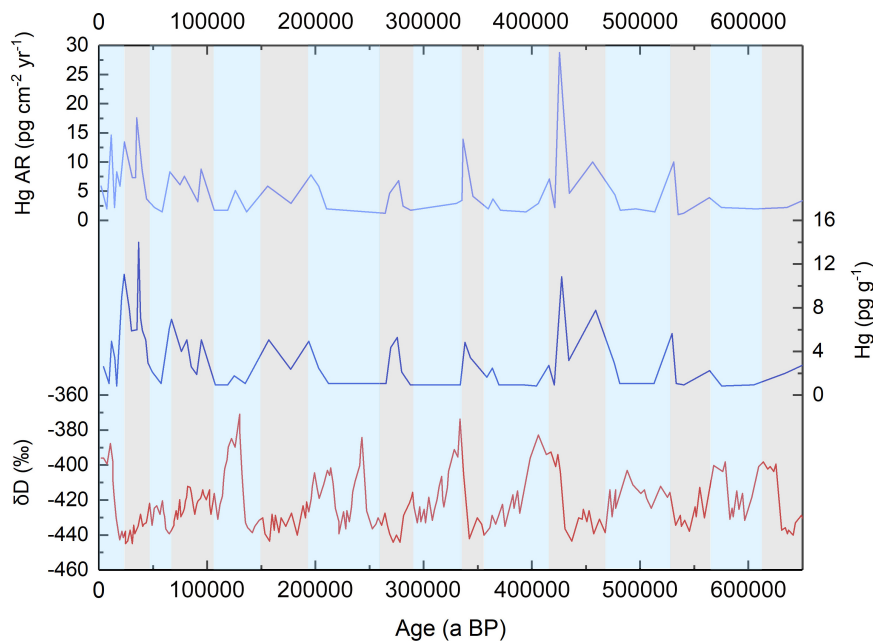


FIGURE 2 | Mercury deposition and temperature change in Antarctica since 670,000 a BP. The data were based on the research results of Petru Jitaru et al. in Dome C ice core of Antarctica, the red curve represents δD (‰), indicating the temperature change (Jitaru et al., 2009).

with broad climate variability with more Hg accumulation in the glacial periods and less accumulation in the interglacial periods.

11.7 to 8.2 ka BP, the Early Holocene

Post YD cooling, in general there was warming in to an interglacial climate. However, in the Early Holocene, the climate was still unstable and there were many climate oscillations sourced in the North Atlantic area, including the 11.1 ka BP event, 10.3 ka BP event, 9.4 ka BP event, 8.2 ka BP event, and possibly other climate variability (Bond et al., 1997). According to the mercury deposition records of the Dajiu Lake Basin, the Swiss Jura Mountains, Lake Titicaca, Yanacocha of Peru, and Maine United States (Figure 3B), both the mercury concentration and mercury accumulation rate for the Early Holocene were relatively low and Hg-depositional variability was linked to abrupt changes, especially during the 8.2 ka BP cooling event in the N. Atlantic. The mercury deposition in the Dajiu Lake Basin, the Swiss Jura Mountains, Lake Titicaca, Yanacocha in Peru, and Maine in the United States reached a peak. Through comparison of the regional data, it was discovered that in the Early Holocene, the mercury accumulation rate in the Dajiu Lake Basin of China was higher than that in the Swiss Jura Mountains and Maine in the United States, among other locations. Meanwhile, data from various regions showed that atmospheric mercury deposition was unstable during this period, with substantial fluctuations.

8.2 to 4.2 ka BP, the Mid-Holocene

Many studies showed that the climate in the Mid-Holocene was generally warm and humid (Dwyer et al., 1996; Haug et al., 2001; Kaufman et al., 2004). Global atmospheric mercury deposition was also characterized by relatively consistent values in the

Mid-Holocene (Figures 3B,C). It is worth noting that the changes in mercury accumulation in some areas during this period were related to specific conditions. For example, the enrichment of mercury deposition from 8 to 6 ka BP in the Swiss Jura Mountains was related to volcanic eruptions (Roos-Barraclough et al., 2002); the mercury peaks of Lake Titicaca from 6 to 5 ka BP were caused by rich orographic precipitation in the eastern part of the Andes (Guédron et al., 2018). The comparison of the regional data showed that in the Mid-Holocene, the characteristics of atmospheric mercury deposition in the Dajiu Lake Basin and Lesser Khingan Range of China, the Swiss Jura Mountains, Spain, Norway, Maine, and Ontario, among other locations, were similar, and the atmospheric mercury accumulation rate and mercury concentrations were at a relatively low level, with the atmospheric mercury deposition being weaker and significantly lower than that during the Early Holocene.

4.2 to 0 ka BP, the Late Holocene

The Late Holocene is a stage when human civilization has developed rapidly and human activities have had a profound impact on natural systems. Since the Industrial Revolution, the population has increased dramatically and the impact of human activities on earth system reached an unprecedented magnitude. Under the multiple effects of natural systems and human activities, regional and global atmospheric mercury deposition changed significantly and the rate of atmospheric mercury accumulation increased to its highest value during historical periods. Atmospheric mercury deposition between 4.2 and 0 ka BP in all regions of the world continued to rise (Figures 3C,D), especially in the past hundreds of years. The atmospheric mercury accumulation rate and the mercury

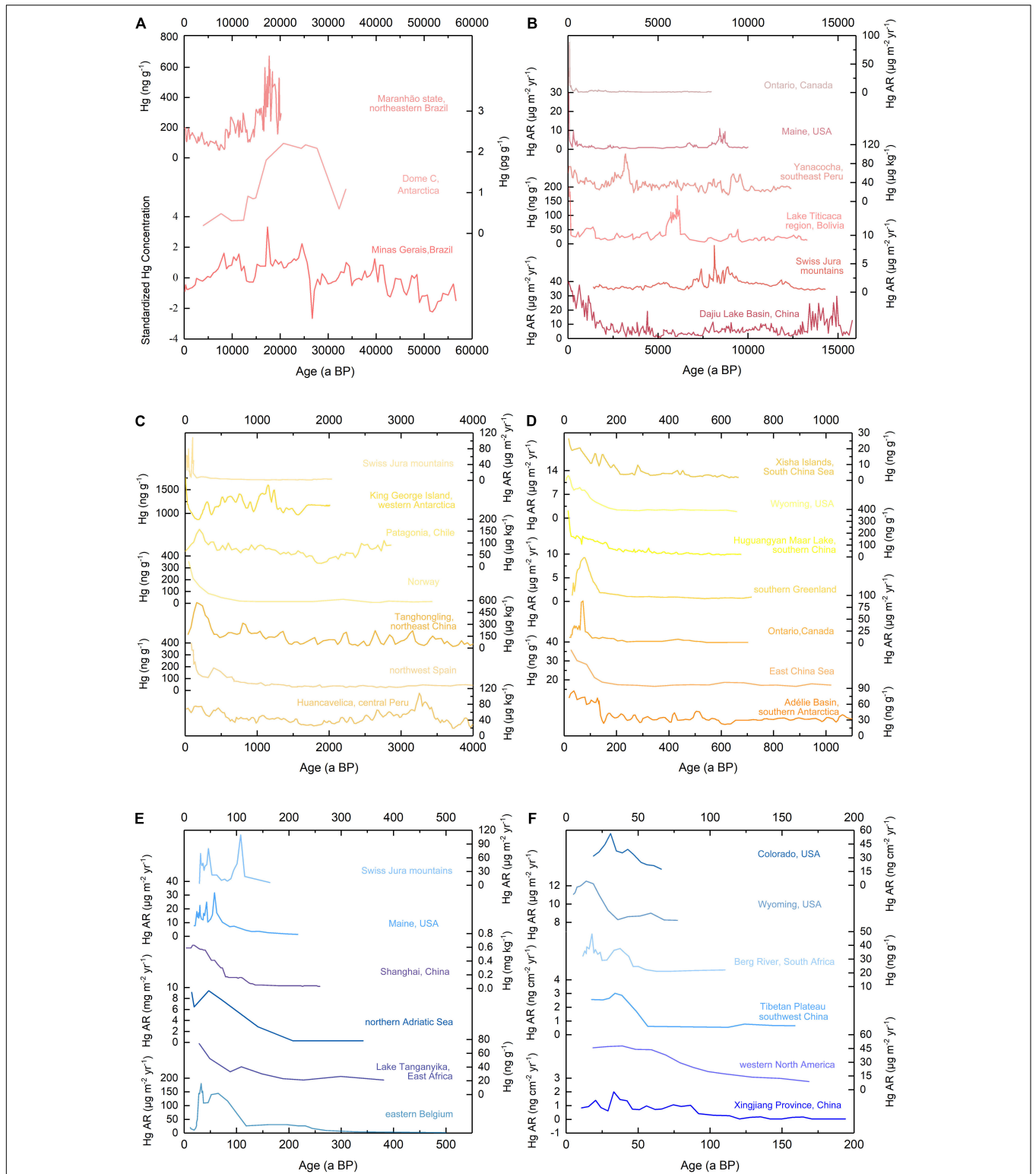


FIGURE 3 | Changes of mercury deposition in different time scales around the world. **(A)** From bottom to top: Standardized Hg concentrations in Minas Gerais, Brazil (Peirez-Rodriguez et al., 2015); Hg concentrations in Dome C, Antarctica (Vandal et al., 1993); and Hg concentrations in Maranhão state, northeastern Brazil (De Lacerda et al., 2017). **(B)** From bottom to top: Hg Accumulation Rate (Hg AR) in Dajiu Lake Basin, China (Li et al., 2016); Hg AR in Swiss Jura Mountains (Roos-Barraclough et al., 2002); Hg concentrations in Lake Titicaca region, Bolivia (Guédron et al., 2018); Hg concentrations in Yanacocha, southeast Peru (Beal et al., 2014); Hg AR in Maine, United States (Roos-Barraclough et al., 2006); and Hg AR in Ontario, Canada (Givelet et al., 2004). **(C)** From bottom to top: Hg (Continued)

FIGURE 3 | Continued

concentrations in Huancavelica, central Peru (Beal et al., 2014), northwest Spain (Martínez-Cortizas et al., 1999), Tanghongling, northeast China (Tang et al., 2012), Norway (Steinnes and Sjøbakk, 2005), Patagonia, Chile (Biester et al., 2003), King George Island, western Antarctica (Sun et al., 2006); and Hg AR in Swiss Jura Mountains (Roos-Barracough et al., 2002). **(D)** From bottom to top: Hg concentrations in Adélie Basin, southern Antarctica (Zaferani et al., 2018), East China Sea (Kim et al., 2019); Hg AR in Ontario, Canada (Givelet et al., 2004), southern Greenland (Pérez-Rodríguez et al., 2017); Hg concentrations in Huguangyan Maar Lake, southern China (Zeng et al., 2017); Hg AR in Wyoming, United States (Kurz et al., 2019); and Hg concentrations in Xisha Islands, South China Sea (Huang et al., 2013). **(E)** From bottom to top: Hg AR in eastern Belgium (Allan et al., 2013); Hg concentrations in Lake Tanganyika, East Africa (Conaway et al., 2012); Hg AR in northern Adriatic Sea (Covelli et al., 2012); Hg concentrations in Shanghai, China (Yang H. D. et al., 2016); and Hg AR in Maine, United States (Roos-Barracough et al., 2006), Swiss Jura Mountains (Roos-Barracough et al., 2002). **(F)** From bottom to top: Hg AR in Xingjiang Province, China (Zeng et al., 2014), western North America (Drevnick et al., 2016), Tibetan Plateau, southwest China (Wang et al., 2010); Hg concentrations in Berg River, South Africa (Kading et al., 2009); and Hg AR in Wyoming, United States (Kurz et al., 2019), Colorado, United States (Gray et al., 2005).

concentration reached their highest values during the Mid- and Late Holocene (**Figures 3D,F**). This was mainly due to the significant increase in mercury emissions from human activities after the Industrial Revolution, which led to an increase in the rate of atmospheric mercury accumulation of several times or even tens of times.

Through the comparison of atmospheric mercury deposition data at a global scale, it could be inferred that the characteristics of mercury deposition in European countries represented by Switzerland, Greenland, Spain, and Belgium, among others, are similar to those of the North American countries represented by the United States and Canada. Specifically, beginning with the Industrial Revolution in the mid-19th century, the mercury deposition rate increased rapidly, and mercury accumulation reached a peak value in the mid- and late 20th century and then showed a downward trend. With the increasing emphasis on the environmental pollution caused by a large number of mercury emissions in European and North American countries, measures were taken in various regions to reduce mercury emissions (Roos-Barracough and Shotyk, 2003; Bindler et al., 2004; Ettlér et al., 2008; Sunderland et al., 2008; Farmer et al., 2009; Drevnick et al., 2012; Allan et al., 2013; Corella et al., 2017). The atmospheric mercury deposition rate in the Asian regions represented by Shanghai, the Xisha Islands, Huguangyan Maar Lake, and the East China Sea increased slowly after industrialization. The rate began to increase rapidly during the 1960s to 1970s, with mercury deposition continuing to rise. From the above comparison, it could be deduced that since the 1970s, the center of global mercury production has gradually shifted from Europe and North America to Asia (Xu et al., 2010; Zeng et al., 2017).

Mercury Deposition and Climate Change

In studies around the world, the impact of climate change on mercury deposition has been the focus of researchers with various outcomes. Most scholars believe that a cold and dry climate is conducive to mercury accumulation, while a warm and humid climate limits the accumulation of mercury (Roos-Barracough et al., 2002, 2006; Jitaru et al., 2009; Gallego et al., 2013; Peirez-Rodríguez et al., 2015; Li et al., 2016; De Lacerda et al., 2017). Some scholars believe that there is more mercury accumulation during humid-climate periods and less accumulation of mercury during dry-climate periods (Guédron et al., 2018). Related to this issue, in the following section, two aspects of climate, namely, temperature change and humidity change, are analyzed and summarized.

Dry and Wet Deposition of Atmospheric Mercury

Mercury emitted from anthropogenic and natural sources to the atmosphere eventually falls back to the surface through dry and wet deposition. Dry deposition refers to the deposition of aerosol particles. An aerosol is a gas dispersion composed of solid or liquid particles suspended in a gas medium, which has a complex chemical composition (Wang, 2011). Wet deposition refers to the process of removing particles from the atmosphere through rainfall, snowfall, and other phenomena.

Atmospheric mercury deposition is a process through which various forms of mercury are removed from the atmosphere. The dry deposition of mercury mainly includes the direct deposition of Hg(0) and RGM, which occurs throughout the year as long as it does not rain heavily. The forms of mercury in wet deposition tend to be soluble and granular Hg(II). Due to the long-distance diffusion and water solubility of gaseous Hg(II), most of the mercury is deposited via Hg(II) dissolved in atmospheric water or adsorbed on the surface of raindrop particles (Fitzgerald et al., 1991). Mercury can react with O₃ in the atmosphere to form water-soluble Hg(II), which can be adsorbed on particles or dissolved in water and then deposited. Inorganic mercury compounds, such as methylmercury, deposit rapidly because of their solubility (Munthe et al., 1995). Although the wet deposition process of Hg(II) contributes greatly, it represents less than 5% of the total mercury in the atmosphere, and the continuous dry deposition of mercury almost all year is still dominant. Recent studies employing field monitoring and mercury isotopes showed that Hg(0) dry deposition far exceeds wet deposition by precipitation and the former can even be five times greater than the latter (Selin and Jacob, 2008; Enrico et al., 2016; Obrist et al., 2017; St. Louis et al., 2019).

At present, the deposition mechanism and long-term monitoring sites of mercury wet deposition flux in many countries are well established. As one of the countries emitting the most mercury in the world, China has yet to establish a relatively complete monitoring system for wet mercury deposition. Additionally, the measurement and evaluation of dry mercury deposition are mostly local or achieved by atmospheric models, and there are few published relevant records.

Impact of Temperature Change on Mercury Deposition

To evaluate the impact of temperature change on mercury deposition, the mercury deposition records of Maine of the United States, southeastern Peru, the Swiss Jura Mountains,

Patagonia of Chile, the Shennongjia Dajiu Lake Basin of China, Maranhão of Brazil, Dome C of Antarctica, and Minas Gerais of Brazil were selected for comparison, as shown in **Figure 4**. During the LGM (~26–16 ka BP), the mercury concentration in Antarctica, as well as Minas Gerais and Maranhão, remained at a high level. From 17 to 13 ka BP, the mercury concentration showed a downward trend until it reached its peak again from 13 to 11.5 ka BP, consistent with the YD event, and the mercury accumulation in the Dajiu Lake Basin and Patagonia also showed an obvious upward trend before and after the YD event. From 11.5 to 9 ka BP, the mercury concentration dropped gradually, but the decrease was small, and it increased significantly and reached its peak from 9 to 8 ka BP, corresponding to the 8.2 ka BP event, with the strongest cooling effect during the Holocene.

After 8.2 ka BP, the mercury accumulations generally showed a slow downward trend. The peak values of mercury accumulation in the Mid- and Late Holocene were mainly affected by volcanic eruptions and human activities (Vandal et al., 1993; De Lacerda et al., 2017). Volcanic emissions are thought to be an important natural source of atmospheric Hg. On millennial timescales, dust from volcanic eruptions might affect mercury deposition in paleoclimate records. Hg enrichment in Patagonia occurred during the periods of three volcanic eruptions two eruptions of Mt. Burney (8300 a BP and 4250 a BP) and one eruption of the Mt. Hudson volcano (7800 a BP; **Figure 4D**; Biester et al., 2003). Many volcanoes erupted between 8 and 6 ka BP in Europe, which explained the peaks of the Hg accumulation rate in the Swiss Jura Mountains (**Figure 4C**; Roos-Barracough et al., 2002).

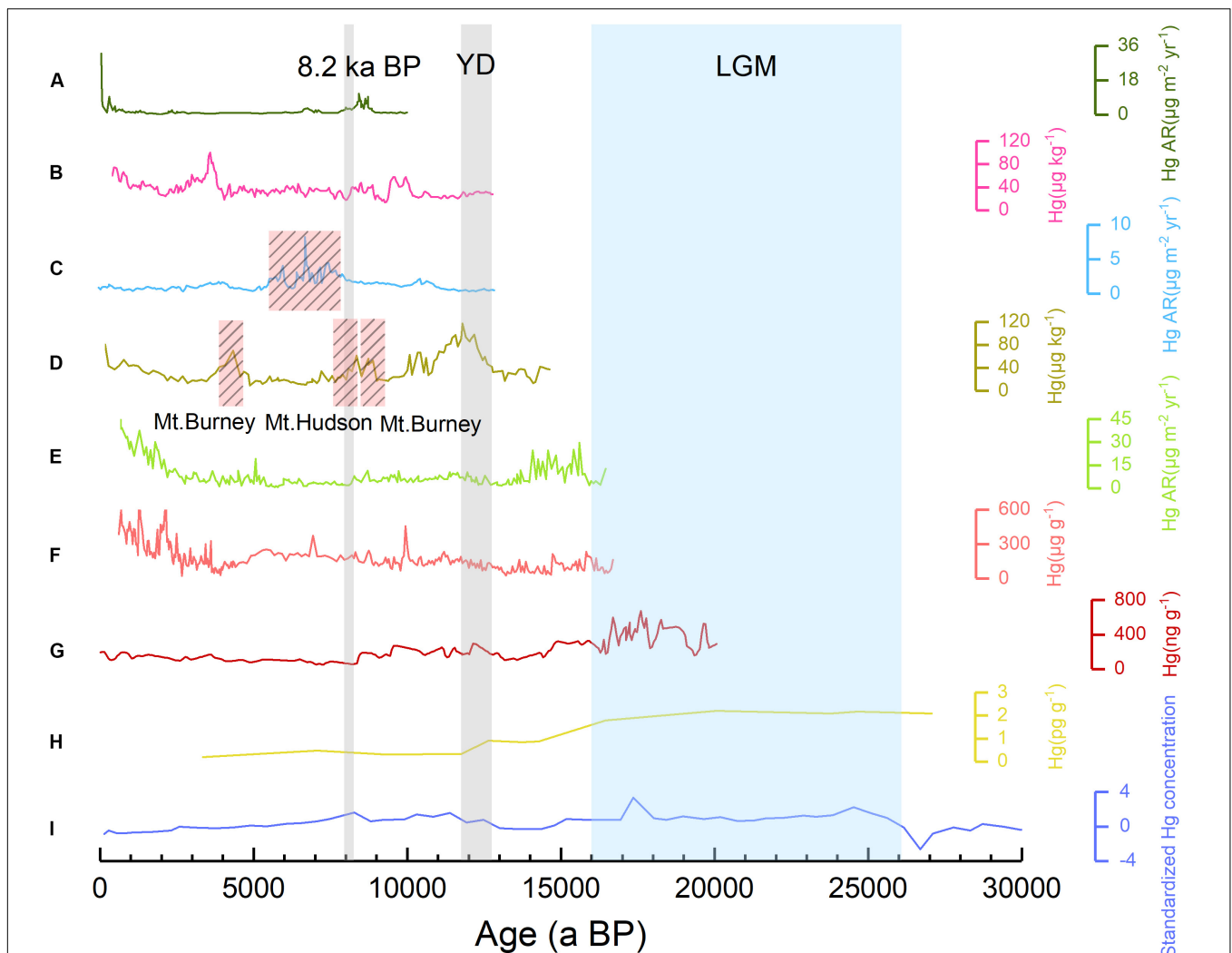


FIGURE 4 | The relationship between mercury deposition and temperature change. **(A)** Hg AR in Maine, United States (10000 a BP; Roos-Barracough et al., 2006); **(B)** Hg concentrations in Yanacocha, southeast Peru (12500 a BP; Beal et al., 2014); **(C)** Hg AR in Swiss Jura Mountains (12000 a BC; Roos-Barracough et al., 2002); **(D)** Hg concentrations in Patagonia, Chile (14500 a BP; Biester et al., 2003); **(E)** Hg AR in Shennongjia Dajiu Lake Basin of China (16000 a BP; Li et al., 2016); **(F)** Hg concentrations in Shennongjia Dajiu Lake Basin of China (16000 a BP; Li et al., 2017); **(G)** Hg concentrations in Maranhão state, northeastern Brazil (20000 a BP; De Lacerda et al., 2017); **(H)** Hg concentrations in Dome C, Antarctica (27000 a BP; Vandal et al., 1993); **(I)** Standardized Hg concentrations in Minas Gerais, Brazil (30000 a BP; Peirez-Rodriguez et al., 2015).

At longer timescales, it could be concluded that the temperature change from 30 to 8 ka BP was consistent with the fluctuation of atmospheric mercury deposition, especially during the LGM, YD event, and 8.2 ka BP event, and atmospheric mercury deposition showed a strong response. Petru Jitaru et al. reconstructed mercury deposition records and temperature changes since 670 ka BP at Dome C, Antarctica. As shown in **Figure 2**, there is a good correlation between peak mercury accumulation and cold periods. Based on these analyses, it could be concluded that a cold climate is conducive to mercury accumulation. According to the studies of different scholars, the following four reasons could be summarized regarding the specific mechanism: (1) Due to the high volatility of mercury, the mercury in the atmosphere is easily attached to objects such as particulate matter under cold conditions and then settles on the ground surface, known as the condensation effect (Roos-Barraclough et al., 2002; Gallego et al., 2013); (2) The atmospheric dust increases during cold periods, and this dust can carry the mercury in the atmosphere to the ground surface so that mercury accumulation is increased (Roos-Barraclough et al., 2002; Jitaru et al., 2009; Peirez-Rodriguez et al., 2015; De Lacerda et al., 2017; Li et al., 2017); (3) In colder environments, marine mercury emissions are higher (Vandal et al., 1993; Roos-Barraclough et al., 2002; Peirez-Rodriguez et al., 2015; De Lacerda et al., 2017) and the high-productivity marine emissions of gaseous mercury are important sources of atmospheric mercury (Vandal et al., 1993); and (4) The mercury released by soil degassing is lower during cold periods (Gallego et al., 2013). In conclusion, more exogenous dust inputs and an increase in mercury in the atmosphere during cold periods lead to mercury accumulation on the scale of tens of thousands of years, while on millennial timescales, mercury emitted from volcanic eruption to the atmosphere also has a certain impact on mercury deposition records.

Impact of Humidity Change on Mercury Deposition

The main effect of precipitation on mercury deposition is that an increase in precipitation promotes a rise in forest litterfall Hg deposition. Terrestrial vegetation often represents the first ecosystem compartment with which new atmospheric Hg interacts following deposition. It was recently demonstrated that a portion of newly wet-deposited Hg(II) may not initially pass directly through the forest canopy to the forest floor, but rather is retained over the growing season, only to be deposited later with litterfall (Graydon et al., 2006). Plant foliage provides an excellent surface for photochemical reduction of newly deposited Hg(II) remaining in the forest canopy following precipitation events (Lindberg et al., 1998). Litterfall Hg deposition is the major pathway for Hg loading into the forest (Zhou et al., 2013). Recent studies on the air-surface exchange of mercury and litterfall Hg deposition revealed that an increase in precipitation can elevate forest biomass, which in turn pumps more Hg(0) into litterfall and then soils (Lindberg et al., 1998; Graydon et al., 2006). It is generally believed that vegetation litter represents a net sink of atmospheric Hg (Lindberg et al., 1998; Demers et al., 2007; Pokharel and Obrist, 2011; Ma et al., 2017; Zhang et al., 2019). Therefore, an increase in precipitation may enhance absolute wet Hg deposition rather than wet deposition relative to dry

deposition via vegetation. However, research on the mechanism of mercury deposition affected by long-term humidity change is lacking. The results of studies conducted in Brazil and Lake Titicaca showed that the atmospheric wet deposition of mercury during humid periods increased, so a humid climate is considered to be conducive to mercury accumulation (Peirez-Rodriguez et al., 2015; Guédron et al., 2018). However, most scholars found a significant relationship between a cold-dry climate and peak mercury deposition. Unfortunately, the influence of temperature could not be excluded to judge the relationships between humidity changes and mercury deposition. Furthermore, due to the small amount of data and conclusions available for reference, the relationship between humidity change and mercury deposition cannot be determined. The authors believe that studies of the relationship between precipitation changes and mercury deposition changes can be strengthened over long timescales and a more accurate conclusion can be drawn by comparing the two types of change.

Mercury Deposition and Anthropogenic Activities

According to a large number of previous studies, atmospheric mercury deposition caused by human activities began approximately 3,500 years ago. Before industrialization, gold, silver, and mercury mining and the widespread use of cinnabar were the main sources of anthropogenic mercury. After industrialization, coal combustion, mercury mining, non-ferrous metal smelting, liquid mercury production, steel manufacturing, cement manufacturing, gold mining, waste incineration, and the development of the chlor-alkali industry led to a sharp increase in anthropogenic emissions of mercury, which in turn caused a sharp increase in mercury deposition.

Preindustrial Era

With the development of human civilization, mercury is being increasingly used in all aspects of society. Cinnabar has a bright red color and never fades, so it has long been used as a pigment. According to the literature, the use of cinnabar can be traced back to the Shang Dynasty (~1600–1046 BC) in China, and some characters engraved with cinnabar were found on unearthed animal bones or turtle shells (Li et al., 2016). Cinnabar was also used to color figurines and pottery; for example, the Terracotta Warriors were painted with cinnabar (Chen, 2017). In ancient times, mercury was considered an antiseptic, and mercury gas volatilized in underground palaces could prevent buried corpses and funerary objects from rotting for a long time. Since mercury is a highly toxic substance that can cause death in cases of inhalation at high volume, it was also used to poison tomb raiders (Liu and Wang, 2001; Duan, 2007). During the Qin and Han Dynasties of China (221 BC–220 AD), Chinese ancestors extracted elemental mercury from cinnabar, utilizing the easy solubility of gold and silver in mercury and the volatility of mercury to extract gold and silver from ores by using mercury, which was later called the “amalgamation process” (Li et al., 2016). For quite a long time in ancient times, the amalgamation process was the main method of gold smelting worldwide, until it was replaced by a more effective method with less mercury

pollution called the “cyanidation method” in the early 20th century (Sun et al., 2006). Mercury was also used in medicine. According to the manuscripts on silk named “Prescriptions for Fifty-two Diseases” in the Han Dynasty Changsha Mawangdui Tomb unearthed in 1973, transcription was performed during the Qin and Han Dynasties. It was the most ancient Chinese medicine prescription that has been discovered, in which mercury was used in four prescriptions. Moreover, mercury was once considered by Tibetans to be able to heal fractures, prolong life, and maintain good health (Wang et al., 2010; Yang et al., 2010).

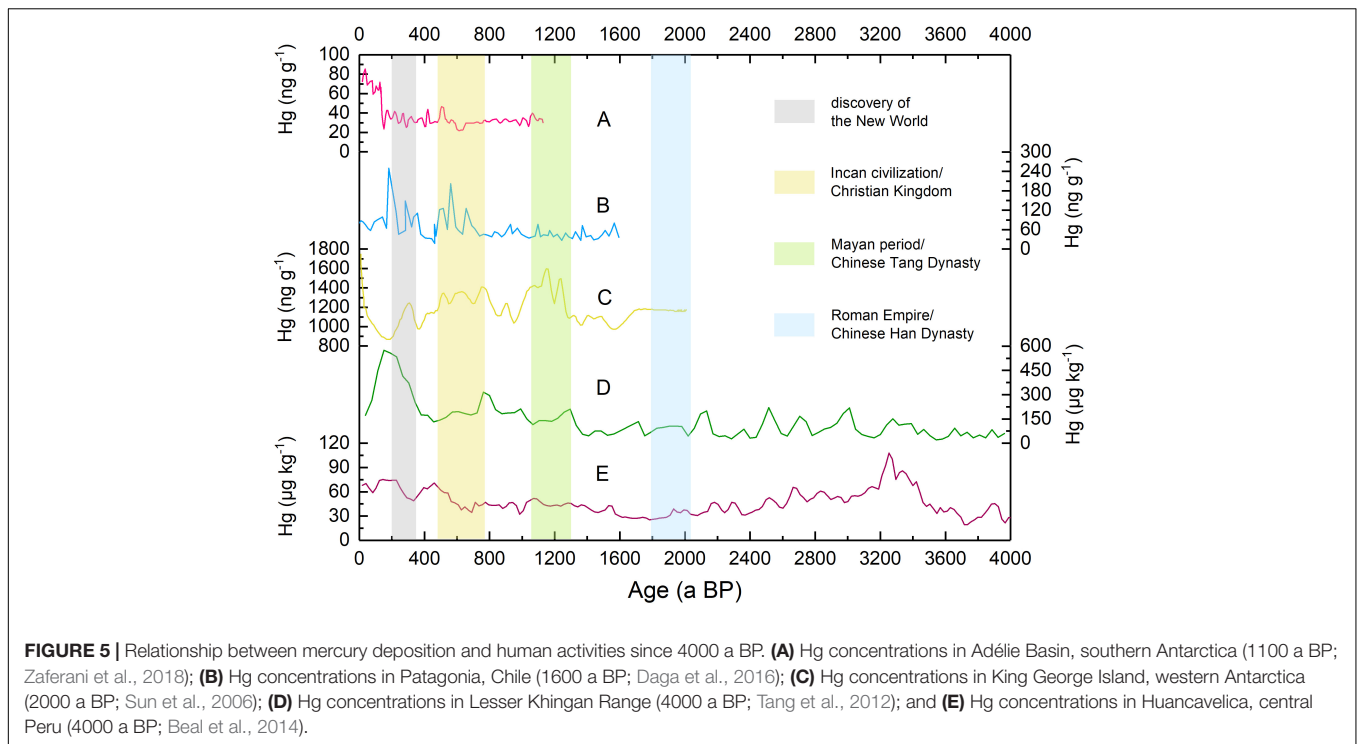
At approximately 1400 BC, the Huancavelica cinnabar mine in Peru began to be mined. During the period of rapid development and expansion of mining and metallurgy in the Andes (~500–1000 AD, ~1000–1400 AD), mercury emissions increased continuously (Cooke et al., 2009). In Europe, the increase in anthropogenic mercury 2500 years ago was consistent with the high mercury mining activities and metallurgical history of Spain (Martínez-Cortizas et al., 1999). **Figure 5** shows that the mercury concentrations in various regions were higher in some stages between 2000 years ago and the beginning of industrialization, namely, the Roman Empire (27 BC–1453 AD), the Chinese Han Dynasty (202 BC–8 AD, 25–220 AD), the Mayan period (from approximately the 10th century BC to the 16th century AD), the Chinese Tang Dynasty (618–907 AD), and after the discovery of the New World (1650–1800 AD). The high mercury concentrations were closely related to the large-scale mining activities during these periods. However, between these periods, with the decrease in mining activities, the mercury concentrations showed a downward trend. Between 18 and 300 AD, both the Roman Empire and the Chinese Han Dynasty engaged in many mining activities when alchemy became popular at the time (Martínez-Cortizas et al., 1999; Sun et al., 2006). After 300 AD, the mercury concentrations declined. Between 300 and 750 AD, the mercury concentrations were relatively low, corresponding to the collapse of Rome and the decline in China’s gold production (Brading and Cross, 1972; Sun et al., 2006). Between 1050 and 1250 AD, low mercury concentrations corresponded to the fall of Mayan civilization and the wars during the Southern Song Dynasty. In addition, the depletion of European silver mines and the suppression of gold mining also led to a decrease in mercury concentration (Sun et al., 2006). Since the beginning of 1250 AD, the mercury concentrations have increased, and during the period of Incan civilization in South America (from the 12th to 15th century), a large amount of gold was produced (Xiang and Huang, 2014). Meanwhile, new silver mines were discovered in Europe, and thousands of people joined the “Silver Tide” activities of the Christian Kingdom (Sun et al., 2006). After 1500 AD, the mercury concentrations fell to another low. Pizarro, a Spanish colonist, conquered the Inca in 1532 AD (Xiang and Huang, 2014), resulting in a reduction in mercury consumption. In the early 19th century, with the outbreak of the South American War of Independence, mercury consumption suddenly stopped and Spain, the main supplier of mercury in the world, also reduced mercury production (Martínez-Cortizas et al., 1999), leading to a decline in mercury concentration. With the rise of the Industrial Revolution in the mid-19th century, mercury concentrations began to rise sharply.

Industrial Age

Since the Industrial Revolution of 1840, coal combustion, non-ferrous metal smelting, and the chlor-alkali industry have become the main sources of anthropogenic mercury. Intensive industrial activities have caused unprecedented mercury pollution. According to previous studies, in Spain, the mercury accumulation after industrialization was 10 times as high as the level before industrialization (Serrano et al., 2013). In Norway, the figure was 15 (Steinnes and Sjøbakk, 2005); in Scotland, the figure was 20 (Farmer et al., 2009); and in Belgium, the figure was 63 (Allan et al., 2013). After the 1960s and 1970s, since European and North American countries began to pay attention to environmental issues and adopted a series of measures to reduce mercury emissions, such as closing the main mercury mines, reducing mercury emissions from flue gas emissions, reducing incinerator emissions, gradually phasing out mercury in waste streams, shutting down old facilities, and installing additional pollution control equipment (Benoit et al., 1998; Conaway et al., 2004; Sunderland et al., 2008; Yang H. D. et al., 2016; Yang J. et al., 2016; Corella et al., 2017; Obrist et al., 2018), mercury deposition was reduced to some extent. However, after the 1970s, with the rise of Asian countries, especially China and India, and the rapid development of industrialization and urbanization, Asia became the largest contributor to anthropogenic mercury, with its mercury emission accounting for more than half of the global emission in 2000 (Xu et al., 2010; Obrist et al., 2018). In recent decades, the massive production of mercury in Asia, coupled with the high mercury emissions from coal combustion, smelting, and waste incineration, has led to a significant increase in anthropogenic mercury emissions (Liu et al., 2012). This means that the mercury production center has gradually moved from Europe and North America to Asia.

Factors Influencing Mercury Deposition in Different Phases or at Different Timescales

Based on the previous analysis, it could be concluded that the main factors influencing mercury deposition include temperature change, human activities, and volcanic eruption, while the influencing factors vary among phases and timescales. According to **Figures 2, 4**, before the Middle and Late Holocene, when human activities had limited environmental impact, mercury deposition was consistent with temperature changes. That is, on the scale of hundreds of thousands of years, mercury accumulated more in cold periods and less in warm periods, and mercury mainly came from exogenous dust inputs. On millennial timescales, the correspondence between mercury deposition and temperature change shows limited significance, while the former is more closely related to volcanic eruption and human activities (**Figures 4, 5**; Vandal et al., 1993; De Lacerda et al., 2017). The frequent volcanic eruptions in the Middle Holocene led to the peak of mercury accumulation in this period (Roos-Barraclough et al., 2002). With the change in human activities in the past 3,500 years, mercury deposition has shown strong correspondence.



SUMMARY AND PROSPECTS

Summary

This study summarized the characteristics of mercury deposition from a global perspective by analyzing previous studies on mercury deposition on yearly to 100,000-year timescales. It was found that regarding the accumulation of mercury, there are certain commonalities within the regional range, while there are also differences among regions. In addition, both climate change and human activities have a significant impact on mercury deposition.

- (1) The common feature of global mercury deposition in the Holocene is that the accumulation was generally lower before the start of the Industrial Revolution of 1840 and posthaste increased rapidly. Alternatively, due to regional volcanic activities or human activities, such as mining and metallurgy, there are differences in mercury accumulations among regions. It is worth noting that since the 1970s, emission reduction measures have been adopted in Europe and North America to reduce mercury accumulation, and Asia has gradually become the global center of anthropogenic mercury emissions.
- (2) On the scale of hundreds of thousands of years, mercury accumulated more in cold periods and less in warm periods. On millennial timescales, the correspondence between mercury deposition and temperature change appears non-significant, and the former is more closely related to volcanic eruption and human activities. An increase in precipitation leads to a rise in forest litterfall Hg deposition. Little research has been performed on the effect

of long-term humidity change on mercury deposition, with inconsistent conclusions, so the specific impact of humidity change on mercury deposition is unknown.

- (3) The atmospheric mercury deposition caused by human activities can be traced back to 3500 years ago. Before industrialization, gold, silver, and mercury mining and the widespread use of cinnabar were the main anthropogenic mercury sources; after industrialization, coal combustion, non-ferrous metal smelting, waste incineration, and chlor-alkali industry development led to a significant increase in anthropogenic emissions of mercury, which in turn led to a sharp increase in mercury deposition.

Prospects

Although many studies have been conducted on the relationships of mercury deposition with climate change and human activities, with significant results, there remains some urgent challenges from a global perspective.

- (1) The mercury deposition monitoring and research sites are unevenly distributed. The research sites are mostly distributed in the Northern Hemisphere, including Western Europe, North America, and China, while the number of research areas distributed in the Southern Hemisphere is relatively small, located in only a few regions of South America, Africa, and Antarctica. There are no research sites in Eastern Europe, Northern and Central Asia, Northern Africa, or Oceania. In future studies, mercury deposition research in these gap areas can be strengthened to provide data for the global reconstruction of the evolutionary history of mercury deposition.

- (2) Most previous studies focused on the history of mercury deposition in the Holocene. There have been few studies on a longer timescale and even fewer studies on the relationship between precipitation change and mercury accumulation. As a result, the impact of climate change, especially humidity change, on the mercury accumulation mechanism has yet to be investigated. Therefore, it is suggested that studies of mercury deposition on a longer timescale and of the impact of precipitation on mercury accumulation be strengthened.
- (3) There is a lack of high-resolution mercury deposition reconstruction. In previous studies, fen peat and lacustrine deposits were widely used as natural archives, with few ice core records with high resolution and almost no stalagmite records available. To accurately analyze the relationships of mercury deposition with climate change and human activities, mercury deposition reconstruction at a higher resolution should be performed.

AUTHOR CONTRIBUTIONS

FL and CM designed the research. FL presented a synthesis of the state of the art of mercury deposition worldwide. FL and PZ completed the data collection, analysis, and interpretation.

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All authors listed have made substantial, direct and intellectual contributions to the work, and approved it for publication. All authors contributed to the article and approved the submitted version.

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Conflict of Interest: The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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