

Enhancing Atmospheric Mercury Research in China to Improve the Current Understanding of the Global Mercury Cycle: The Need for Urgent and Closely Coordinated Efforts

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The current understanding of the global mercury (Hg) cycle remains uncertain because Hg behavior in the environment is very complicated. The special property of Hg causes the atmosphere to be the most important medium for worldwide dispersion and transformation. The source and fate of atmospheric Hg and its interaction with the surface environment are the essential topics in the global Hg cycle. Recent declining measurement trends of Hg in the atmosphere are in apparent conflict with the increasing trends in global anthropogenic Hg emissions. As the single largest country contributor of anthropogenic Hg emission, China's role in the global Hg cycle will become more and more important in the context of the decreasing man-made Hg emission from developed regions. However, much less Hg information in China is available. As a global pollutant which undergoes long-range transport and is persistence in the environment, increasing Hg knowledge in China could not only promote the Hg regulation in this country but also improve the understanding of the fundamental of the global Hg cycle and further push the abatement of this toxin on a global scale. Then the atmospheric Hg research in China may be a breakthrough for improving the current understanding of the global Hg cycle. However, due to the complex behavior of Hg in the atmosphere, a deeper understanding of the atmospheric Hg cycle in China needs greater cooperation across fields.

■ HG AS A GLOBAL POLLUTANT

As a well-known toxin, mercury (Hg) exposures cause many adverse health effects in humans and wildlife.¹ All forms of Hg in the aquatic environment have the potential to convert to methylmercury (MMHg), a highly toxic form of Hg that accumulates in food chains. Consumption of fish with elevated MMHg is the principal pathway of human exposure to Hg.^{1,2}

Unlike other heavy metals, which are usually associated with air particles, Hg in ambient air exists predominantly in the gaseous form (>95%). The most significant releases of Hg emissions are to the air.³ Therefore, the atmosphere is an

important long-range transport route for this toxin. In the atmosphere, Hg has been analytically defined as three different species, including gaseous elemental Hg (GEM or Hg(0)), reactive gaseous Hg (RGM) or gaseous oxidized Hg (GOM, analytically defined, but may exist as Hg(II), for example, HgCl₂, HgBr₂, HgBrOH, HgO, etc.) and particulate Hg (PHg, analytically defined, chemical speciation unknown, likely also Hg(II)).³ The sources of atmospheric Hg include both primary natural emission (e.g., volcano eruption, geological Hg-enriched land emissions), primary anthropogenic emission (e.g., coal combustion, waste treatment and the production of metal, cement and Hg) and reemission from previously deposited Hg on various surface environments (e.g., soil, water, and vegetation).³ Hg emissions into the atmosphere from human activities generally include all three forms. The primary natural emission and reemission mainly emit GEM.³ The three Hg species have very different atmospheric behaviors.³ RGM has a short atmospheric lifetime and deposits near emission sources because of its high water solubility. The atmospheric lifetime of PHg is generally short and strongly depends on the meteorological condition and its chemical composition. GEM has a long atmospheric lifetime because of its low solubility in water and lack of reactivity in the atmosphere. Under normal atmospheric conditions it persists long enough (0.5–1 years) to be mixed well and disperses globally before it is oxidized by atmospheric oxidants (such as Br, OH, O₃, BrO, etc.) to RGM. Therefore, GEM has been identified as a global pollutant.³

Hg is largely deposited to the terrestrial and aquatic surfaces as Hg(II) from the atmosphere. After being deposited on soil, water and vegetation, Hg(II) can reduce to Hg(0) and convert to MMHg by abiotic and biotic processes.⁴ Recently deposited Hg(II) is more available for reduction and methylation than those already present in ecosystems.⁵ Due to its low solubility and low reactivity, the formed Hg(0) can be released to the overlaying air and be redistributed on a global scale through atmospheric transport.³ These special properties of Hg mean that once released into the environment, Hg can continue to cycle through the air, water and land for a long time until returning to the long-lived reservoir (e.g., deep-ocean sediments).

In the past several decades, many environmental authorities have listed Hg as a priority pollutant.⁶ In the period of 1950s–1980s, Hg regulations were mainly subject to domestic and regional environmental control in developed regions.⁶ As the scientific knowledge has grown, especially new data on health risks from low-level Hg exposure,⁷ many developing countries are also beginning to take measures to address Hg pollution.² Now, it is widely accepted that only expanded regional and

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international collaborative action can effectively address global Hg pollution.⁶ At present, although it is facing potential difficulties, many countries are participating the negotiation of a global legally binding instrument on Hg under the coordination of UNEP.⁸

■ THE INCREASING IMPORTANCE OF CHINA'S ROLE IN THE GLOBAL HG CYCLE

As an emerging economic giant of the world, China plays a significant, sometime dominant, role in shaping environmental outcomes for the planet at present and in the foreseeable future. This includes the Hg issue.

In the past three decades, the growth in demand for energy and raw materials (e.g., metal, cement), the coal-dominated energy structure and the massive production of Hg for industry and consumer goods (e.g., polyvinyl chloride and chlor-alkali production, battery, fluorescent lamp, thermometer, electrical and electronic switches) for domestic and international markets make China a major producer and consumer of this metal. China is also a significant and growing man-made emitter of this toxin.^{2,3} Meanwhile, for protecting human health and ecosystems, mitigating climate change and pursuing sustainable development, China is putting forward substantial efforts to meet the environmental challenge. This includes developing clean, renewable and low-carbon energies, promoting afforestation, introducing safer alternative and cleaner technologies and implementing more stringent pollutant emission standards and effective waste management. These efforts have a large direct and indirect environmental benefit, including reduction in Hg emissions.

The surface environments, such as soil, forest, and ocean, play a crucial role in the atmospheric Hg cycle via serving as source and sink of atmospheric Hg. The ecosystem change induced by climate change or human activities may alter the source–sink relationship of Hg. As the third largest country in size (960 Mha in total land area and 300 Mha in ocean area), China has various types of ecosystems, ranging from glaciers, tundra and deserts to grasslands, wetlands, forest, lakes, and oceans. Due to climate change, overgrazing, land reclamation for agriculture and extension of urban and industrial complex, desertification and deforestation have affected more than a quarter of China. The ecosystem change in China may have remarkable influence on the regional and global Hg cycle.

Although there are many significant improvements in the understanding of the global Hg cycle in the past several decades, substantial uncertainty remains on some important processes.³ For example, a recent analysis of worldwide trends in atmospheric Hg concentrations showed a 20–38% decline in atmospheric Hg concentrations from 1996 to 2009 at background stations in the Northern and Southern Hemisphere.⁹ A smaller declining trend was also reported at other background regions (e.g., the Mace Head station in Ireland¹⁰ and the Alert station in the Canadian Arctic¹¹). However, a recent estimate given by Streets et al.¹² showed that the global emission of all forms of Hg (GEM/RGM/PHg) from human activities have increased since 1950 and surged dramatically since 2000.¹² The increase was dominated by emission from Asia, especially from China.¹² As mentioned above, GEM, the dominant form of atmospheric Hg, has an atmospheric lifetime of up to one year and should to be mixed well in the global atmosphere before removal, then these declining measurement trends are in apparent conflict with the increasing trends in primary anthropogenic Hg(0) emissions. Hg has been effectively regulated in developed countries since the 1970s. The man-made Hg emissions from

developed countries are being reduced due to the effective regulation since the 1970s.^{2,3,6,12} Therefore, in this context China will play an increasingly important role in the global Hg cycle in the foreseeable future. As the major producer, consumer and emitter of Hg, the limited knowledge in China, especially about the atmospheric processes, is a critical barrier to current understanding of the global Hg cycle. Then enhancing atmospheric Hg research in China (e.g., the source and fate of atmospheric Hg, the Hg speciation profile in flue gases and Hg exchange between air and surface environment) will significantly improve the understanding of the global Hg cycle.

■ THE NEED FOR URGENT AND CLOSE COORDINATED EFFORTS

Since the late 1970s, scientists have been concerned about Hg pollution in China.¹³ As the research progressed in the past four decades, the topics also gradually expanded from point sources of Hg pollution to the fate and cycling of Hg in various environmental compartments and ecosystems.^{13–15} However, due to the lack of technical, financial, and institutional resources, the research and science knowledge related to Hg in China is limited, and the available Hg information is fragmentary and inconclusive.^{13,14} The dilemma of the growing importance of China's role and the knowledge gap not only affects Hg regulation in China but also the deep understanding of fundamental processes in the global Hg cycle (e.g., atmospheric Hg oxidation pathway and the interaction between atmosphere–land–ocean). This hampers the achievement of a fair global Hg treaty. Therefore, it is essential to develop a framework for interpreting available information in the context of high uncertainty and an effective strategy for prioritizing research to reduce these large uncertainties of Hg in China.

Recently, many studies have been published that discuss the status of Hg emission, pollution, and environmental and human health risk in China.^{13–17} In this paper, rather than discussing the latest progress in Hg science, we will briefly review the deficiencies of atmospheric Hg research in China, which is a critical barrier to the current understanding of the global Hg cycle, and then recommend several research priorities that have close connections. Hopefully, the following proposed research priorities, especially the integration of these topic (see Figure 1), could promote the understanding of Hg in China and subsequently lead to major advances in Hg science and reduce the uncertainties in our current understanding of the global Hg cycle.

■ RESEARCH NEEDS

Reduce the Uncertainties of Anthropogenic Hg Emission Inventory. A reliable emission inventory is fundamental for understanding the regional and global Hg issue both in scientific (e.g., model application, the estimate of global atmospheric Hg trend) and policy (e.g., international Hg treaty) perspectives. It will also be essential to meet future needs of the domestic and international cap and trade system. Globally, uncertainties in emissions from stationary combustion, such as coal-fired power plant and various industrial processes are estimated at 25% and 30%, respectively, and uncertainties in other sectors, such as waste treatment, small-scale gold mining, and large-scale Hg mining, are much higher, up to a factor of 2–5.³ Anthropogenic Hg emissions sources in developed countries such as the United States and those in Europe are better known than those in developing countries.^{2,3}

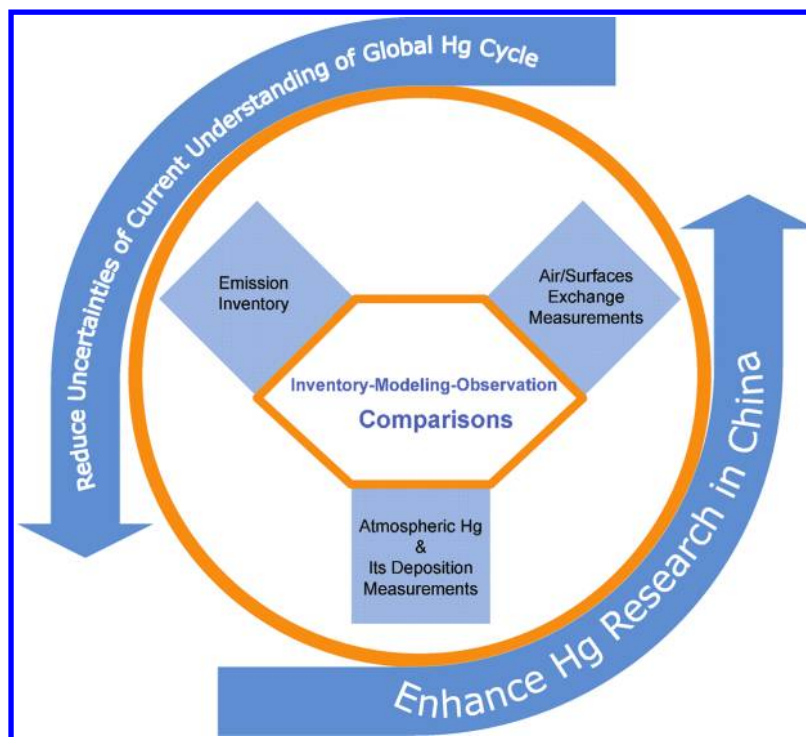


Figure 1. Enhancing the close coordinated Hg researches in China could significantly reduce the uncertainties of the current understanding of the global Hg cycle.

In the past decade, considerable efforts have been directed at estimating the man-made Hg emission from China. As the current best-available estimate, the emission data (2003 as the reference year) compiled by UNEP Global Partnership for Mercury Air Transport and Fate Research (UNEP-MFTP) shows that the Hg emission from China is 623 t.³ Some greater values are also reported by Wu et al. (700 t)¹⁶ and Pacyna et al. (800 t).¹⁸ Overall, the Hg emission from China accounts for ca. 30–40% of the global anthropogenic emission.³ Estimates for future global Hg emissions suggest that China's rising energy needs will continue to drive the increase of man-made Hg emission in the foreseeable future and the main driving force is the expansion of coal-fired electricity generation.^{18,19}

Since the Hg emission from China accounts for a large fraction in the global Hg emission, the uncertainty in China significantly influences the accuracy of the global Hg emission inventory. The atmospheric Hg measurements performed downwind of China²⁰ and modeling focused on East Asia^{21,22} also address the large discrepancy between field measurements, model results and inventories. These observations and modeling generally suggested that the Hg emission from China were underestimated in the original inventory.²³

By the extensive efforts from many researchers and agencies, the uncertainty level of anthropogenic Hg emission from China has diminished over time, but the paucity of some key data leads current inventories to remain large uncertainties.^{3,16} Considering several important factors (e.g., emission factor and activity level) that influence the estimation of emissions, Streets et al.²⁴ reported an uncertainty level of $\pm 44\%$ at the 95% confidence interval (or 696 ± 307 t) in the estimate of total anthropogenic Hg emissions of China in 2003. This uncertainty levels are clearly higher than those of global inventories (25–30%),³ as mentioned above.

As two leading man-made emission sources in China, the uncertainties of emission from the coal-fired power plants are

approximately 40–70% and those of other industrial sources are even higher, up to 450%.^{3,25} Various factors contribute to the uncertainties, including the Hg concentrations in coal and raw materials and Hg removal efficiency of pollution control devices.^{3,25} Therefore data sets of coal and raw material of Hg concentration covering large spatial extents and sectors are urgently needed. Recently, field test for Hg emission from the coal-fired power plant in China begins to receive attention.^{26–28} The limited studies suggest that Hg emission is mainly dependent on coal properties and air pollution control devices configuration.²⁸ But, tests for other industrial processes are scarce. For cutting atmospheric pollutant (such as SO₂, particulate matter, NO_x, etc.) emissions, China is accelerating the installation of air pollutant control devices, such as flue gas desulfurization, electrostatic precipitators and selective catalytic reduction systems in coal-fired power plants and industrial boilers.²⁹ However, the effect of these measures on cutting future Hg emission has not been fully evaluated.²⁸

Another paucity of data is on the Hg speciation profile in flue gases, which closely relates to the atmospheric behaviors of emitted Hg. Because the traditional test method for Hg speciation emission (e.g., Ontario Hydro Method) is laborious, advanced techniques, such as commercially available continuous emission monitoring for Hg speciation, should be introduced.

Overall, the survey of Hg concentrations in coal and raw material and field tests for emission in various industrial sectors should continue to expand. Field test for waste treatment and large Hg mining also should receive attention in the future. Ultimately, some important parameters (e.g., the Hg concentration in coal and raw material and the emission factors) used in current calculation of inventory for China, which mainly obtain from the technology performance in North America, Europe, and Japan, should update to the field data of China. Notably, the estimate of future trends of Hg emissions should carefully consider the dynamic of the energy transition and the

increasing implementation of pollution control measures in China.

Increase the Data set of Atmospheric Hg and Its Deposition. The most significant releases of Hg to the environment are to air, and the special property of Hg causes the atmosphere to be the most important medium for worldwide dispersion and transformation.³ Then understanding the Hg behavior in the atmosphere is crucial for investigating the global biogeochemical cycle of this toxin.³⁰

Asia has many developing countries, including the two largest, China and India, which are undergoing the rapid industrial development. The current emission inventory suggests that emissions from Asia represent about 50–60% of global anthropogenic Hg emissions.^{3,18} Particularly, recent studies have highlighted that the Hg emissions from China and India may increase dramatically in the future due to a sharp increase in energy production from coal combustion.^{18,19} However, much less information concerning the atmospheric Hg in Asia is available.

The measurements of atmospheric Hg and its deposition in China are quite limited (Figure 2).¹⁷ The selection of

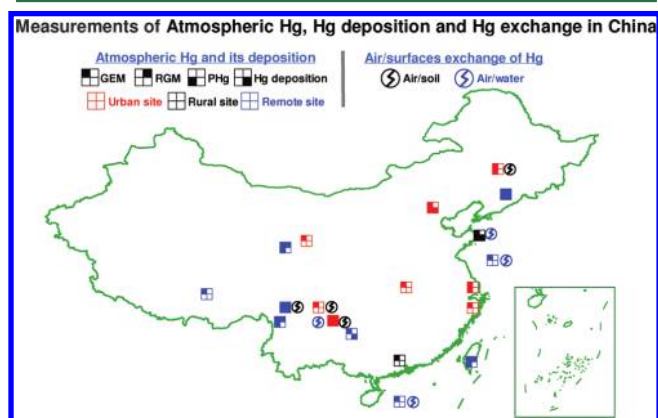


Figure 2. Map of China showing the sampling sites for atmospheric Hg research in China.

observation sites is also generally based on the researchers' interests rather than a systematic national network. In earlier studies, most field measurements were conducted at urban/suburban regions^{31–34} or mining/industrial regions.³⁵ Recently, more studies concerned the rural, remote, and oceanic regions.^{36–43} More recently, several studies were performed to investigate the atmospheric Hg speciation.^{44,45} This data has clearly shown the seriousness of atmospheric Hg pollution in China.^{15,17} The short-time measurements (<1 year) and the limited number of observation sites can not reveal any spatial and long-term trends of atmospheric Hg pollution.¹⁷

Considering the cost and instrumental availability and reliability, the priorities for future atmospheric Hg research in China are to observe GEM or total gaseous Hg (TGM = GEM + RGM) and weekly or monthly integrated wet total Hg (THg) deposition. These data sets could provide a basic state of atmospheric Hg pollution in China and initially satisfy the evaluation of the most current Hg models. The full range of measurements (including atmospheric Hg speciation, dry deposition, event-based wet deposition, gaseous Hg flux at the air/surfaces, other relevant air pollutants and meteorology) should be made in several carefully chosen sites. These intensive measurements will advance understanding of the

fundamental atmospheric Hg chemistry and provide detailed information needed to calibrate, test and develop regional and global Hg models. Some mountain-top sites in China, such as Waliguan station of Global Atmospheric Watch (3810 m above sea level, located on the Qinghai–Tibetan plateau), should receive additional attention because these sites have the potential to explore the Hg cycle in the free troposphere.⁴⁶ Recently, more aircraft measurements have been performed in China to investigate the atmospheric physics and chemistry (e.g., ref 47). If Hg measurement can be integrated into the future aircraft measurements,^{48,49} this could help understand the vertical Hg distribution in China/East Asia and further reveal the free–troposphere Hg chemistry.

Ideally, to provide the statistically significant temporal trends and spatial patterns of atmospheric Hg and its deposition, the national or regional monitoring network is the most appropriate (e.g., Mercury Deposition Network in the United States and Canada⁵⁰). For the large size and the variety of climate patterns and source regions, China also potentially needs a coordinated national network for atmospheric Hg monitoring. The evaluation of Hg models will significantly benefit from the network data. More importantly, only the network data can verify the attainment of the reduction goals and ensure coordinated implementation in future international Hg treaty. Notably, attention should be paid to challenges and problems of current atmospheric Hg measurements (especially for RGM and PHg) methods^{30,51,52} and the effective management of networks (e.g., the detailed measurement protocols, operator training, and thorough quality assessment and control).⁵⁰

At present, incorporating Hg monitoring into existing observation networks, such as the National Air Quality Network or the Acid Rain Monitoring Network in China, would be the most expeditious and efficient approach. The data of other air pollutants (SO₂, CO, NO_x, aerosol, etc.) and meteorological parameters monitored in the networks could be powerful to explore the emission, transportation, and transformation of atmospheric Hg. For regional atmospheric pollution, the countries in East Asia have successfully built a network for acid rain monitoring (Acid Deposition Monitoring Network in East Asia, EANET⁵³). This project has promoted the understanding of acid deposition issue in East Asia and provides useful inputs for decision making to prevent or reduce the adverse impacts of acid rain on the regional environment. As the largest emission source of Hg, regional cooperation related to atmospheric Hg monitoring in Asia and the active role of China in this cooperation are key to success. Recently, several stations in China (e.g., the Waliguan station and Zhuzhang station) had joined the Global Mercury Observation System (GMOS).⁵⁴ Furthermore, If possible, more intensive sites in China should join the coordinated regional or global Hg monitoring network because improving the understanding of atmospheric Hg cycle and global transport will require an integrated observation network covering a large spatial scale.

Determine Hg Flux between Air and Surfaces. Hg emission from the various surfaces includes the primary emission from geological Hg-enriched region, the regions impacted by direct Hg discharge (e.g., landfills, waste/tailings piles, sewage irrigation area, etc.) and the reemission from regions generally impacted by common atmospheric Hg deposition. Understanding the role of various ecosystems as source or sink of atmospheric Hg as well as the factors controlling its exchange is a basis of the global Hg cycle.^{3,55}

The lack of Hg flux data in China has influenced the accuracy of Hg models and the region/global Hg budget.^{55–57} For primary emission, several field studies regarding the geological Hg-enriched areas and mining areas in Southwest China^{55,58} were performed. The field data of other directly impacted regions (e.g., sewage irrigation area, landfill) is extremely sparse.⁵⁹ More studies are needed to understand the factors controlling Hg emission from contaminated areas and to further estimate its contribution to local and regional atmospheric Hg pool.

The reemission of Hg from background regions is spatially and temporally heterogeneous because this process is controlled by many factors, such as the Hg content, land-use types and climate.³ The role of various ecosystems of China in the Hg cycle has not received enough attentions in previous studies. Relative to the large size and the variety of ecosystems, the number of quantitative studies determining Hg flux at air and surfaces is rare in China (Figure 2).¹⁷ Only a few field studies have been performed to examine the Hg exchange between the air/soil^{60,61} and air/lake^{62,63} interfaces. Recently, the air/sea exchange of Hg downwind of China has received attention.^{40,43,64} But the field data relevant to other ecosystems is almost nonexistent.¹⁷ Although some model studies have estimated the Hg emission from natural sources (including vegetation, soil, and water surfaces) in East Asia, the accuracy of these estimates is difficult to evaluate due to the lack of field data.⁵⁶ Another noteworthy issue is that the high man-made atmospheric Hg emission and subsequent enhanced deposition flux could change the characteristics of air/surfaces Hg exchange in China, implying it may be different from other regions.

Therefore, greater focus is needed on the quantitative determination of Hg exchange between surfaces and air in China. This representative data will contribute to an in-depth understanding of Hg fate in regional and global scales, and also to improve the future model development.⁵⁷ Incorporation of Hg flux measurement into the established observation networks, such as ChinaFLUX (a network measuring the CO₂, H₂O and energy exchange between atmosphere and various ecosystems⁶⁵), would also be the most expeditious and efficient approach.

To date, the primary approach for investigating Hg flux between air and surfaces in China is the dynamic flux chambers (DFCs) method,^{60,61} because it is inexpensive, portable, easy to set up and operate. But the DFCs method has a series of inherent limitations. For example, the chamber isolates the surface from the atmospheric turbulence and rain/dew/snow, which changes the surface boundary layer condition and subsequently affects the mass transfer of Hg across the air and surfaces. The chamber also alters many other environmental conditions relating to Hg production/consumption and emission/deposition at the interface, such as solar radiation, surface temperature, humidity, and so on.⁶⁶ At present, there is not a standard design and operating protocol for DFCs, which limits the comparison of data from different works using various types of chamber designs and operating protocols.⁶⁷ More advanced approaches, such as micrometeorological methods and isotope techniques, should be introduced to overcome some limitations of the DFCs method and to further provide the insight of Hg air/surfaces exchange.

Enhance the Inventory–Modeling–Observation Comparisons. As a key component of the regional and global research framework, model studies and their results generally

are used to set public policy. Several widely used models for chemistry and transport of atmospheric species (e.g., GEOS–Chem, CMAQ, GRAHM, CTM) have been updated to include Hg.³ However, the lack of reliable spatially and temporally resolved Hg data and unclear atmospheric chemical transformation result in large uncertainties.³

As the largest and increasing source region of Hg, many model studies have focused on East Asia (mainly China) to understand the atmospheric Hg transport, transformation, deposition, (re)emission and outflow in this region.^{21,22,55,57} However, the accuracy of these models is difficult to examine owing to the lack of synchronous observations of atmospheric Hg. Therefore there is a need of greater coordinated modeling observation of atmospheric Hg. The emission inventory is also an important uncertainties source of model results.³ The integration of the updated anthropogenic Hg emission inventory into the modeling–observation studies also helps to examine and improve the accuracy of emission inventory. The large size and various types of sources region and climate in China provide a unique opportunity to improve the understanding of Hg cycle via the coordinated inventory–modeling–observation comparisons. These improvements will include (a) promoting the understanding of fundamental chemical mechanisms of atmospheric Hg, which is one of the major uncertainties in the current knowledge of Hg cycle; (b) optimizing the locations for short-term process sites and long-term observational sites; (c) assessing and improving the reliability of emission inventories and (d) understanding the role of China as source or sink for Hg on regional and global scales. Therefore, the close cooperation between the emission inventory scientists, field scientists, and modelers is a priority of the development of atmospheric Hg research program in the future.

■ SUMMARY

In summary, Hg is considered as a global pollutant because of its long-range cycling and persistence in the environment. For effectively addressing this global environmental issue, it is widely accepted that expanded and integrated science and policy efforts across local, regional and global scales are needed. Hg emission, transformation, and fate in Chinese environment are a critical part of the global Hg cycle. Owing to the rapid industrial development, anthropogenic Hg emissions in China may increase in the next few decades unless some drastic and effective measures are taken. In the context of the decreasing Hg emission from developed regions, China's role in the global Hg cycle is increasingly important. However the lack of high quality data in China leads to large uncertainties of current understanding of the global Hg issue and further affects the accuracy of global Hg assessment. This strongly impacts the subsequent development of an international Hg treaty. Enhancing the study of Hg in China, particularly the atmospheric process, not only helps China cope with the environmental challenge of this toxin but also reduces uncertainties of the understanding of global Hg cycle and further facilitates global Hg regulation. Although the Hg issue in China is attracting widespread attention and more research programs have focused on this issue, factors influencing the behavior, fate, and trend of Hg in Chinese environment are multitudinous and intertwined. Therefore, for more effectively and deeper understanding the Hg cycle in China, we need close coordinated efforts across field scientists, industrial departments, modelers, policy-makers, and other stakeholders from both domestic and international communities.

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Biography

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