## Response to the topical editor's comments:

We thank the topical editor for his comments. We have revised our manuscript according to his comments. In the following we would like to reply the comments point by point.

**Comments to the Author:**

I am pleased to accept your manuscript for publication subject to the following corrections:

1) please check all units in tables and figures. ug should be µg (micrograms, $\upmu$g) in text and on figures (e.g. fig 6, fig 8).

**Response:** Thanks for the comments. We have checked all units in the texts, tables and figures. All 'ug' has been revised to 'µg' in the revised manuscript.

2) the unit should be repeated in the caption for clarity.

**Response:** Thanks for the comments. We have revised the captions of all the tables and figures. The units now are repeated in the captions in the revised manuscript.

3) unit for deposition should be per unit time (eg Fig 9 and 12).

**Response:** Thanks for the comments. The unit for deposition has been revised to µg $m^{-2}$ $yr^{-1}$ or µg $m^{-2}$ $mon^{-1}$.

4) please enlarge all labels on fig 10.

**Response:** Thanks for the comments. All labels on Figure 10 have been enlarged in the revised manuscript.

The Revised Manuscript

# GNAQPMS-Hg v1.0, a global nested atmospheric mercury transport model: Model description, evaluation and application to trans-boundary transport of Chinese anthropogenic emissions

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# Abstract

Atmospheric mercury (Hg) is a toxic pollutant and can be transported over the whole globe due to its long lifetime in the atmosphere. For the purpose of assessing Hg hemispheric transport and better characterizing regional Hg pollution, a global nested atmospheric Hg transport model (GNAQPMS-Hg) has been developed. In GNAQPMS-Hg, the gas and aqueous phase Hg chemistry representing the transformation among three forms of Hg: elemental mercury (Hg(0)), divalent mercury (Hg(II)), and primary particulate mercury (Hg(P)) are calculated. A detailed description of the model, including mercury emissions, gas and aqueous phase chemistry, and dry and wet deposition is given in this study. Worldwide observations including extensive data in China have been collected for model evaluation. Comparison results show that the model reasonably simulates the global mercury budget and the spatial-temporal variation of surface mercury concentrations and deposition. Overall, model predictions of annual total gaseous mercury (TGM) and wet deposition agree with observations within a factor of two, and within a factor of five for oxidized mercury and dry deposition. The model performs significantly better in North America and Europe than in East Asia. This can probably be attributed to the large uncertainties in emission inventories, coarse model resolution and to the inconsistency between the simulation and observation periods in East Asia. Compared to the global simulation, the nested simulation shows improved skill at capturing the high spatial variability of surface Hg concentrations and deposition over East Asia. In particular, the root mean square error (RMSE) of simulated Hg wet deposition over East Asia is reduced by 24% in the nested simulation. Model sensitivity studies indicate that Chinese primary anthropogenic emissions account for 30% and 62% of surface mercury concentrations and deposition over China, respectively. Along the rim of the western Pacific, the contributions from Chinese sources are 11% and 15.2% over the Korean Peninsula, 10.4% and 8.2% over Southeast Asia, and 5.7% and 5.9% over Japan. But for North America, Europe and West Asia, the contributions from China are all below 5%.

# 1 Introduction

Since the Minamata Event in Japan in the 1960s (Harada, 1995), the toxicity of mercury (Hg) on human health and the environment has caused widespread public concern. Hg is a persistent, bio-accumulated pollutant, and the only heavy metal that can be transported globally in gaseous form (Schroeder and Munthe, 1998). As a result, Hg has been listed as a priority pollutant by many countries and international agencies. After a long struggle, the first global treaty (the Minamata Convention) aimed at reducing Hg emissions and releases, was adopted and signed by 92 countries in 2013 (http://www.mercuryconvention.org/). This made an important advance towards joint action to control global Hg pollution and has brought higher requirements for understanding global Hg source-receptor relationships, especially the impacts of high regional emissions (e.g. from China and India) on global Hg levels. However, besides the remaining uncertainties in emission estimates, poor understanding of the chemical transformation of atmospheric mercury has made assessment of long-range transport very challenging (AMAP/UNEP, 2013).

Atmospheric mercury models are powerful tools to assess the fate and transport of mercury in the atmosphere. A number of atmospheric mercury models have been developed to investigate the emissions, transport, chemistry, deposition and source-receptor relationships of Hg at global and regional scales. Global models include the GEOS-Chem model (Amos et al., 2012; Zhang et al., 2012), the CTM-Hg model (Seigneur et al., 2004), the CAM-Chem-Hg model (Lei et al., 2013), the ECHMERIT model (De Simone et al., 2014), the MSCE-Hg-Hem model (Travnikov and Ilyin, 2009), the DEHM model (Christensen et al., 2004), and the GRAHM model (Dastoor and Durnford, 2014). Regional models include the CMAQ-Hg model (Bash, 2010), the STEM-Hg model (Pan et al., 2008), the CAMx-Hg model (ENVIRON, 2011) and the WRF-Chem-Hg model (Gencarelli et al., 2014). Application of these models has greatly advanced our understanding of the global Hg cycle. However, several model intercomparison studies (Ryaboshapko et al., 2007; Bullock et al., 2008; Pirrone and Keating, 2010) have found that large uncertainties still exist in Hg models and there is much room for improvement, especially for simulation of reactive gaseous mercury (RGM) and dry deposition.

Mercury is released to the atmosphere from both anthropogenic and natural sources. Human activities have increased the amount of mercury cycling through the atmosphere-ocean-terrestrial system by about a factor of three (Selin, 2009), although anthropogenic sources are estimated to account for only 31% of total Hg emissions (Pirrone et al., 2010). China has the world's largest Hg production, consumption and emissions, and suffers the most serious Hg pollution (Jiang et al., 2006), but the impacts of its anthropogenic emissions on global Hg levels are still unclear. Previous modeling studies mainly focused on long-range transport of mercury from Asia. Based on the GEOS-Chem model, about 7-20% of Hg deposition over the United States (US) was found to originate from Asian anthropogenic sources, which was comparable to that from North American sources (Strode et al., 2008; Jaffe and Strode, 2008). Another modeling study using the CTM-Hg model with three emission scenarios indicated that Asian anthropogenic emissions accounted for 14–25% of Hg deposition over the US (Seigneur et al., 2004). Travnikov (2005) reported a contribution to Hg deposition from total Asian sources (including both anthropogenic and natural emissions) of 15% over Europe and 33% over the Artic. Corbitt et al. (2011) further pointed out that Asian emissions are the largest contributors to anthropogenic deposition to all ocean basins and these contributions are expected to further grow in the future. The above studies all treated Asian anthropogenic emissions as a whole, and the effects of anthropogenic emissions from the world’s largest single emitter (China) have not been explicitly assessed before. In addition, due to lack of observational data, little model validation has been conducted over East Asia (especially China) in these studies and this leads to greater uncertainty in the conclusions. Fu et al. (2012) reviewed previous modeling studies and pointed out that current model simulations tend to underestimate total gaseous mercury (TGM) and total particulate mercury (TPM) concentrations but overestimate reactive gaseous mercury (RGM) concentrations in China. To improve Hg model skill in China, nested simulations with high horizontal resolution might be a good choice. Zhang et al. (2012)

demonstrated that a nested-grid model can capture the variation of Hg wet deposition over North America better than a global model. In this study, online nested Hg simulation with flexible horizontal resolution was developed and evaluated. Compared to traditional multi-scale modeling approach (using a global model to provide initial and boundary conditions to a regional model) (Seigneur et al., 2001), online nested method use the same physical and chemical parameterizations in the global and nested domains which could avoid uncertainties induced by different boundary conditions. Compared to offline nested method used in the GEOS-Chem model (Zhang et al., 2012), online nested method can provide boundary conditions with higher time resolution (10 or 5 minutes) from the global domain to the nested domain. Hence, online nested simulation would potentially improve model performance in regional scale.

Therefore, a comprehensive evaluation and improvement of Hg model performance in China is needed to effectively reduce the uncertainties in Hg trans-boundary transport and a quantitative assessment of Chinese anthropogenic contribution to global Hg concentration and deposition levels is helpful to determine and fulfill the Hg emission reduction tasks under the Minamata Convention.

In this paper, we describe the development of a global nested atmospheric mercury transport model (GNAQPMS-Hg) incorporating the latest available physical and chemical processes essential to the mercury life cycle. The spatial and temporal variability of Hg concentrations and deposition are comprehensively evaluated against available worldwide observations, including extensive data from China. The impact of horizontal resolution (1°x1° in the global domain versus 0.33°x0.33° in the nested domain) on model predictions over East Asia is examined. Finally, the trans-boundary transport of Chinese primary anthropogenic Hg emissions is quantified using the model.

# 2 Model description and setup

## 2.1 General description

The atmospheric physics and chemistry component of GNAQPMS-Hg, with the exception of the mercury module, is based on the Nested Air Quality Prediction Modeling System (NAQPMS) (Wang et al., 2006), developed at the Institute of Atmospheric Physics, Chinese Academy of Sciences. NAQPMS is a 3-D regional Eulerian model which has been rigorously evaluated and widely applied to simulate the chemical evolution and transport of ozone (Li et al., 2007; Tang et al., 2010), the distribution and evolution of aerosol and acid rain over East Asia (Wang et al., 2002; Li et al., 2011; Li et al., 2012) and to provide operational air quality forecasts in mega cities such as Beijing, Shanghai and Guangzhou (Wang et al., 2010; Wu et al., 2012; Wang et al., 2009). GNAQPMS is the global version of NAQPMS and uses the same model framework and physical and chemical parameterization schemes.

As a multi-scale model, GNAQPMS can simulate the transportation and formation of primary and secondary pollutants from urban to global scale using an online nesting approach. It includes advection, diffusion and convection processes, gas/aqueous/aerosol chemistry, and modules for dry and wet deposition. The advection process is parameterized based on an accurate mass conservative, peak-preserving algorithm provided by Walcek and Aleksic (1998). The gas phase chemical mechanism is the CBM-Z mechanism (Zaveri and Peters, 1999), including 133 reactions for 53 species. The dry deposition module uses the parameterization of Wesely (1989). The wet deposition and aqueous-phase chemistry module is constructed based on a revised version of the RADM mechanism (Chang et al., 1987; Wang et al., 2002; Ge et al., 2014). A mercury module has been developed and coupled into the GNAQPMS model in this study, as described in Sect. 2.2 to 2.4. Hereafter, we call this new model GNAQPMS-Hg. Note that meteorology, emissions, deposition and chemistry are self-consistent between the global and nested domains.

## 2.2 Mercury chemistry

### 2.2.1 Basic mechanism

In GNAQPMS-Hg, three forms of mercury are explicitly treated: elemental mercury (Hg(0)), divalent mercury (Hg(II)), and primary particulate mercury (Hg(P)). Transformations between these three forms include the gas phase oxidation of Hg(0) to Hg(II), the aqueous phase oxidation of Hg(0) to Hg(II), the aqueous phase reduction of Hg(II) to Hg(0), the aqueous phase equilibria of Hg(II) species and the aqueous phase adsorption of Hg(II) to PM. Fig. 1 depicts the mercury reaction pathways both in the gas and aqueous phase while the detailed reactions and their rate constants are summarized in Table 1. In line with most global mercury models, GNAQPMS-Hg does not include dynamic air-surface exchange during Mercury Depletion Events (MDEs) in Polar regions (Schroeder et al., 1998) due to lack of fundamental data.

In the gas phase, Hg(0) is oxidized to Hg(II) by $O_3$, OH, hydrogen peroxide ($H_2O_2$), hydrogen chloride (HCl) and molecular chlorine ($Cl_2$). The oxidized products of these five reactions are assumed to be in the gas phase. According to Lin et al. (2004), OH and $O_3$ are the dominant oxidants in the continental troposphere while Cl and Br dominate Hg(0) oxidation in the marine boundary layer and the upper troposphere. In the aqueous phase, Hg(0) is oxidized to Hg(II) by dissolved $O_3$, OH, and $Cl_2$, and Hg(II) can be reduced back to Hg(0) via reaction with $HO_2$ and by the formation of sulfite complexes. In addition, adsorption of Hg(II) species on atmospheric particulate matter (PM) is simulated using an adsorption coefficient ($K = 34\ L\ g^{-1}$) recommended by Seigneur et al. (1998).

As shown in Table 1, the mercury chemistry requires the concentrations of several non-mercury species, among which $O_3$, OH, $HO_2$, $H_2O_2$, $SO_2$, HCl and PM are simulated online with GNAQPMS-Hg. However, $Cl_2$ is not explicitly simulated, and a typical vertical profile of $Cl_2$ concentrations is therefore prescribed. The $Cl_2$ concentrations are specified to be 100 ppt at the surface, 50 ppt aloft at night, 10 ppt during daytime over the oceans, and zero over land (Seigneur et al., 2001).

### 2.2.2 Bromine oxidation

In order to test the effect of bromine (Br) oxidation reactions on global Hg concentrations, five Br chemical reactions in the gas phase are added in addition to the $O_3$-OH oxidation mechanism. The detailed description of the Br chemical reactions and their implementation in the model is shown in Section S1.1 in the supplement. A model sensitivity experiment with additional Br oxidation reactions was conducted and compared to the base case simulation with $O_3$-OH oxidation mechanism. Fig. S1 in the supplement shows the difference of surface TGM concentrations resulting from introducing Br oxidation reactions. Decrease in TGM concentrations is found in the whole globe. This is because additional Br chemistry transforms more Hg(0) into Hg(II), which subsequently enhances the deposition of Hg(II), leading to the reduction of TGM concentrations. Larger TGM reduction is found in the Northern Hemisphere than in the Southern Hemisphere. In general, the change in TGM concentration is less than 0.2 ng $m^{-3}$ in most areas which indicates that introducing Br chemistry seems to have little impact on overall TGM magnitudes and patterns. These results are similar to Lei et al. (2013) which test the impact of Br chemistry using the CAM-Chem-Hg model. Although adding the Br chemistry does not significantly change the TGM pattern, but it may affect the gaseous Hg partitioning between Hg(0) and Hg(II), and hence may affect the global Hg deposition patterns. More in-depth tests and analysis are needed to address these impacts in the future. In the following sections, we still use the base case simulated results without considering the possible effects of Br chemistry.

### 2.2.3 Gas-particle partitioning of Hg(II)

Recent studies suggested that gas-particle partitioning of Hg(II) is an important process affected global Hg concentrations and deposition (Amos et al., 2012). To test these effects, an empirical mechanism of gas-particle partitioning of Hg(II) was added to the GNAQPMS-Hg model. The detailed description of this mechanism and its implementation in the model is shown in Section S1.2 in the supplement. A model sensitivity experiment with Hg(II) gas-particle partitioning module was conducted and compared to the base case simulation with all Hg(II) existing in the gas phase. Figs. S2-3 in the supplement show the change fraction of surface TGM concentrations and oxidized Hg concentrations resulting from introducing the Hg(II) gas-particle partitioning mechanism. As expected, TGM concentrations decrease while oxidized Hg concentrations increase in the whole globe. The change fractions of TGM and oxidized Hg concentrations are smaller than 0.1 over the middle latitude of the Northern Hemisphere, and even smaller than 0.05 over China. Considering that the base case simulation has overestimated oxidized Hg concentrations in most areas, introducing the mechanism of gas-particle partitioning of Hg(II) would further increase this model discrepancy. Therefore, we still use the base case simulated results without considering the possible effects of gas-particle partitioning of Hg(II) in the following sections.

## 2.3 Mercury deposition

Deposition is the leading removal process of atmospheric mercury, and also a major cause of mercury contamination in soil and water. Studies have shown that both dry and wet removal pathways are equally significant for the total deposition of mercury (Pirrone and Keating, 2010; Lin et al., 2006).

Dry deposition of Hg(0), Hg(II) and Hg(P) is accounted for in the GNAQPMS-Hg model, and simulated with the Wesely (1989) resistance model, which considers the effect of different land cover types and characterizes the diurnal variation of dry deposition velocities. The Henry's Law constant for Hg(0) is set to be 0.11 M atm$^{-1}$ (Lin and Pehkonen, 1999) with a temperature factor of -4970 K (Clever et al., 1985), and the surface reactivity is set to zero. Hg(II) represents $HgCl_2$ and $Hg(OH)_2$. Its Henry's Law constant is assumed to be the same as $HNO_3$ because they have similar solubility (Bullock and Brehme, 2002). Like $HNO_3$, Hg(II) has a strong tendency to stick to surfaces and its dry deposition occurs readily, so the surface resistance for Hg(II) in the dry deposition scheme is set to zero. The Hg(P) dry deposition velocity is set equal to that for sulfate, similar to that applied in the CMAQ-Hg and

STEM-Hg model (Bullock and Brehme, 2002; Pan et al., 2008). More detailed description of the dry deposition scheme used in the model is given in Section S2.1 in the supplement. Model intercomparison studies demonstrate that there are still very large uncertainties in Hg dry deposition estimates (Bullock et al., 2008), and this can be ascribed to the wide range of treatments and physical parameters for dry deposition used in different models.

The wet deposition of Hg includes in-cloud and below-cloud scavenging. In-cloud scavenging is dependent on cloud and rain water content, species solubility and chemical transformation in the liquid phase, while below-cloud scavenging depends mainly on total rainfall intensity and washout efficiency. Among the three forms of mercury, wet deposition of Hg(0) is minor compared to Hg(II) and Hg(P) due to its low solubility. Therefore, Hg(0) oxidation will enhance total Hg wet deposition. In the GNAQPMS-Hg model, wet deposition of Hg species is calculated through adapting the RADM mechanism. The physical properties (e.g. Henry's Law constant, surface reactivity, molecular diffusivity) used are the same as those in the dry deposition module. More detailed description of the wet deposition scheme used in the model is given in Section S2.2 in the supplement. Currently, the uncertainties of Hg wet deposition simulation are mainly from the assumptions made in the cloud scavenging process and the uncertainty associated with the precipitation fields (Seigneur et al., 2001; Lin et al., 2006).

## 2.4 Mercury emissions

We include anthropogenic emissions, biomass burning emissions, geogenic emissions, land reemission and ocean emissions (including reemission) of Hg in the model. Emissions from artisanal mining and volcanoes are neglected due to lack of fundamental data. The former is estimated to be 400 Mg yr$^{-1}$, and the latter 90 Mg yr$^{-1}$, and they account for about 5% and 1% of global total Hg emissions (Pirrone et al., 2010). Note that biomass burning emissions, geogenic emissions, land and ocean emissions are all treated as Hg(0). Global Hg emissions in the model are compared to previous studies in Table 2, and their spatial distributions are given in Figs. S4-S6 in the supplement.

Anthropogenic emissions in 2000 are derived from the Arctic Monitoring and Assessment Programme (AMAP) inventory (Pacyna et al., 2006; Wilson et al., 2006). This inventory has a horizontal resolution of 0.5°x0.5° and no seasonal variation. Following Selin et al. (2008), we increase the Asian (0～60$^0$N，65～150$^0$E) Hg(0) emissions in the AMAP inventory by 50% (about 300 Mg yr$^{-1}$) to account for the regional underestimation identified by Jaffe et al. (2005). The modified inventory has a total emission of 2488 Mg yr$^{-1}$, with Hg(0), Hg(II) and Hg(P) accounting for 63%, 29% and 8% respectively. The major source regions are Asia and Africa, accounting for 59% (1480 Mg yr$^{-1}$) and 16% (399 Mg yr$^{-1}$), while Europe and North America contribute only 7% and 6%. China has the largest emissions at country level (about 785 Mg yr$^{-1}$), contributing 53% and 32% to the Asian and global anthropogenic Hg emissions, respectively. It is noted that the emissions over South Africa in this inventory were reported to be flawed (AMAP/UNEP, 2008) and much higher than reality. The effects of these flawed emissions on the simulated results were assessed in Section S3 in the supplement.

Biomass burning emissions are specified by mapping an annual mean value of 675 Mg yr$^{-1}$ (Friedli et al., 2009) to the spatial and temporal distribution of CO biomass burning emissions from the IPCC-AR5 (Intergovernmental Panel on Climate Change Fifth Assessment Report) emissions inventory (Lamarque et al., 2010). The regional and monthly emission amounts are prescribed based on Friedli et al. (2009). A similar method has been used by Jung et al. (2009).

The geogenic emissions here represent mobilization of Hg by degassing from geological reservoirs. Following Selin et al. (2007), we consider a geogenic source of 500 Mg yr$^{-1}$ distributed according to the locations of Hg mines (Frank, 1999) as an indicator of Hg deposits. No temporal variation is applied to the geogenic emissions.

Land and ocean emissions are not dynamically calculated in the model due to the large uncertainties associated with current parameterizations. Consistent with several previous studies (Selin et al., 2007; Seigneur et al., 2001), the global annual land reemission of Hg is assumed to be 1500 Mg yr$^{-1}$. The biogenic CO emissions from the Global Emission InitiAtive (GEIA) inventory (Guenther et al., 2006) are used as spatial and temporal surrogates to map the land reemission. Regional emission totals from different latitude zones and land uses are prescribed based on Mason (2009).

Ocean emissions in our model are specified as 5000 Mg yr$^{-1}$ (including reemission), close to the estimates of Selin et al. (2008). Similarly, ocean emissions are mapped according to the distribution of ocean biogenic CO emissions from the Precursors of Ozone and their Effects in the Troposphere (POET) inventory (Granier et al., 2005). Additionally, ocean emissions are adjusted to reflect several distribution characteristics: 1) ocean emissions are high in summer but low in winter (Strode et al., 2007), 2) ocean emissions are largest in the Tropics and downwind of industrial regions (Strode et al., 2007; Soerensen et al., 2010b), and 3) ocean emissions are large at mid and high latitudes in the Southern Hemisphere due to high wind speeds (Selin et al., 2008).

### 2.5 Model setup

Two nested domains covering the whole globe and East Asia are configured in this study. The horizontal resolutions are 1°x1° and 0.33°x0.33°, respectively. Vertically, the model uses 20 terrain-following layers from the surface to 20 km a.s.l., with a decreasing resolution with height. Roughly, the lowest 14-18 layers are in the troposphere and the remaining layers are in the stratosphere. The time step in the model calculation is 600 s. The input/output frequency is 6h in the global domain but 3h in the nested domain. The meteorological fields are provided by the global version of the Weather Research and Forecasting (WRF) model. The atmospheric lifetime of Hg(0) is 0.5-2 year (Schroeder and Munthe, 1998), and so to ensure mixing through the global troposphere and approach steady-state, we conduct the simulation for a 4-year period, with the first 3 years used for initialization and the last year (2001) used for analyses.

Emissions of reactive gases and aerosols used in this study are from several databases: 1) the IPCC-AR5 anthropogenic and biomass burning emissions for 2000 (Lamarque et al., 2010); 2) the GEIA biogenic emissions for 2000 (Guenther et al.,

2006) and lightning emissions of nitric oxide ($NO_x$) for 1983–1990 (Price et al., 1997); 3) the POET ocean emissions of volatile organic compounds (VOCs) for 2000 (Granier et al., 2005); 4) the soil $NO_x$ emissions for 2001 from Yan et al. (2005). All emissions are interpolated and remapped to match the model grids of the global and nested domains. The initial and top boundary conditions for $O_3$, $NO_x$, and CO are taken from a global chemical transport model (MOZART-V2.4) with 2.8° resolution (Horowitz et al., 2003).

Two model simulations, with and without Chinese primary anthropogenic Hg emissions, are carried out in this study. The differences between the two simulations are attributed to the influence of Chinese primary anthropogenic Hg emissions.

# 3 Model evaluation

## 3.1 Observational data

Compared to reactive gases and aerosols, atmospheric Hg measurements are still quite sparse. Routine monitoring networks for atmospheric Hg concentrations and deposition have only been established in Europe and North America. Lack of Hg observational data is a great restriction against advancing our understanding of global Hg cycling and improving our skill in modeling. There is an urgent need to establish a coordinated global Hg monitoring network for current Hg study (Sprovieri et al., 2010; Keeler et al., 2009).

The observational dataset in this study is based partly on the database shared by the GEOS-Chem Hg modeling group (public access athttps://github.com/noelleselin/HgBenchmark; Selin et al., 2007; Selin et al., 2008; Holmes et al., 2010). This is supplemented with scattered Hg observations across East Asia collected from the literature. The observations used in this study are summarized as follows: 1) long-term TGM/GEM (gaseous elemental mercury) measurements at 51 land sites, with 49 in the Northern Hemisphere and 2 in the Southern Hemisphere; 2) long-term RGM/TPM measurements at 26 land sites, all in the Northern Hemisphere; 3) short-term Hg species measurements from 6 ship cruises; 4) wet deposition measurements from the MDN (the Mercury Deposition Network in North America, http://nadp.sws.uiuc.edu/nadpdata/mdnalldata.asp) and EMEP (the European Monitoring and Evaluation Programme, http://www.nilu.no/projects/ccc/emepdata.html) monitoring networks, with 51 and 8 sites respectively; 5) dry and wet deposition measurements at 19 sites in East Asia. Further information about the measurement sites and data sources is given in Tables S2-S5 in the supplement. It should be noted that the time periods of the measurements do not all match with those of the simulation, and this difference may partially explain any model–observation discrepancies. The influence of the mismatch of time periods when comparing the simulated results with the observations was qualitatively analyzed and shown in Section S4.1 in the supplement.

### 3.2 Global mercury budget

Fig. 2 gives the global mercury budget in GNAQPMS-Hg, including the cycling among atmosphere, ocean and land. The total atmospheric burden of Hg is 5546 Mg, with Hg(0), Hg(II), and Hg(P) contributing 90%, 9% and 1%, respectively. Therefore, mercury in the atmosphere exists mainly as Hg(0). Total emissions and deposition of Hg are 5163 Mg yr$^{-1}$ and 2866 Mg yr$^{-1}$ over land (a net source), and are 5000 Mg yr$^{-1}$ and 7297 Mg yr$^{-1}$ over ocean (a net sink), indicating that Hg is transported from land to ocean. For total deposition of Hg species, Hg(0) and Hg(II)/Hg(P) account for 38% and 62% over the earth's surface. Over land, deposition of Hg(II)/Hg(P) is more prominent than that of Hg(0), while they are both important over the ocean. Our results for total Hg deposition over ocean and Hg(II)/Hg(P) deposition over land are very close to that of GEOS-Chem (Selin et al., 2008). However, Hg(0) deposition over land derived from GNAQPMS-Hg is much smaller. This may be due to the lower reactivity coefficient used in the dry deposition module in GNAQPMS-Hg (zero in GNAQPMS-Hg but $10^{-5}$ in GEOS-Chem), which produces a lower dry deposition velocity for Hg(0).

Table 2 compares the GNAQPMS-Hg TGM budget and lifetime to those from previous modeling studies. The TGM sources, sinks, burden and lifetime estimated from GNAQPMS-Hg are all in the range determined by previous studies. Taking the TGM lifetime as an example, the reported range is 0.5-1.7 years and it is 0.54 years for GNAPQMS-Hg. In addition, similar to the results of GEOS-Chem (Selin et al., 2007) and CAM-Chem-Hg (Lei et al., 2013), Hg dry deposition in GNAQPMS-Hg dominates globally over wet deposition. Dry and wet deposition account for 78% and 22%, respectively.

### 3.3 Total gaseous mercury (TGM)

As shown in Fig. 3, the main characteristics of the spatial distribution of TGM are captured well by the model. High surface TGM concentrations are found in or downwind of areas with intensive mercury-relative mining (e.g. Western USA) and rapid industrialization (e.g. East Asia). In particular, TGM concentrations even exceed 3 ng m$^{-3}$ in eastern China. Both model simulation and observations show a significant surface interhemispheric gradient in TGM (Fig. 3 and Fig. 4). Based on background observations, Lindberg et al. (2007) reported that mean Hg(0) concentrations were 1.5–1.7 ng m$^{-3}$ in the Northern Hemisphere and 1.1–1.7 ng m$^{-3}$ in the Southern Hemisphere. Lamborg et al. (2002) also estimated the range of north-south interhemispheric TGM concentration ratios for surface air as 1.2–1.8. Our model results share a general similarity with these studies. In GNAQPMS-Hg, surface mean TGM concentrations in the Northern and Southern Hemisphere are 1.56 and 1.23 ng m$^{-3}$, and the derived interhemispheric ratio is 1.27. However, it should be noted that GNAQPMS-Hg is systematically biased low relative to cruise observations in the Northern Hemisphere, which leads to underestimation of the TGM interhemispheric ratio compared with the range (1.49±0.12) reported by Temme et al. (2003) based on observations from several Atlantic cruises. This disagreement was also found by several previous modeling studies (Seigneur et al., 2004; Selin et al., 2007), and can be attribute to the inability of current models to reproduce the air-sea exchange of Hg reasonably (Soerensen et al., 2010a). More specifically, this discrepancy is due to upwelling mercury from the sub-surface ocean, possibly reflecting the legacy of past anthropogenic emissions (Holmes et al., 2010), and has been partially demonstrated by Soerensen et al. (2012). In general, the simulated TGM concentrations match observations within a factor of two (Fig. 10). The correlation coefficient (R) and normalized mean bias (NMB) between model results and observations from 51 land sites are 0.7 and -18%, respectively (Table 3).

Fig. **5** illustrates the mean seasonal variations of surface TGM concentrations in North America, Europe, East Asia, the Arctic, the Antarctic (Neumayer) and South Africa (Cape Point). In northern mid-latitudes, TGM concentrations are high in winter but low in summer. This seasonality can be reproduced well by GNAQPMS-Hg. The summer low is caused by high OH concentrations and frequent precipitation (Bergan and Rodhe, 2001). Compared with observations, the simulated TGM monthly variations are stronger in North America but weaker in East Asia. The site by site comparisons in East Asia are shown in Fig. S9 in the supplement. We can see that nested simulation can well improve model performance in simulated TGM monthly variation in East Asia. At Arctic and Antarctic sites, TGM shows a spring minimum driven by MDEs and a summer maximum driven by reemission from the snowpack (Steffen et al., 2005). The summer maximum is captured by GNAQPMS-Hg because high reemission in polar summer has been taken into account in our land reemission inventories. However, due to missing halogen chemistry, the model fails to reproduce the spring minimum. At Cape Point, both observed and simulated TGM show little seasonal variation. However, simulated monthly TGM concentrations are systematically biased high (NMB is 87%), which can be attributed to the flawed anthropogenic emissions in the AMAP emission inventories over South Africa (AMAP/UNEP, 2008). By updating the anthropogenic emissions over South Africa, the simulated TGM concentrations at Cape Point decrease from 1.77 ng m$^{-3}$ to 1.23 ng m$^{-3}$, more close to the observed values (See Section S3 in the supplement).

Additional evaluation and analyses of simulated diurnal and vertical variation of TGM concentrations are given in Section S4.2 in the supplement.

## 3.4 Oxidized mercury

Fig. 3 also shows the global distribution of oxidized mercury (defined as the sum of RGM+TPM in the observations and Hg(II)+Hg(P) in the model). Similar to TGM, a pronounced north-south interhemispheric gradient is found for surface concentrations of oxidized mercury, which is consistent with the global distribution of emissions. Both model simulation and observations indicate that oxidized mercury concentrations are much higher in East Asia than North America and Europe. Compared to scarce available observations, oxidized mercury concentrations are overestimated by GNAQPMS-Hg in most parts of the world (except East Asia). This discrepancy may partially be attributed to excessive oxidation of Hg(0) by relatively high concentrations of OH and $O_3$ (especially over the ocean) and uncertainties concerning Hg chemical speciation in emission inventories. The simulated tropospheric mean OH concentration is $1.41\times10^{6}$ molec $cm^{-3}$. This is at the high end of the concentration range ($0.65-1.56\times10^{6}$ molec $cm^{-3}$) summarized by Lawrence et al. (2001) and is about 27% higher than the ensemble mean ($11.1\pm1.8\times10^{5}$ molec $cm^{-3}$) of the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) models (Voulgarakis et al., 2013). The simulated mean surface $O_3$ in the North Pacific and North Atlantic is overestimated by 27% and 34% compared to observations from the WDCGG (World Data Centre for Greenhouse Gases) network, although concentrations over land are reproduced relatively well (see Figs. S10-S11 in the supplement). Besides, uncertainties of Hg chemistry (e.g. gas-particle partitioning of RGM, in-plume reduction of RGM) and deposition processes in the present model might also contribute to this discrepancy. Overall, the simulated oxidized mercury concentrations agree with observations within a factor of five (Fig. 10). The statistical indicators, R and NMB, calculated from 26 land sites are 0.53 and 3% (Table 3), respectively.

## 3.5 Wet deposition

Wet deposition is mainly determined by the distribution of precipitation and Hg concentrations. Fig. 6 and Fig. 7 evaluate the simulated annual Hg wet deposition and accumulated precipitation over North America, Europe and East Asia. In general, GNAQPMS-Hg reproduces the spatial patterns of Hg wet deposition relatively well.

Over North America, the maximum wet deposition occurs in the southeast, corresponding to high OH concentrations and frequent precipitation there, while less wet deposition occurs in the west and north, where there is much less precipitation. GNAQPMS-Hg predicts the magnitude of mean wet deposition within 5% and shows a good spatial correlation (R=0.76) (Table 3). These results are similar to those of GEOS-Chem (Selin et al., 2007). However, it should be also noted that precipitation in the southeast is slightly overestimated by the model.

Over Europe, model performance for wet deposition and precipitation are better than over North America and East Asia. High spatial correlation between the simulated and observed results are found for both wet deposition (R=0.78) and precipitation (R=0.86), and the NMBs are both less than 5% (Table 3).

Over East Asia, Hg wet deposition is not only related to the precipitation pattern but also the local Hg emissions, especially in the southwest and Jilin province of China, and in Central Japan. Model performance for wet deposition over East Asia is poorer than over Europe and North America. Although the spatial distribution and magnitude of precipitation over East Asia are seemingly well reproduced (R=0.64 and NMB=-6%), a large underestimation (NMB=-61%) of wet deposition is found here. Specifically, this is because the model fails to capture the high wet deposition at certain sites. For example, the observed wet deposition over Shanghai and Changchun are 251 and 108 µ~~u~~g m$^{-2}$ yr$^{-1}$ while the corresponding simulated values are only 25 and 13 µg m$^{-2}$ yr$^{-1}$ ~~ug m$^{-2}$~~. This suggests that it is hard for models with coarse horizontal resolution to characterize the high local mercury pollution in China. The difference between the simulated and observed time periods and uncertainties in the emission inventories may also contribute to these discrepancies.

Fig. 8 further compares the simulated seasonal cycle of wet deposition with measurements at MDN sites over North America and EMEP sites over Europe. No monthly wet deposition observations are available over East Asia. Wet deposition and precipitation share similar monthly variations, with high values in summer and autumn and low values in winter, as shown by both observations and simulation. In summer and autumn, the variation in wet deposition and precipitation among sites is larger than for other seasons, and this is evident from the greater variability in Fig. 8. GNAQPMS-Hg tends to overestimate wet deposition and precipitation in July and August over North America.

### 3.6 Dry deposition

Due to limited observations, only Hg dry deposition over East Asia is evaluated in this study. It should be noted that data (Table S5 in the supplement) used to evaluate model simulation of dry deposition is not directly measured, but is inferred or estimated based on measurements of total Hg in through fall and rainwater, wet deposition and atmospheric concentrations. Associated with local Hg emissions, high dry deposition mainly occurs over central eastern China and central Japan (Fig. 9). The modeled dry deposition has a good spatial correlation with observations (r=0.81), but there is a substantial negative bias (NMB=-42%, Table 3). In general, the simulated dry deposition agrees with observations within a factor of five (Fig. 10). Over Japan, the model results are biased high by a factor of 2-5, which may be caused by overestimation of Hg(II) and Hg(P) emissions and missing model mechanism to deal with fast in-plume reduction of Hg(II) (Vijayaraghavan et al., 2008; Amos et al., 2012; Zhang et al., 2012). Taking Tokyo as an example, observed Hg(P) is only 98 pg m$^{-3}$ while the simulated value is as high as 648 pg m$^{-3}$. Modeling studies conducted by Pan et al. (2008) using the STEM-Hg model also found large overestimation in dry deposition over Japan. Conversely, the model results are biased low by a factor of 2-5 over China, which indicates probable underestimation of Chinese Hg emissions.

### 3.7 Model performance summary and comparison

In this section, we summarize the statistical performance of GNAQPMS-Hg for TGM, oxidized mercury, and wet and dry deposition, compare the model performance over East Asia, North America and Europe, and assess the effects of horizontal resolution on model predictions over East Asia. As shown in Fig. 10, the simulated TGM and wet deposition are within a factor of two of the corresponding observations and within a factor of five for oxidized mercury and dry deposition. The statistical performance of GNAQPMS-Hg is comparable with that of other state-of-the-art Hg models (Bullock et al., 2008; Ryaboshapko et al., 2007; Pirrone and Keating, 2010).

### 3.7.1 East Asia versus North America and Europe

As illustrated in Table 3, the model statistical performance for all Hg parameters in North America and Europe is better than in East Asia. For example, the RMSEs between simulated and observed TGM over North America and Europe are 0.58 and 0.17 ng $m^{-3}$ but up to 3.61 ng $m^{-3}$ over East Asia. The poor model performance over East Asia is probably caused by the following reasons. Firstly, there are differences between simulated and observed data periods. Hg measurements over East Asia (especially China) are mainly taken from recent years, and the observed values are higher than in year 2001, which may lead to model underestimation. Hg anthropogenic emissions in China had increased by 164% during 1992-2007 (Liang et al., 2013) is an evidence. Secondly, there is a much higher spatial variation ratio (SVR, see Table 3) for Hg parameters in East Asia than North America and Europe. This implies that there are very intense spatial variations in surface Hg concentrations and deposition over East Asia which cannot be resolved at the coarse horizontal resolution used in global models (see Section 3.7.2). Thirdly, there are large uncertainties in emission inventories over East Asia. Large underestimations in Hg anthropogenic emissions over East Asia have been demonstrated in several previous studies (Jaffe et al., 2005; Pan et al., 2007; Friedli et al., 2004; Song et al., 2015). This is consistent with the simulated results in this study. Except the above factors, missing of some chemical and physical processes (e.g. gas-particle partitioning of Hg(II), in-plume reduction of Hg(II), dynamic land reemission) in the present model might also contribute to the poor model performance over East Asia.

### 3.7.2 Global versus nested simulations

In order to assess the impact of resolution on model predictions, an online nested simulation with higher resolution (0.33°x0.33°) over East Asia was conducted and compared to the global simulation with lower resolution (1°x1°). Emissions, meteorology, deposition and chemistry are self-consistent between the global and nested domains. The nested simulation uses higher resolution model inputs (e.g. topography, meteorology, emissions) and thus has the potential to better resolve high spatial variability of Hg concentrations and deposition in regional and local scales.

Fig. 7 and Fig. 9 compare the spatial distributions of simulated annual mercury wet deposition, accumulated precipitation and dry deposition over East Asia between the global and nested simulations. Although the global and nested simulations predict similar large scale patterns for Hg deposition, the nested simulation resolves many fine features which are lost in the global simulation by horizontal averaging. Firstly, in the nested domain, high deposition fluxes become more concentrated in regions with large emissions or precipitation resulting in higher spatial variability in deposition. Secondly, the nested simulation reveals elevated wet deposition in southwest China due to frequent orographic and convective precipitation. Finally, the nested simulation shows a more detailed land/ocean contrast in deposition over coastal regions. For example, over the coastal regions of southeast China and Japan, wet deposition increases due to scavenging of local emissions and enhanced precipitation (Fig. 7) while dry deposition decreases associated with the lower dry deposition velocity of Hg(0) over land than over ocean (Fig. 9). Our results are similar to those of Zhang et al. (2012) who conducted a nested simulation of Hg over North America using the GEOS-Chem model. More comparisons about the differences of dry and wet deposition and Hg budgets over East Asia between the two simulations are given in Fig. S16 and Table S6 in the supplement.

Fig. 11 and Table 3 further quantitatively compare the model performance over East Asia between the global and nested domains. In the Taylor diagram (Taylor, 2001), the position of each circle (or square) quantifies how closely the simulated results match observations. We can see that the simulated precipitation, oxidized Hg, wet and dry deposition agree better with observations in the nested domain than in the global domain (Fig. 11). The largest improvement is found in the simulated wet deposition. Specifically, the statistical parameter R for simulated wet deposition increases from 0.36 to 0.78, the NMB decreases from -61% to -28%, and the RMSE decreases by 24% (from 60.1 to 45.5 µg ~~ug~~ $m^{-2}$ $yr^{-1}$) (Table 3). But for TGM, oxidized Hg and dry deposition, the statistical parameters do not change significantly. For example, the RMSEs of simulated oxidized Hg and dry deposition decrease by 7% and 2% respectively, but increase by 7% for simulated TGM.

### 3.7.3 Online versus offline nested simulations

In order to further justify the online nested method, several model sensitivity experiments were conducted and the simulated results were compared to observations over East Asia. In the base simulation, the online nested method was used and the nested domain get boundary conditions from the global domain every 10 minutes. In the sensitivity simulations, the offline method was used and the boundary conditions were applied at 3 or 6 hours. The differences between the base and the sensitivity simulations were mainly caused by different frequency of boundary conditions. As shown in Table 4, the model performances are similar between the two sensitivity simulations (with 3 and 6 hour offline boundary conditions respectively). While the model performances in the base simulation are slightly better than those in the sensitivity simulations. The largest improvement is found in the simulated oxidized mercury. Specifically, the statistical parameter R for simulated oxidized mercury increases from 0.41 to 0.45, the NMB decreases from -18% to -12%, and the RMSE decreases by 4% compared to the simulated results with 6 hour offline boundary conditions. For wet deposition, little changes are found. This is because Hg wet deposition is not only affected by air concentrations but also precipitation. These results confirmed the effectiveness of the online nested method.

# 4 Impacts of Chinese primary anthropogenic sources on global Hg levels

Fig. 12 shows the contribution of Chinese primary anthropogenic sources (not including reemission) to annual mercury surface concentrations and total deposition in the Northern Hemisphere, and Fig. 13 gives the corresponding mean percentage contributions over different world regions (defined in Fig. S17 in the supplement), as derived from a sensitivity simulation with Chinese anthropogenic emissions shut off. In general, the largest percentage contribution is found in China itself, followed by neighbouring regions like the Korean Peninsula, Southeast Asia, Mongolia and Japan, but they are relatively small in other regions. Specifically, domestic anthropogenic emissions contribute on average 0.6 ng m$^{-3}$ (ranging from below 0.1 to above 3.0) to surface Hg concentrations and 18.4 µg ~~ug~~ m$^{-2}$ yr$^{-1}$ (ranging from below 2.0 to above 50.0) to total deposition in China. They account for about 30% and 62% on a national basis, respectively. The domestic contribution to deposition consists mainly of the deposition of directly emitted Hg(II) and Hg(P) near sources and deposition of Hg(II) formed by oxidation of Chinese Hg(0). For neighboring regions, the Chinese anthropogenic contributions to surface Hg concentrations and deposition are also large. For example, the percentage contributions are 11% (0.2-0.6 ng m$^{-3}$) and 15.2% (8-20 µg m$^{-2}$ yr$^{-1}$ ~~ug m$^{-2}$~~) over the Korean Peninsula, 10.4% (0.1-0.6 ng m$^{-3}$) and 8.2% (1-12 µg m$^{-2}$ yr$^{-1}$ ~~ug m$^{-2}$~~) over Southeast Asia, and 5.7% (0.1-0.4 ng m$^{-3}$) and 5.9% (2-15 µg m$^{-2}$ yr$^{-1}$ ~~ug m$^{-2}$~~) over Japan. For regions far away from China, the percentage contributions are small. They are 4.2% (0.06-0.1 ng m$^{-3}$) and 4.8% (0.5-4 µg m$^{-2}$ yr$^{-1}$ ~~ug m$^{-2}$~~) over North America, and 3.5% (below 0.08 ng m$^{-3}$) and 3.0% (below 2.0 µg m$^{-2}$ yr$^{-1}$ ~~ug m$^{-2}$~~) over Europe. The percentage contributions over North America determined from our simulation are comparable with the modeling study of Lei et al. (2013). They estimated that around 7% of TGM concentrations and 9% of total Hg deposition in the United States resulted from transpacific transport of Asian anthropogenic emissions. Given that about 53% of Asian anthropogenic Hg emissions are from China, it is reasonable that our estimated contributions are a little smaller than those reported by Lei et al. (2013).

Finally, there are another two issues which need to be addressed. Firstly, the above analysis mainly focuses on regional average contributions. However, the percentage contributions vary geographically inside the region. As shown in Fig. 12, contributions of domestic anthropogenic emissions to total deposition in Central Eastern China can exceed 40 μg m$^{-2}$ yr$^{-1}$ ~~ug m$^{-2}$~~, but they are below 5 μg m$^{-2}$ yr$^{-1}$ ~~ug m$^{-2}$~~ in Western China. Similarly, previous studies have found that Asia emissions make a much larger contribution to Hg deposition in the Western USA than in the Eastern USA (Seigneur et al., 2004; Strode et al., 2008). Secondly, the contributions from reemission of previously deposited anthropogenic Hg (treated as natural land or ocean reemission in GNAQPMS-Hg) are not taken into account in this study. Of the natural emissions, only one-third is considered not to be influenced by anthropogenic activities at all (Jung et al., 2009). In addition, according to the modeling study of Selin et al. (2008), 31% (including 22% primary and 9% recycled) of the deposition over USA is from anthropogenic emissions outside of North America. When considering reemission of previously deposited anthropogenic Hg, this suggests that the foreign anthropogenic contribution would increase by about 42% (from 22% to 31%). If we apply the same scaling factor to our attribution results, then the estimated Chinese anthropogenic contributions to Hg deposition over North America would increase from 4.8% to 6.8%. Therefore, it is also important to consider the reemission of previously deposited anthropogenic Hg.

# 5 Conclusions

A global nested atmospheric mercury transport model including Hg emissions, chemical transformation and deposition is introduced in this study. The treatment of Hg chemistry employs the $O_3$-OH oxidation and $SO_3^{2-}$-$HO_2$ reduction mechanisms. The gas phase reactions of Hg are added to the CBM-Z mechanism, while the aqueous phase reactions and wet deposition of Hg are calculated through adapting the RADM mechanism. The Wesely (1989) resistance model is used to deal with Hg dry deposition. The same meteorological fields, emissions, chemical and physical parameterizations are used in the global and nested domains.

The GNAQPMS-Hg model has a global mercury source of 10163 Mg $yr^{-1}$, including 2488 Mg $yr^{-1}$ primary anthropogenic emissions, 675 Mg $yr^{-1}$ biomass burning emissions, 2000 Mg $yr^{-1}$ land emissions (of which 75% is reemission), and 5000 Mg $yr^{-1}$ from the ocean. Dynamic bidirectional air-surface exchange of Hg is not included in the model. Instead, we simply apply static net emission fluxes to account for natural sources (including reemission) of Hg, with total emission amounts determined based on published estimates.

Based on existing routine monitoring networks (e.g. MDN, EMEP) and the published literature, global observations including surface Hg concentrations and deposition are collected for model evaluation. Compared with previous studies, many more observations over East Asia (especially China) are included in our dataset. Model evaluation shows that the spatial distribution and seasonal cycle of Hg concentrations and deposition can be reproduced reasonably well by GNAQPMS-Hg. Overall, the simulated annual TGM and wet deposition match observations within a factor of two, and within a factor of five for oxidized mercury and dry deposition. This performance is comparable with other state-of-the-art Hg models. Some model deficiencies have also been identified. GNAQPMS-Hg is systematically biased low relative to cruise observations in the Northern Hemisphere, due to poor representation of the air-sea exchange mechanism for Hg. GNAQPMS-Hg overestimates oxidized mercury concentrations in most parts of the world which may partially be caused by excessive oxidation of Hg(0) by relatively high concentrations of OH and $O_3$ and uncertainties associated with Hg chemical speciation in emission inventories. The model performs significantly better in North America and Europe than in East Asia. This can probably be attributed to the large uncertainties in emission inventories, coarse model resolution and inconsistency between the simulation and observation periods in East Asia. An online nested simulation with higher resolution (0.33°x0.33°) over East Asia was conducted to examine the impact of horizontal resolution on model predictions. Relative to the global simulation, the nested simulation can better resolve high spatial variability of Hg concentrations and deposition over East Asia, can better capture features such as higher wet deposition due to orographic and convective precipitation, and land/ocean contrast. Statistically, the RMSE of simulated wet deposition over East Asia is reduced by 24% in the nested simulation.

To quantify the impacts of Chinese anthropogenic sources on global Hg levels, a model sensitivity simulation was conducted with Chinese anthropogenic emissions shut off. The results show that these sources contribute 30% and 62% of surface mercury concentrations and deposition over China. Outside of China, the largest percentage contributions of 11% and 15.2% are found in the Korean Peninsula, following by Southeast Asia (10.4% and 8.2%), Mongolia (6.1% and 8.6%), and Japan (5.7% and 5.9%). For regions far away from China, the percentage contributions are relatively small (e.g. 4.2% and 4.8% over North America; 3.5% and 3.0% over Europe).

To perfect the model, future improvements will be focused on the following aspects: 1) employing dynamic parameterizations for bidirectional air-surface (sea and land) exchange of Hg (Selin et al., 2008; Bash, 2010; Strode et al., 2007) to better reflect natural emissions (including reemission), 2) including fast in-plume reduction of Hg(II) to better characterize Hg(II) distribution near large point sources (Amos et al., 2012), and 3) reducing uncertainties in the anthropogenic Hg emission inventory, especially the Hg speciation profile. Finally, establishment of routine Hg monitoring networks would be also very helpful for enhancing and improving modeling studies in East Asia.

## Code availability

Please contact Huansheng Chen (E-mail: chenhuansheng@mail.iap.ac.cn) to obtain the source code of GNAQPMS-Hg.

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**Table 1.** Reactions and rate constants used in the GNAQPMS-Hg model.

| NO. | Reaction | Rates (k or K)[a] | References |
|---|---|---|---|
| **Gas-phase reactions** | | | |
| RG1 | $\text{Hg(0)(g)+O}_3\text{(g)}\rightarrow\text{Hg(II)(g)}$ | $3\text{x}10^{-20}\ \text{cm}^3\ \text{molec}^{-1}\ \text{s}^{-1}$ | Hall (1995) |
| RG2 | $\text{Hg(0)(g)+HCl(g)}\rightarrow\text{HgCl}_2\text{(g)}$ | $1\text{x}10^{-19}\ \text{cm}^3\ \text{molec}^{-1}\ \text{s}^{-1}$ | Hall and Bloom (1993) |
| RG3 | $\text{Hg(0)(g)+H}_2\text{O}_2\text{(g)}\rightarrow\text{Hg(OH)}_2\text{(g)}$ | $8.5\text{x}10^{-19}\ \text{cm}^3\ \text{molec}^{-1}\ \text{s}^{-1}$ | Tokos et al. (1998) |
| RG4 | $\text{Hg(0)(g)+Cl}_2\text{(g)}\rightarrow\text{HgCl}_2\text{(g)}$ | $2.6\text{x}10^{-18}\ \text{cm}^3\ \text{molec}^{-1}\ \text{s}^{-1}$ | Ariya et al. (2002) |
| RG5 | $\text{Hg(0)(g)+OH(g)}\rightarrow\text{Hg(OH)}_2\text{(g)}$ | $8\text{x}10^{-14}\ \text{cm}^3\ \text{molec}^{-1}\ \text{s}^{-1}$ | Sommar et al. (2001) |
| **Gas-liquid equilibria** | | | |
| GL1 | $\text{Hg(0)(g)}\leftrightarrow\text{Hg(0)(aq)}$ | $0.11\ \text{M atm}^{-1}$ | Sanemasa (1975) |
| GL2 | $\text{HgCl}_2\text{(g)}\leftrightarrow\text{HgCl}_2\text{(aq)}$ | $1.4\text{x}10^{6}\ \text{M atm}^{-1}$ | Lindqvist and Rodhe (1985) |
| GL3 | $\text{Hg(OH)}_2\text{(g)}\leftrightarrow\text{Hg(OH)}_2\text{(aq)}$ | $1.2\text{x}10^{4}\ \text{M atm}^{-1}$ | Lindqvist and Rodhe (1985) |
| **Aqueous-phase equilibria** | | | |
| AE1 | $\text{HgCl}_2\text{(aq)}\leftrightarrow\text{Hg}^{2+}\text{+2Cl}^-$ | $1\text{x}10^{-14}\ \text{M}^2$ | Sillen et al. (1964) |
| AE2 | $\text{Hg(OH)}_2\text{(aq)}\leftrightarrow\text{Hg}^{2+}\text{+2OH}^-$ | $1\text{x}10^{-22}\ \text{M}^2$ | Sillen et al. (1964) |
| AE3 | $\text{Hg}^{2+}\text{+SO}_3^{2-}\leftrightarrow\text{HgSO}_3$ | $2.1\text{x}10^{13}\ \text{M}^{-1}$ | Van Loon et al. (2001) |
| AE4 | $\text{HgSO}_3\text{+SO}_3^{2-}\leftrightarrow\text{Hg(SO}_3)_2^{2-}$ | $1\text{x}10^{10}\ \text{M}^{-1}$ | Van Loon et al. (2001) |
| **Aqueous-phase reaction** | | | |
| RA1 | $\text{Hg(0)(aq)+O}_3\text{(aq)}\rightarrow\text{Hg}^{2+}$ | $4.7\text{x}10^{7}\ \text{M}^{-1}\ \text{s}^{-1}$ | Munthe (1992) |
| RA2 | $\text{Hg(0)(aq)+OH(aq)}\rightarrow\text{Hg}^{2+}$ | $2\text{x}10^{9}\ \text{M}^{-1}\ \text{s}^{-1}$ | Lin and Pehkonen (1997) |
| RA3 | $\text{HgSO}_3\text{(aq)}\rightarrow\text{Hg(0)(aq)}$ | $0.0106\ \text{s}^{-1}$ | Van Loon et al. (2000) |
| RA4 | $\text{Hg(II)(aq)+HO}_2\text{(aq)}\rightarrow\text{Hg(0)(aq)}$ | $1.7\text{x}10^{4}\ \text{M}^{-1}\ \text{s}^{-1}$ | Pehkonen and Lin (1998) |
| RA5 | $\text{Hg(0)(aq)+HOCl(aq)}\rightarrow\text{Hg}^{2+}$ | $2.09\text{x}10^{6}\ \text{M}^{-1}\ \text{s}^{-1}$ | Lin and Pehkonen (1998) |
| RA6 | $\text{Hg(0)(aq)+OCl}^-\rightarrow\text{Hg}^{2+}$ | $1.99\text{x}10^{6}\ \text{M}^{-1}\ \text{s}^{-1}$ | Lin and Pehkonen (1998) |
| **Adsorption of Hg(II) on PM in the aqueous-phase** | | | |
| AD1 | $\text{Hg(II)(aq)}\leftrightarrow\text{Hg(II)(p)}$ | $34\ \text{L g}^{-1}$ | Seigneur et al. (1998) |

[a] The reaction rate constants are for temperatures in the range of 20 to 25$^0$C. No temperature dependence information is available.

**Table 2.** Global budgets of TGM in the literature (Unit: Mg yr$^{-1}$).

| | Bergan et al. (1999) | Shia et al. (1999) | Lamborg et al. (2002) | Mason et al. (2002) | Seigneur et al. (2004) | Selin et al. (2007) | Selin et al. (2008) | This work |
|---|---|---|---|---|---|---|---|---|
| **Total Sources** | 6050 | 6100 | 4400 | 6600 | 6411 | 7000 | 11200 | 10163 |
| **anthropogenic** | 2150 | 2100 | 2600 | 2400 | 2143 | 2200 | 3400 | 2488 |
| **land** | 2500 | 2000 | 1000 | 1600 | 2290 | 2000 | 2800 | 2675 |
| **ocean** | 1400 | 2000 | 800 | 2600 | 1978 | 2800 | 5000 | 5000 |
| **Total Sinks** | 6050 | 6100 | 4200 | 6600 | 6411 | 7000 | 11200 | 10163 |
| **Wet deposition** | | 2800 | | 3920 | | 2100 | | 2283 |
| **Dry deposition** | | 3300 | | 2680 | | 4700 | | 7880 |
| **TGM Burden** | 6050 | 10400 | 5220 | 5000 | 7690 | 5360 | 5600 | 5507 |
| **TGM lifetime(yr)** | 1 | 1.7 | 1.3 | 0.76 | 1.2 | 0.79 | 0.5 | 0.54 |

**Table 3.** Statistical summary of comparisons of the model results with observations[a].

| Parameter | Region | R | NMB | RMSE | SVR[b] |
|---|---|---|---|---|---|
| TGM | East Asia Nested | 0.51 | -39% | 3.87 | 2.56 |
| | East Asia | 0.54 | -32% | 3.61 | 2.56 |
| | North America | 0.69 | 18% | 0.58 | 0.48 |
| | Europe | 0.57 | -8% | 0.17 | 0.35 |
| | Global | 0.70 | -18% | 2.22 | - |
| Oxidized mercury | East Asia Nested | 0.45 | -12% | 242 | 3.66 |
| | East Asia | 0.31 | -10% | 259 | 3.66 |
| | North America | 0.53 | 148% | 28 | 1.61 |
| | Europe | 0.91 | 155% | 48 | 1.00 |
| | Global | 0.53 | 3% | 185 | - |
| Wet deposition | East Asia Nested | 0.78 | -28% | 45.5 | 6.69 |
| | East Asia | 0.36 | -61% | 60.1 | 6.69 |
| | North America | 0.76 | -4% | 4.3 | 1.89 |
| | Europe | 0.78 | 4% | 1.5 | 1.40 |
| | Global | 0.38 | -36% | 29.3 | - |
| Dry deposition | East Asia Nested | 0.88 | -42% | 87.0 | - |
| | East Asia | 0.81 | -42% | 88.5 | - |

[a] R, NMB, RMSE, SVR represent correlation coefficient, normalized mean bias, root mean square error, spatial variation ratio. Units of TGM, oxidized mercury, wet and dry deposition are ng m$^{-3}$, pg m$^{-3}$, µ~~u~~g m$^{-2}$ yr$^{-1}$, µ~~u~~g m$^{-2}$ yr$^{-1}$ respectively.

[b] SVR defines as (max-min)/mean observations over all sites.

**Table 4.** Statistical comparisons of the online and offline nested simulation results with observations over East Asia[a].

| Parameter | Case[b] | *R* | NMB | RMSE |
|---|---|---|---|---|
| TGM | Base simulation | 0.51 | -39% | 3.87 |
| | 3 hour boundary conditions | 0.50 | -42% | 3.94 |
| | 6 hour boundary conditions | 0.50 | -42% | 3.95 |
| Oxidized mercury | Base simulation | 0.45 | -12% | 242.15 |
| | 3 hour boundary conditions | 0.42 | -17% | 251.52 |
| | 6 hour boundary conditions | 0.41 | -18% | 252.78 |
| Wet deposition | Base simulation | 0.78 | -28% | 45.47 |
| | 3 hour boundary conditions | 0.78 | -29% | 45.75 |
| | 6 hour boundary conditions | 0.78 | -29% | 45.78 |
| Dry deposition | Base simulation | 0.88 | -42% | 87.02 |
| | 3 hour boundary conditions | 0.88 | -44% | 88.55 |
| | 6 hour boundary conditions | 0.87 | -45% | 88.96 |

[a] *R*, NMB, RMSE represent correlation coefficient, normalized mean bias, root mean square error. Units of TGM, oxidized mercury, wet and dry deposition are ng m$^{-3}$, pg m$^{-3}$, µ~~u~~g m$^{-2}$ yr$^{-1}$, µ~~u~~g m$^{-2}$ yr$^{-1}$ respectively.

[b] In the base simulation, the nested domain gets boundary conditions from the global domain every 10 minutes.

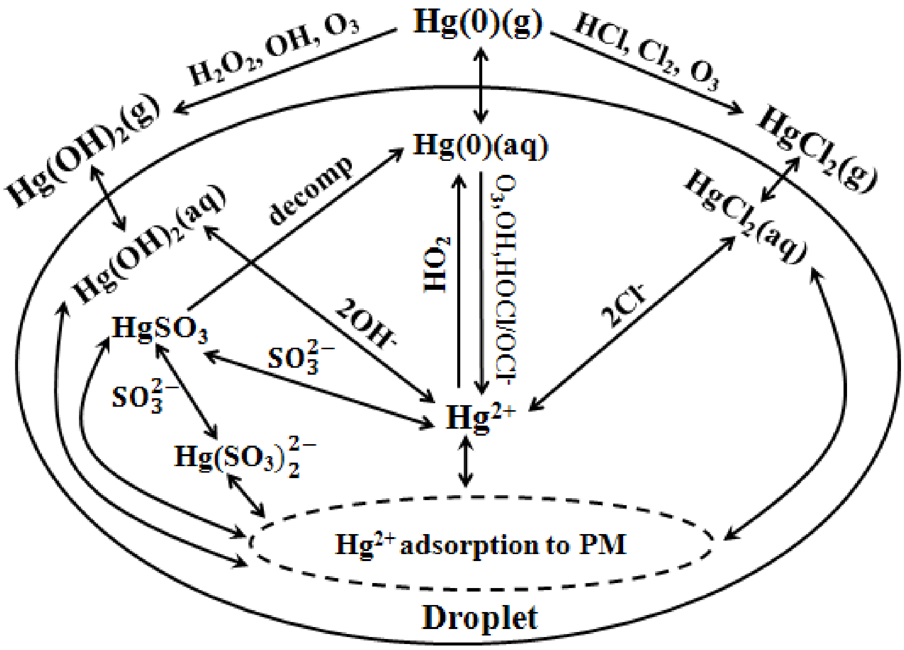


**Fig. 1.** Schematic of different mercury reactions utilized in the GNAQPMS-Hg model.

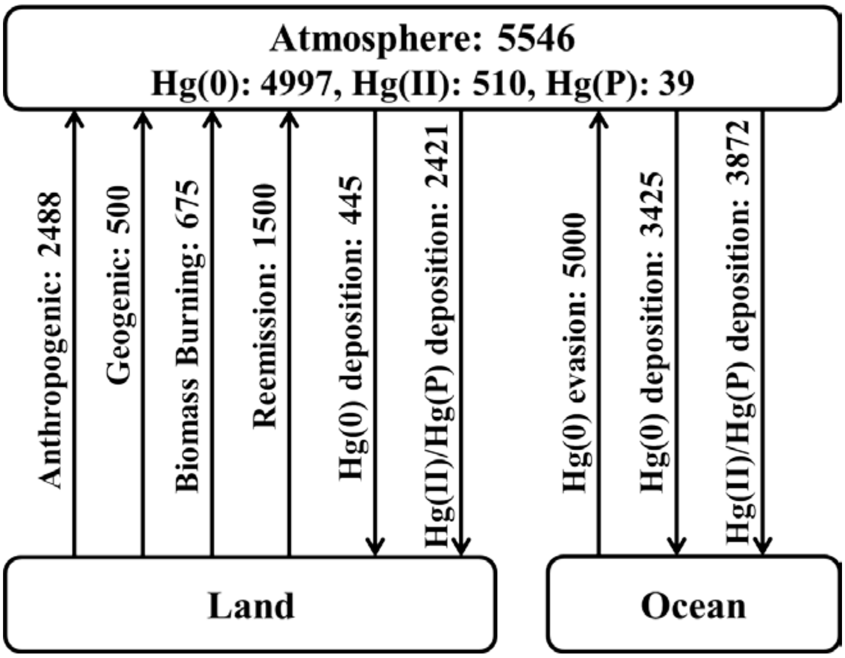
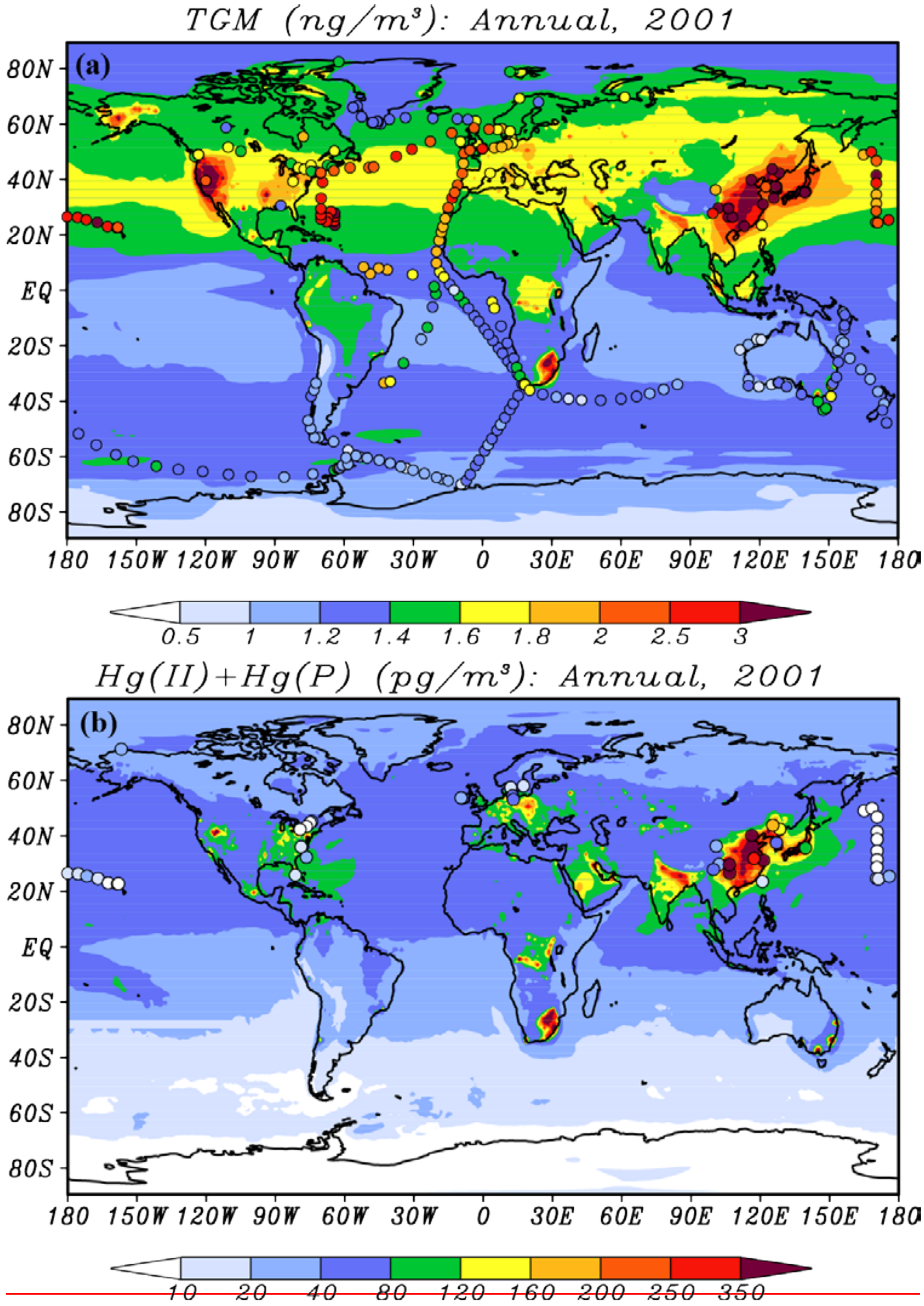


**Fig. 2.** Global atmospheric mercury budget in GNAQPMS-Hg. Units are Mg $yr^{-1}$.


TGM (ng/m³): Annual, 2001
(a)
80N
60N
40N
20N
EQ
20S
40S
60S
80S
180
150W
120W
90W
60W
30W
0
30E
60E
90E
120E
150E
180
0.5
1
1.2
1.4
1.6
1.8
2
2.5
3
Hg(II)+Hg(P) (pg/m³): Annual, 2001
(b)
10
20
40
80
120
160
200
250
350

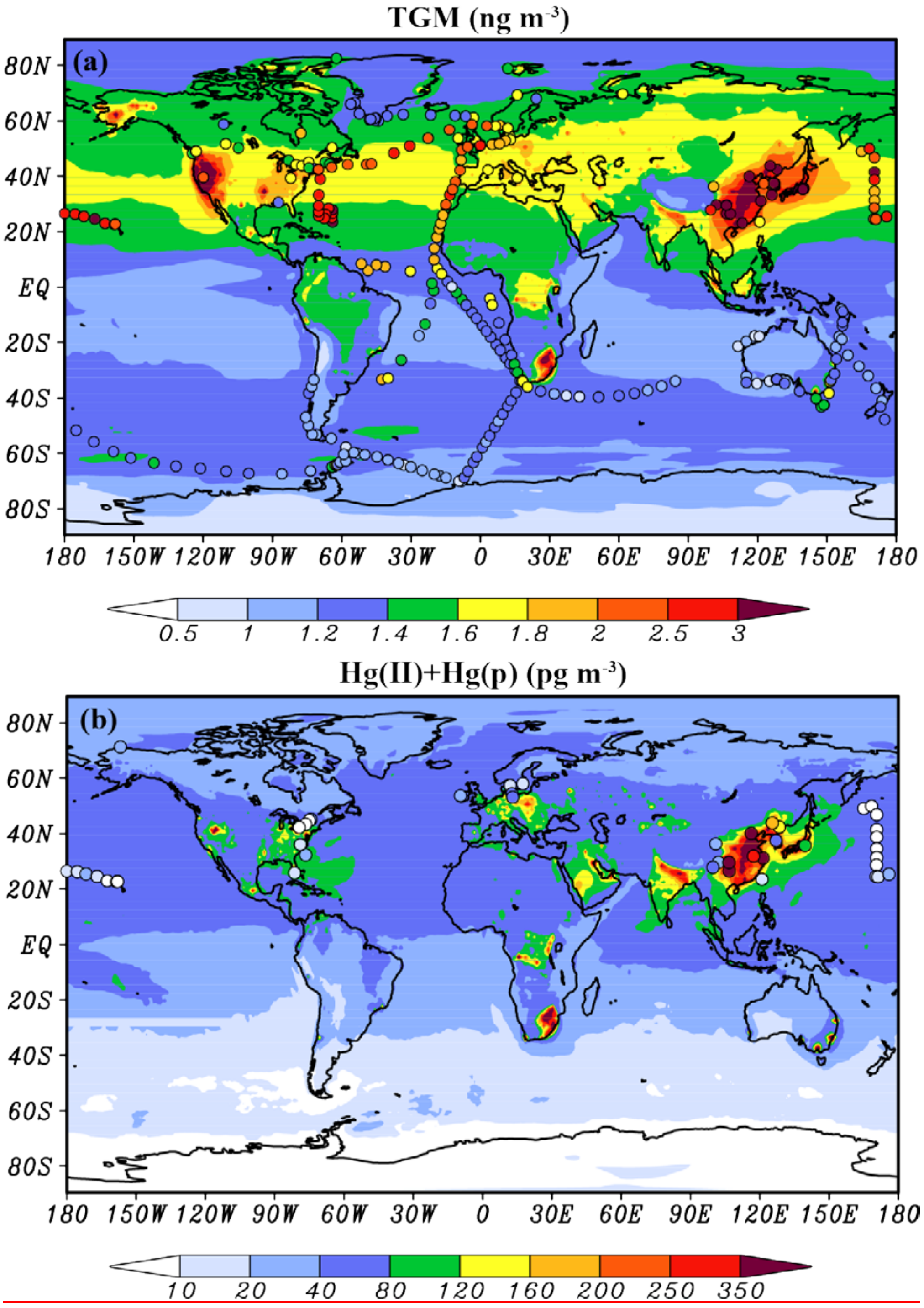
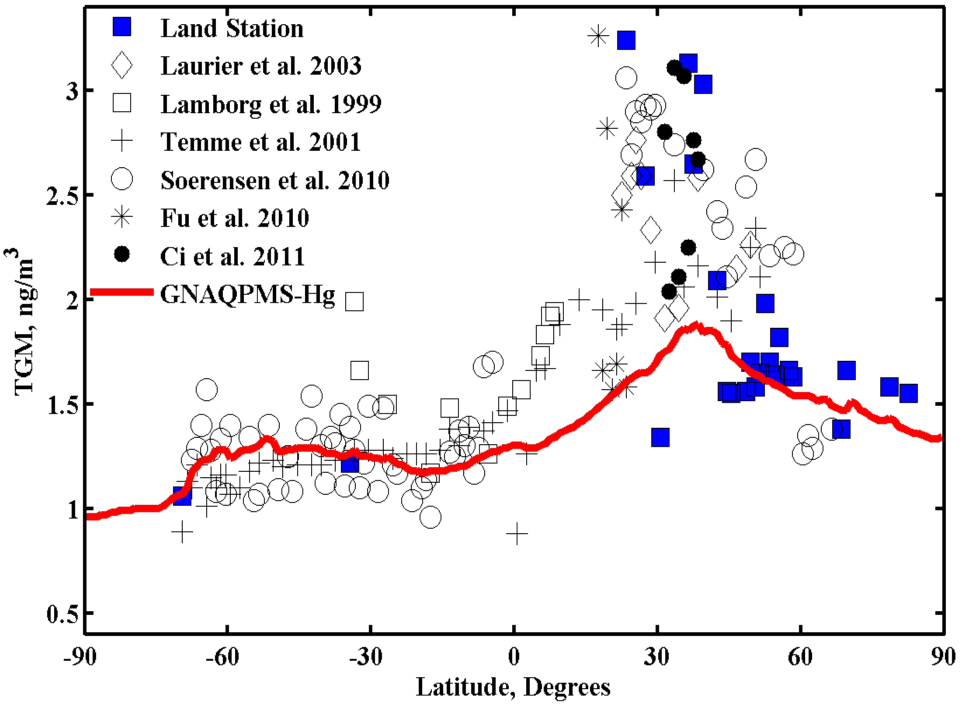


**Fig. 3.** Annual average TGM (a) and oxidized mercury (Hg(II)+Hg(P),b) concentrations in surface air. Model results (background, for year 2001) are compared to observations (circles) from long-term surface sites and short-term ship cruises. Units of TGM and oxidized mercury are ng m$^{-3}$ and pg m$^{-3}$, respectively.


Land Station
Laurier et al. 2003
Lamborg et al. 1999
Temme et al. 2001
Soerensen et al. 2010
Fu et al. 2010
Ci et al. 2011
GNAQPMS-Hg
TGM, ng/m3
Latitude, Degrees
3
2.5
2
1.5
1
0.5
-90
-60
-30
0
30
60
90

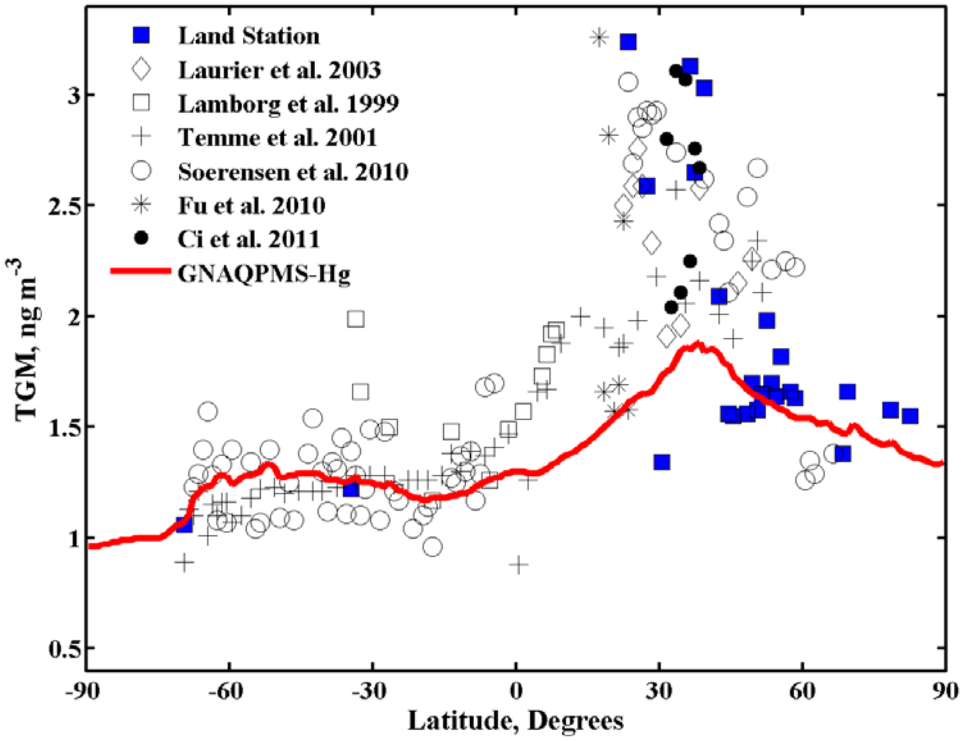
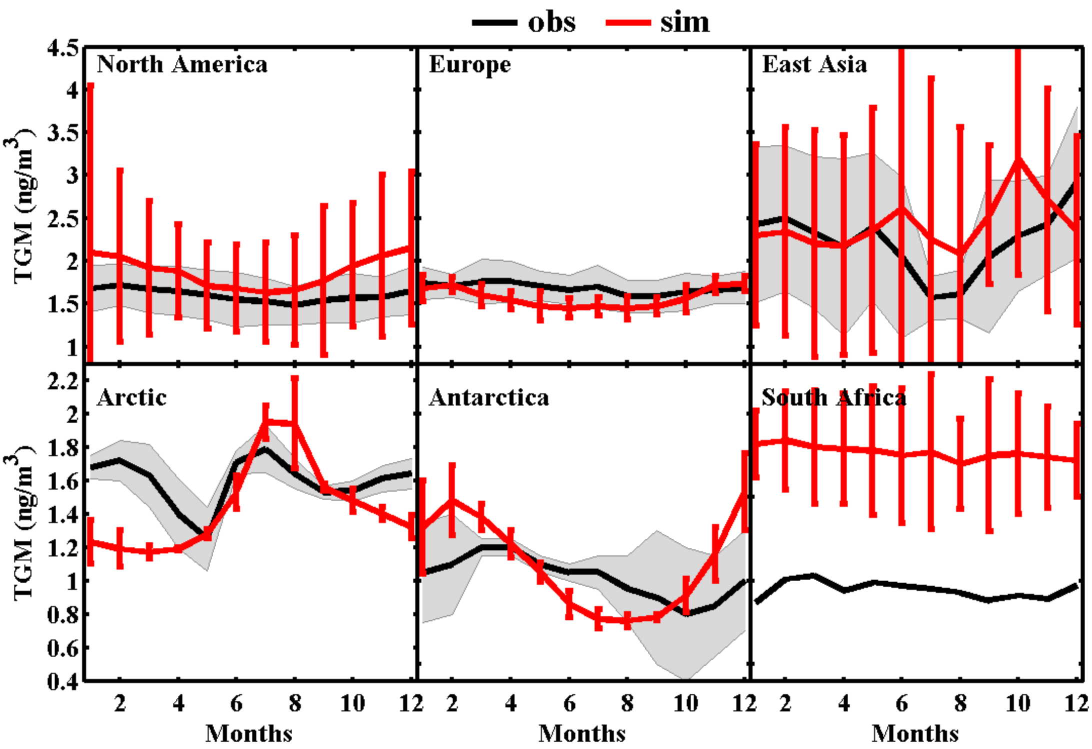


**Fig. 4.** Variation of surface TGM ~~surface~~ concentrations (ng $m^{-3}$) with latitude. Zonally averaged, annual mean_

model results (line) are compared to observations (symbols).



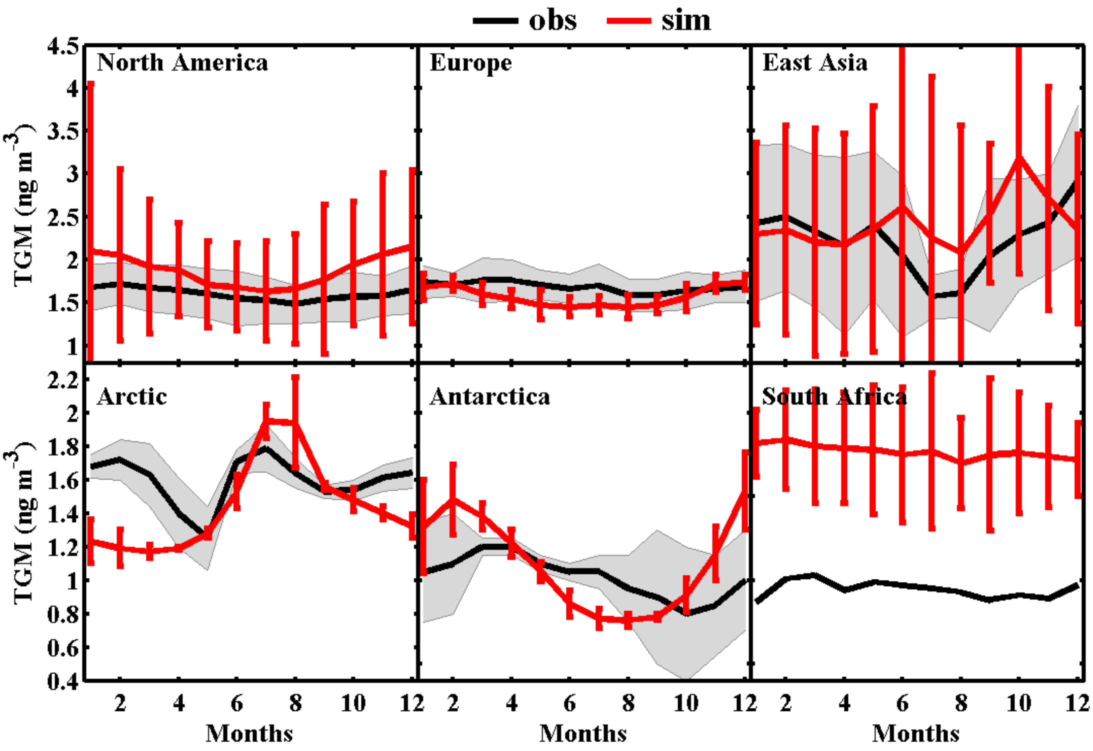
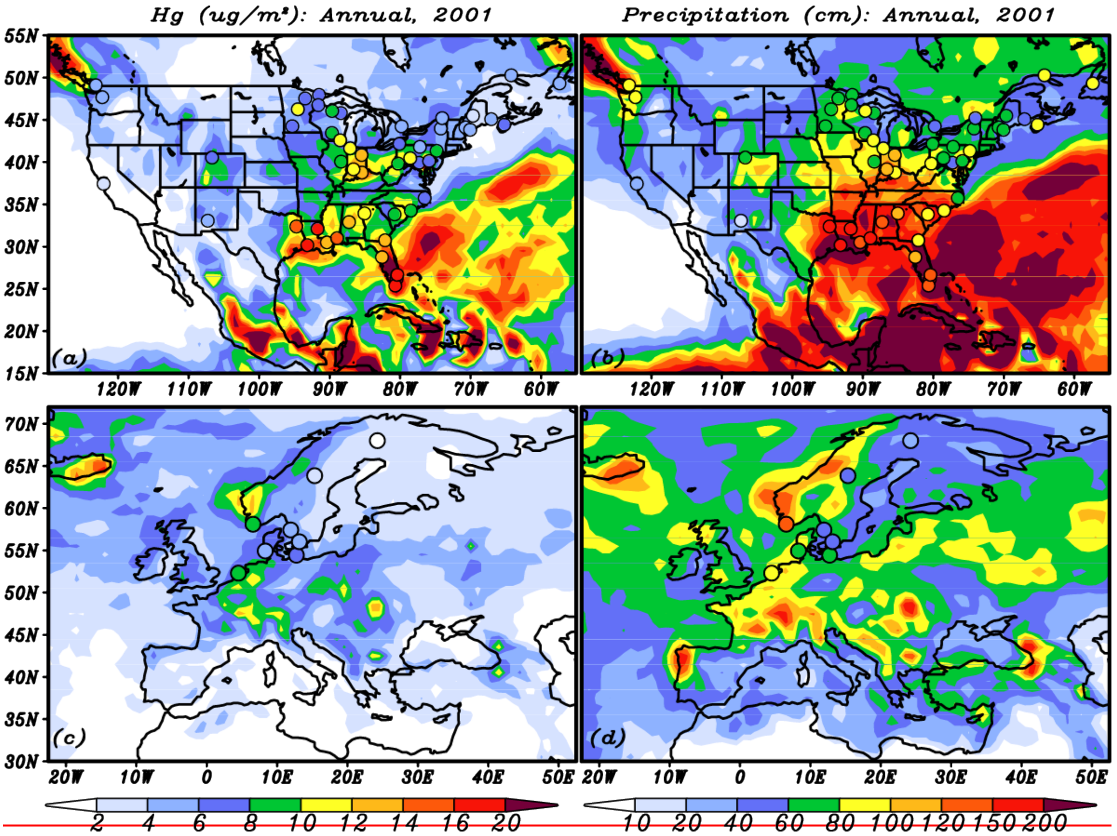


**Fig. 5.** Mean seasonal variation of TGM (ng m$^{-3}$) at North America, Europe, East Asia, Arctic, Antarctica

and South Africa sites. Gray shaded areas and red vertical bars show one standard deviation over the sites for observations and for model results.



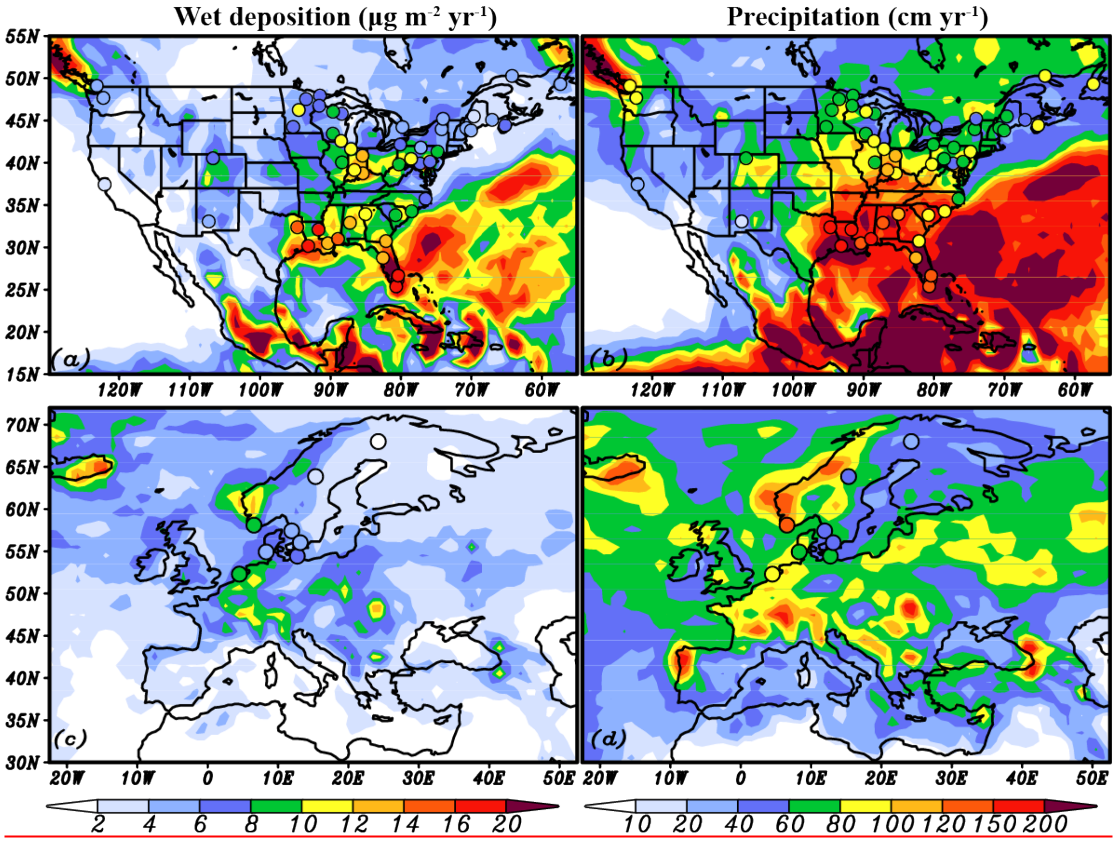
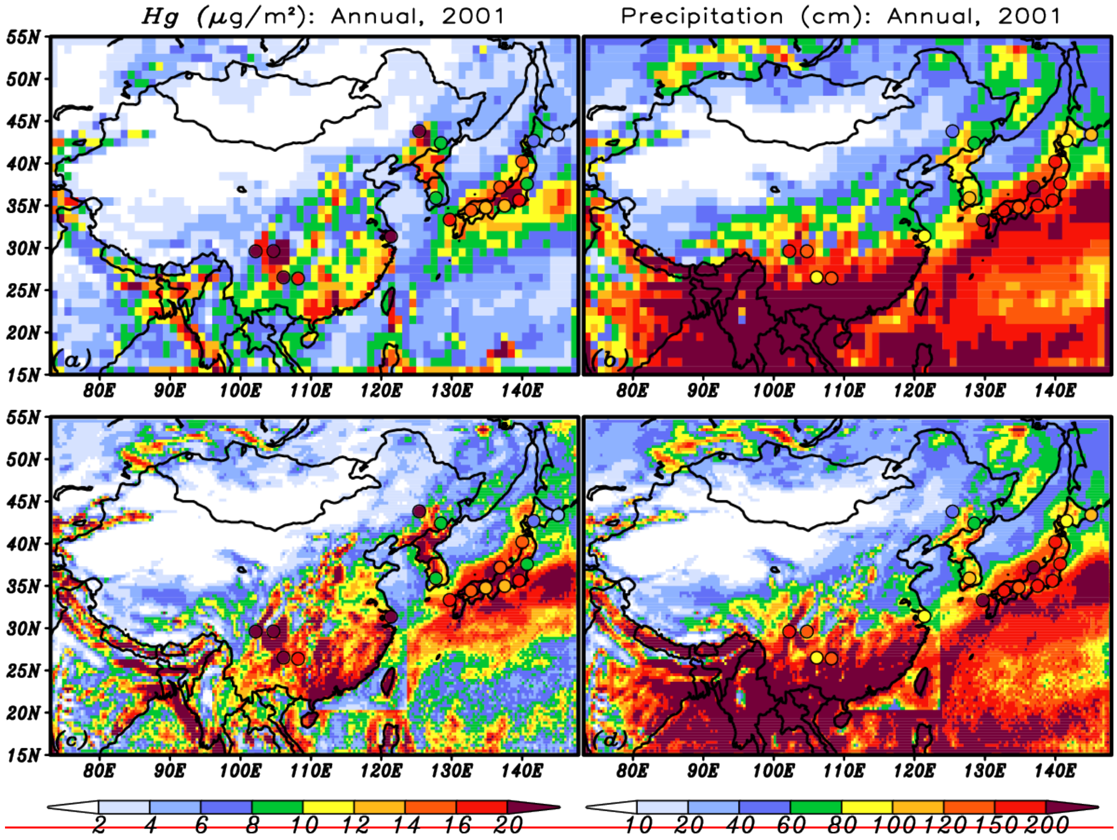


**Fig. 6.** Simulated annual mercury wet deposition (μg m$^{-2}$ yr$^{-1}$) and accumulated precipitation (cm yr$^{-1}$) over North America (a, b) and Europe (c, d) in 2001. Overlaid points show observations for the same year from the Mercury Deposition Network (MDN) over North America, and the European Monitoring and Evaluation Programme (EMEP) over Europe.



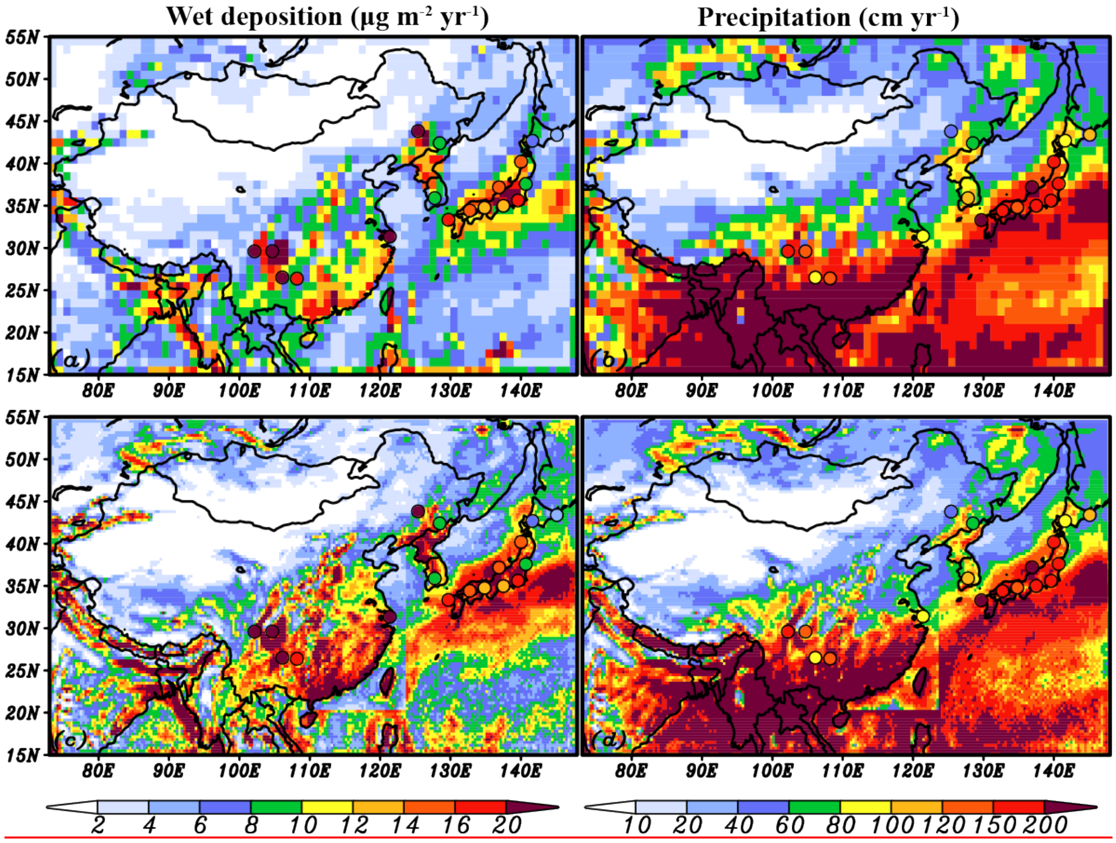
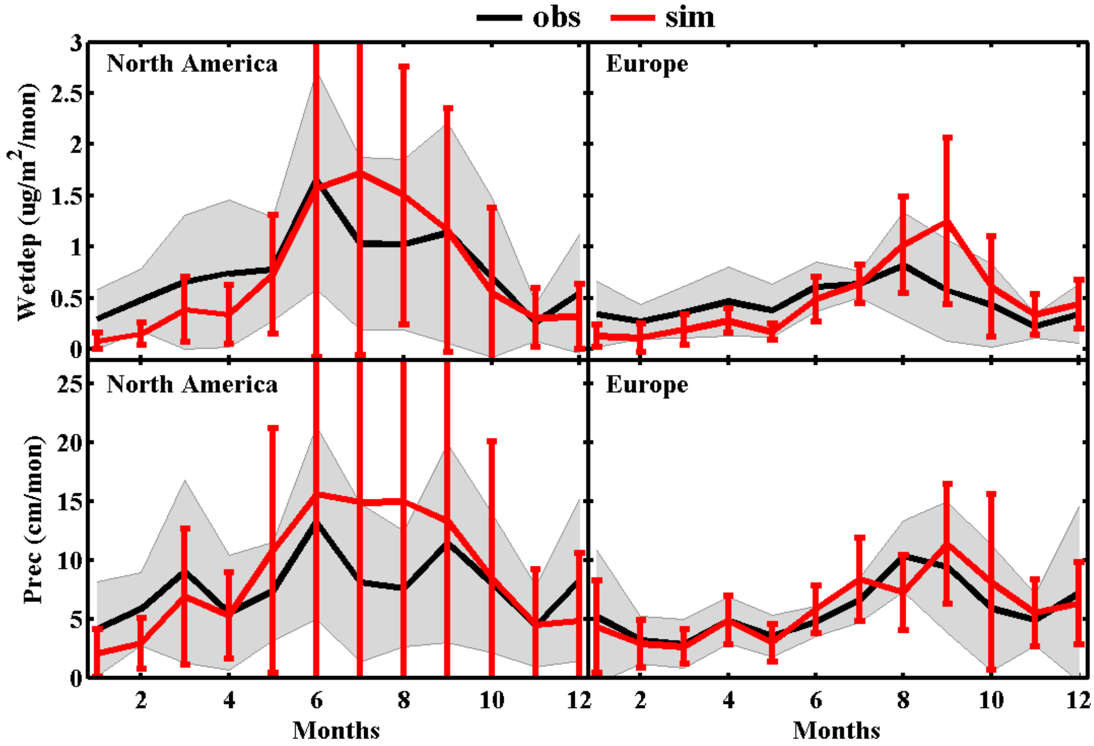


**Fig. 7.** Simulated annual mercury wet deposition (µg $m^{-2}$ $yr^{-1}$) and accumulated precipitation (cm $yr^{-1}$) over East Asia in_

the global (a, b) and nested (c, d) domains in 2001. Overlaid points show observations collected_ from the literature. Note that observations and simulated results are not in the same year.



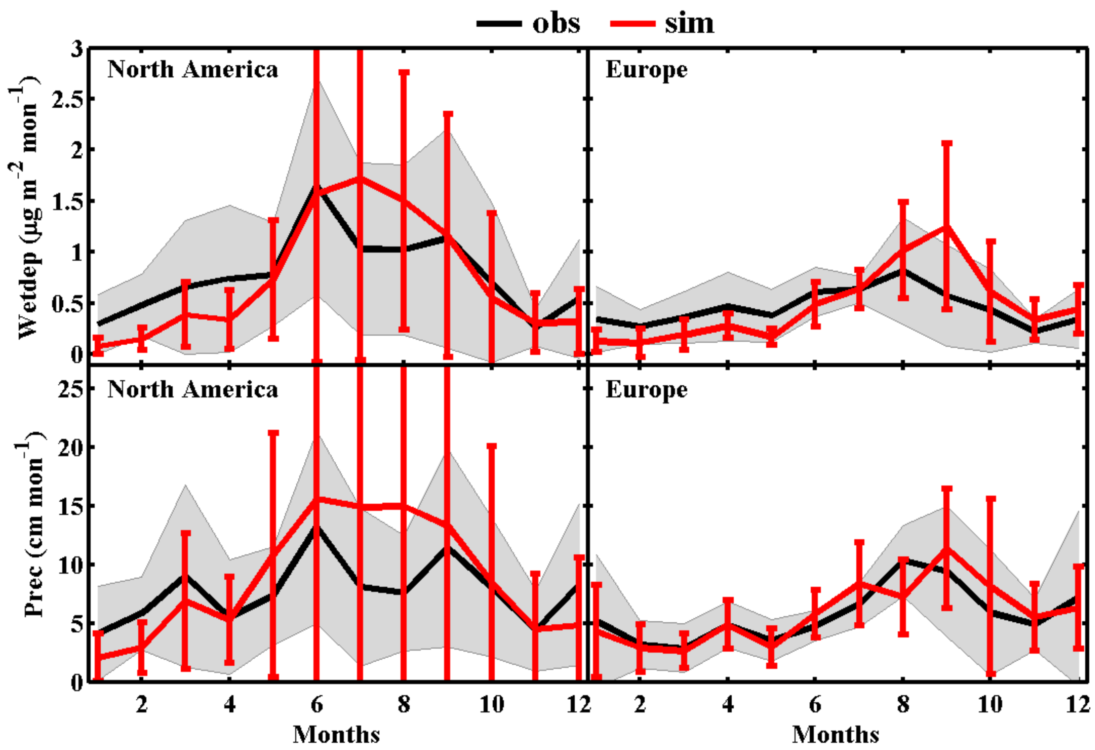
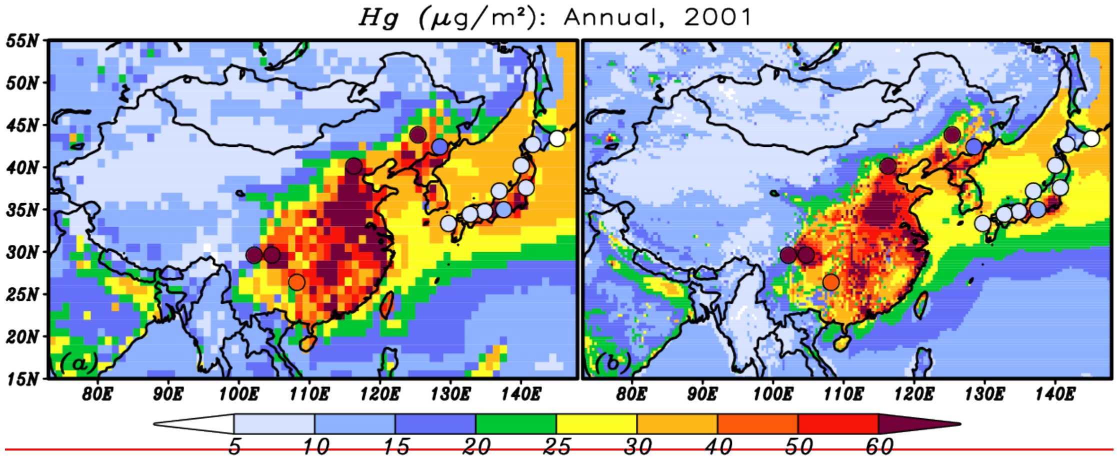


**Fig. 8.** Mean seasonal variation of mercury wet deposition (µg m$^{-2}$ mon$^{-1}$) and accumulated precipitation (cm mon$^{-1}$) at North America (51 sites averaged) and Europe (8 sites averaged) sites in 2001. Gray shaded areas and red vertical bars show one standard deviation over the sites for observations and for model results.



Dry deposition (μg m$^{-2}$ yr$^{-1}$)

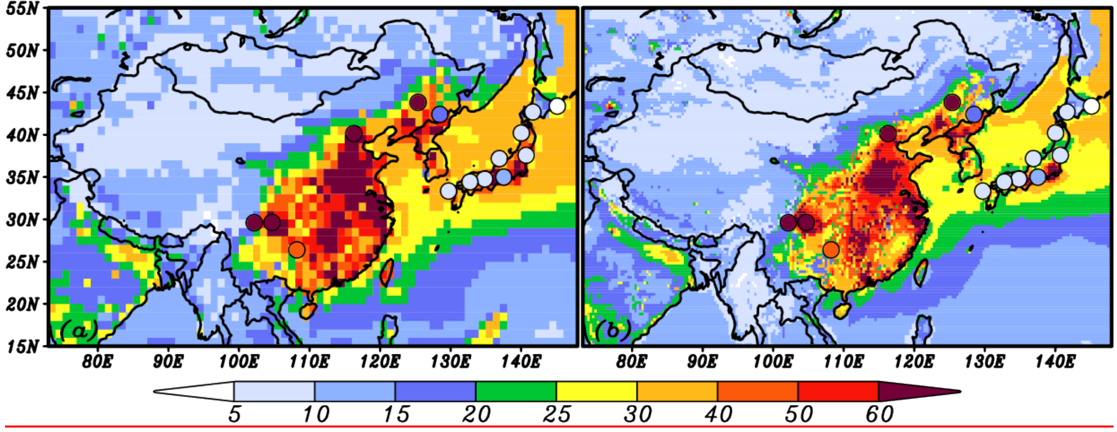
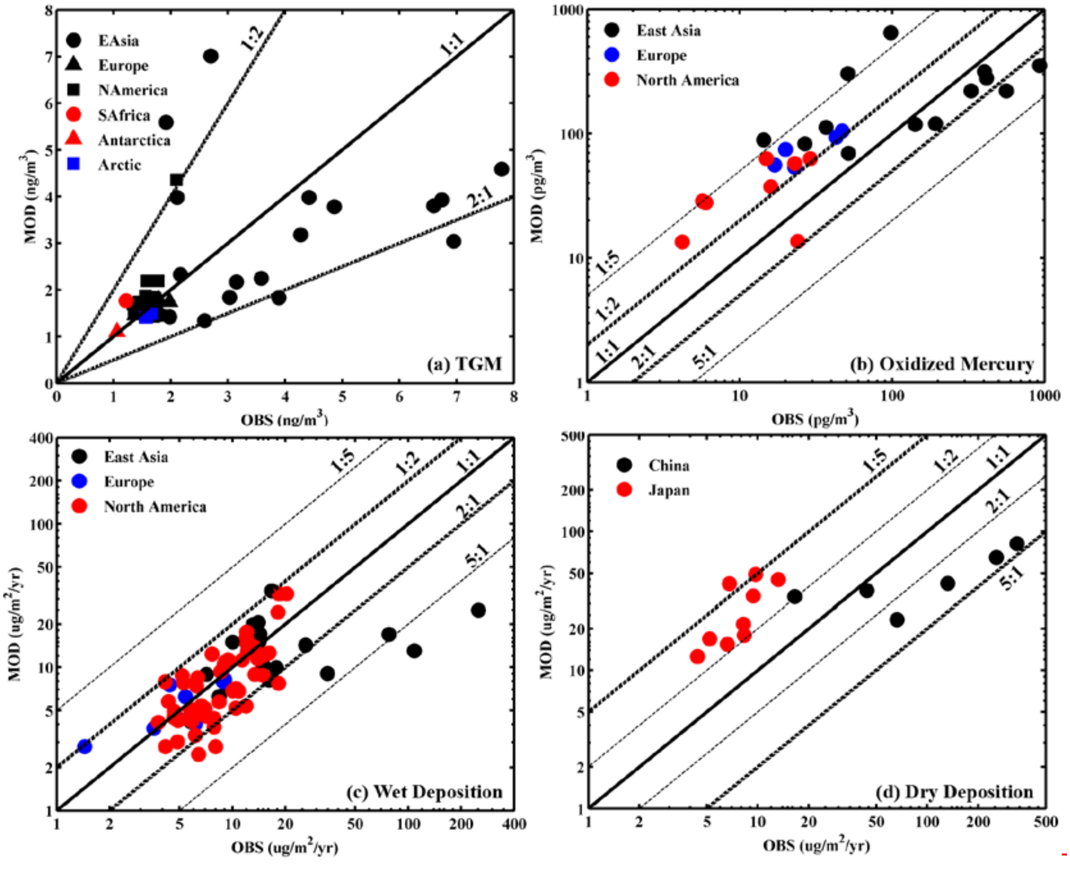


**Fig. 9.** Simulated annual mercury dry deposition (µg m$^{-2}$ yr$^{-1}$) over East Asia in the global (a) and nested (b) domains in 2001. Overlaid points show observations collected from the literature. Note that observations and simulated results are not in the same year.


(a) TGM
EAsia
Europe
NAmerica
SAfrica
Antarctica
Arctic
1:2
1:1
2:1
MOD (ng/m$^3$)
OBS (ng/m$^3$)
0
1
2
3
4
5
6
7
8
(b) Oxidized Mercury
East Asia
Europe
North America
1:5
1:2
1:1
2:1
5:1
MOD (pg/m$^3$)
OBS (pg/m$^3$)
1
10
100
1000
(c) Wet Deposition
East Asia
Europe
North America
1:5
1:2
1:1
2:1
5:1
MOD (ug/m$^2$/yr)
OBS (ug/m$^2$/yr)
1
2
5
10
20
50
100
200
400
(d) Dry Deposition
China
Japan
1:5
1:2
1:1
2:1
5:1
MOD (ug/m$^2$/yr)
OBS (ug/m$^2$/yr)
1
2
5
10
20
50
100
200
500

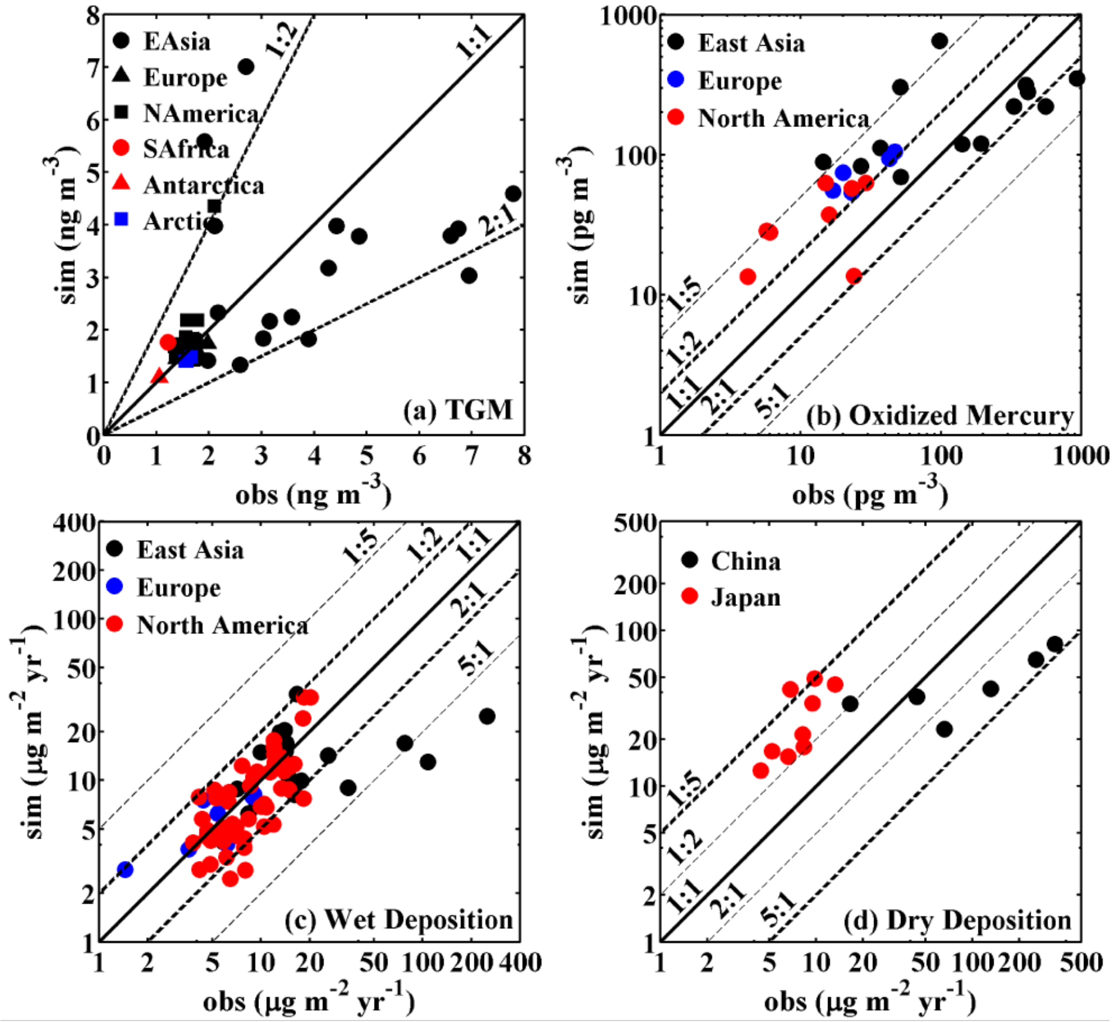


**Fig. 10.** Simulated vs. observed TGM (a), oxidized mercury (b), wet deposition (c), dry deposition (d) in different regions. Note that coordinates are different in different panels. Units of TGM, oxidized mercury, wet deposition and dry deposition are ng $m^{-3}$, pg $m^{-3}$, μg $m^{-2}$ $yr^{-1}$, μg $m^{-2}$ $yr^{-1}$, respectively.

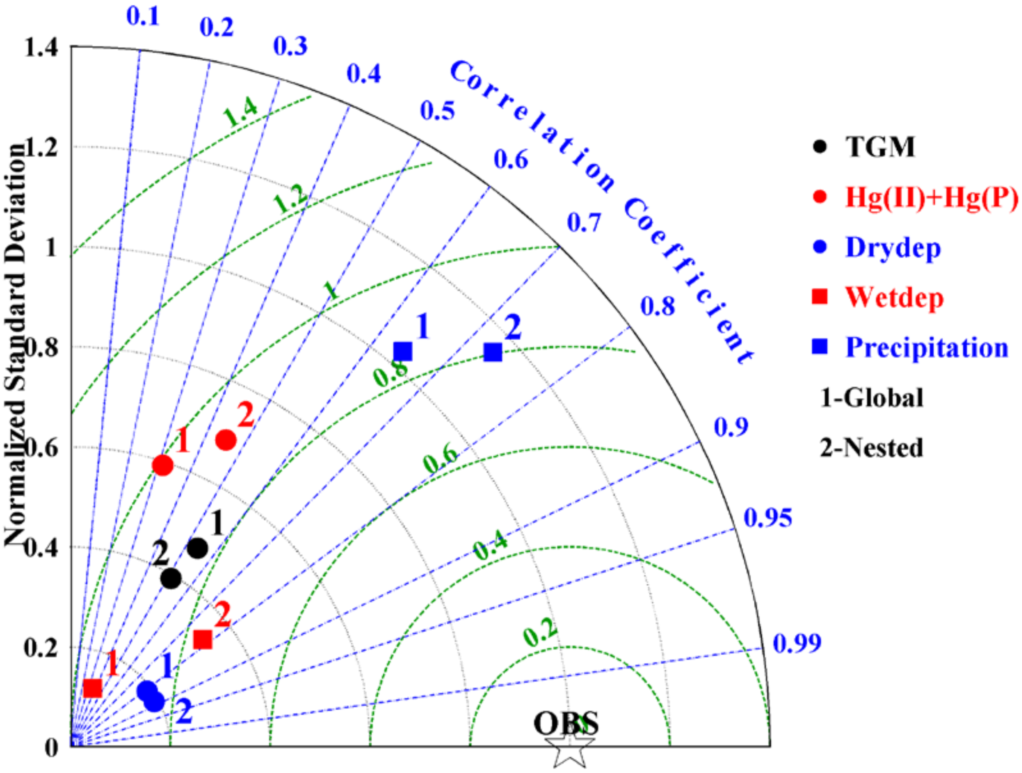


**Fig. 11.** Taylor Diagram of simulated annual TGM, Hg(II)+Hg(P), dry deposition, wet deposition and precipitation over East Asia in the global and nested domains (denoted by 1 and 2).

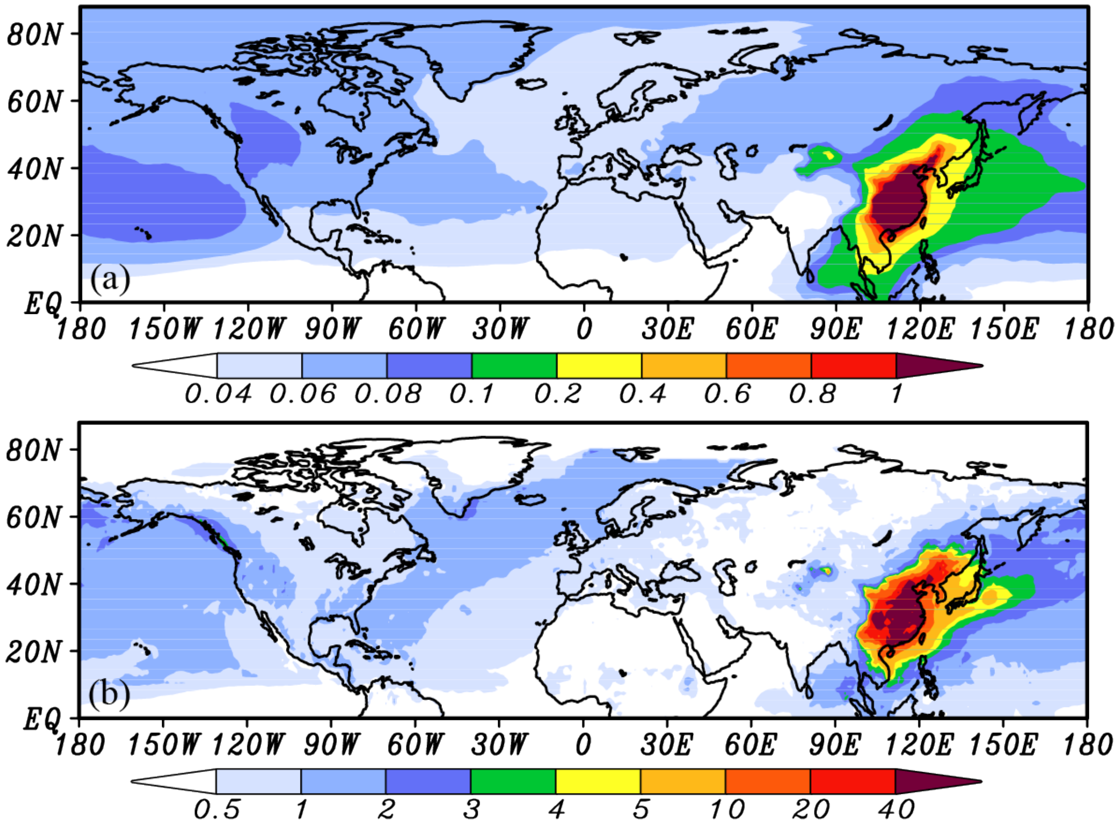


**Fig. 12.** Contributions of Chinese primary anthropogenic sources to (a) annual mercury surface concentrations and (b) total (wet plus dry) deposition in the Northern Hemisphere. The units of~~for~~ mercury concentrations and deposition are ng m$^{-3}$ and µg ~~ug~~ m$^{-2}$ yr$^{-1}$, respectively.

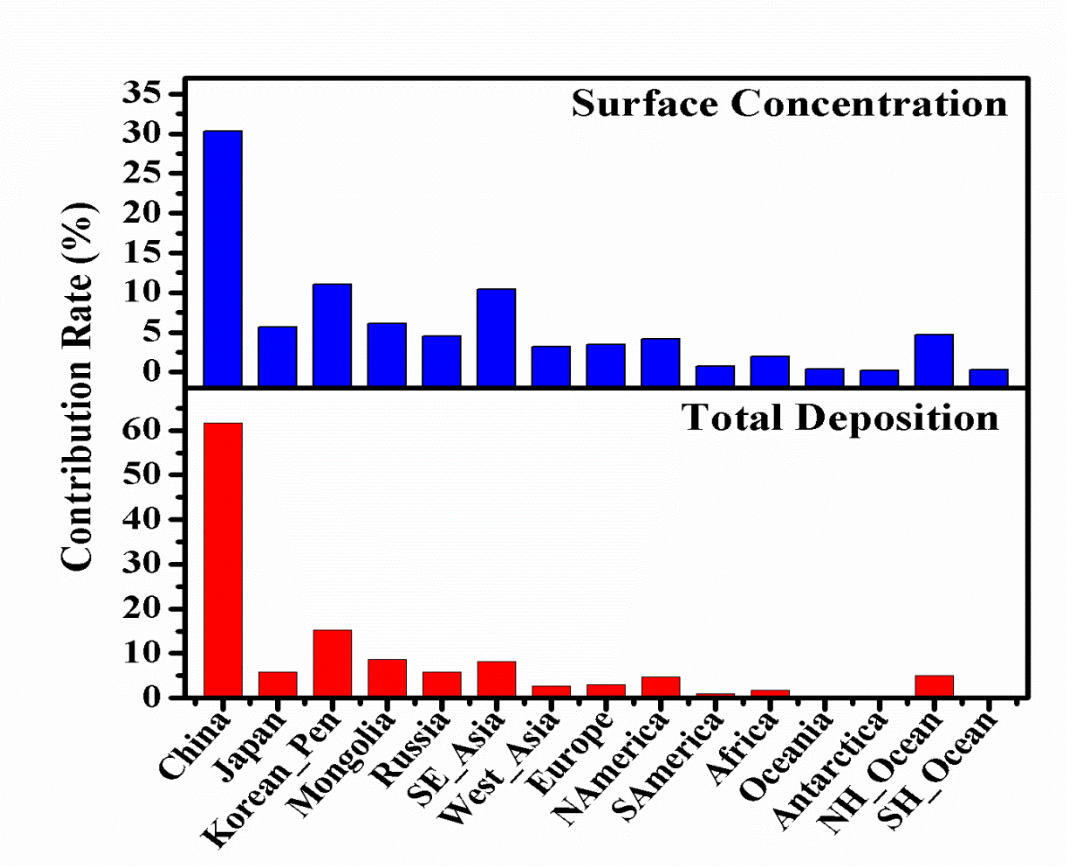


**Fig. 13.** Mean percentage contributions (%) from Chinese primary anthropogenic sources to annual_
mercury surface concentrations and total (wet plus dry) deposition over different world regions.

## The Supplement

# Supplementary Material: GNAQPMS-Hg v1.0, a global nested atmospheric mercury transport model: Model description, evaluation and application to trans-boundary transport of Chinese anthropogenic emissions

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Correspondence to: Z. F. Wang (zifawang@mail.iap.ac.cn)

## S1 Mercury chemistry

### S1.1 Bromine oxidation

As shown in Table S1, we add five Br chemical reactions in the gas phase (Seigneur and Lohman, 2008) in addition to the $O_3$-OH oxidation mechanism to test how the Br oxidation reactions affect the Hg distributions. Similar to the treatment of Holmes et al. (2006, 2010), the five reactions are treated as a single reaction, with an effective Hg(0) first-order rate constant that is a function of the individual reaction rates and the concentrations of Br, BrO and OH based on the assumption that Br, BrO and OH concentrations don't change by their reactions with Hg. This is also the same with the implementation described in CAMx (2014). The effective first-order rate constant is calculated as follows:

$$k_{eff} = \frac{k_1[Br](k_3[Br]+k_4[OH])}{k_2+k_3[Br]+k_4[OH]} + k_5[BrO] \quad \mathrm{s^{-1}}$$

In the GNAQPMS-Hg model, Br and BrO are not explicitly simulated. Therefore, we specify typical vertical profiles of Br and BrO concentrations over land and ocean, with higher values over ocean ($2.9\times10^{-8}$ and $2.9\times10^{-7}$ ppm for Br and BrO) than over land ($5.0\times10^{-9}$ and $5.0\times10^{-8}$ ppm for Br and BrO). During the night, the concentrations of Br and BrO are assumed to be zero, considering that the photolysis of $Br_2$ is the primary source for these radicals.

### S1.2 Gas-particle partitioning of Hg(II)

The mechanism of gas-particle partitioning of Hg(II) implemented in GNAQPMS-Hg is based on the studies of Rutter and Schauer (2007a, b). Similar mechanisms are also used by the CAMx model and Vijayaraghvan et al. (2008). Rutter and Schauer (2007a) suggest that surface area rather than particulate matter (PM) mass controls the Hg(II) partitioning process. The surface-area adsorption coefficient ($K_{sa}$) is calculated as follows:

$$K_{sa} = Hg_{p,ads}/(RGM \times A_{sp} \times PM)$$

where $K_{sa}$ is in $m^3$-$m^{-2}$, $Hg_{p,ads}$ and RGM are in pg $m^{-3}$, $A_{sp}$ is the specific surface area of PM in m$^2$ μg~~μ r~~$^{-1}$ and PM is the ambient urban PM concentration in μg m$^{-3}$. Further, they also found that the $K_{sa}$ obtained for urban PM falls between that of ammonium sulfate and adipic acid and it can be expressed as a function of temperature (K):

$$K_{sa} = 10^{\left(\frac{4250}{T} - 10\right)}$$

Besides, studies also found a ten-fold increase in adsorption of RGM to sodium chloride compared to ammonium sulfate and organic particulate compounds. Therefore, $K_{sa}$ for sea-salt is about 10 times that for urban PM:

$$K_{sa} = 10^{\left(\frac{4250}{T} - 9\right)}$$

For simplicity, we treat all non-sea-salt PM as urban PM and use the above two equations to simulate RGM adsorption to urban PM and sea-salt PM, respectively. Therefore, the effective adsorption coefficient for each aerosol size section is calculated as follows:

$$K_{sa,eff} = 10^{\left(\frac{4250}{T} - 9\right)} \times F_{ss} + 10^{\left(\frac{4250}{T} - 10\right)} \times (1 - F_{ss})$$

where is $F_{ss}$ the fraction of sea-salt in that size section.

## S2 Mercury deposition

### S2.1 Dry deposition

In the model, dry deposition is treated as a first-order removal mechanism. The deposition flux of a pollutant to the surface is the product of a characteristic deposition velocity and its concentration in the surface layer. Deposition velocities are derived from models that account for the reactivity, solubility, and diffusivity of gases, the sizes of particles, local meteorological conditions, and season-dependent surface characteristics. Dry deposition parameterizations of gases and aerosols are based on the work of Wesely (1989) and Slinn and Slinn (1980), respectively.

For gases, deposition velocity $V_d$ is calculated from three primary resistances $r$ (s m$^{-1}$)in series as described below.

$$V_d = \frac{1}{r_a + r_b + r_s}$$

The aerodynamic resistance $r_a$ represents bulk transport through the lowest model layer by turbulent diffusion. The quasi-laminar sub-layer resistance $r_b$ represents molecular diffusion through the thin layer of air directly in contact with the particular surface to which material is being deposited. The surface resistance $r_c$ depends upon the physical and chemical properties of the surface.

For particles, surface deposition occurs via diffusion, impaction, and gravitational settling. Particle size is the dominant variable controlling these processes. Particle deposition velocity for a given aerosol size is calculated using the following resistance equation.

$$V_d = V_{sed} + \frac{1}{r_a + r_b + r_a r_b v_{sed}}$$

$V_{sed}$ is the gravitational settling (or sedimentation) velocity which is dependent on aerosol size and density.

The detail formulations of how to calculate $\mathrm{r_a}$, $\mathrm{r_b}$, $\mathrm{r_s}$ and $\mathrm{V_{sed}}$ for gases and aerosols can be found in Wesely (1989) and Slinn and Slinn (1980) or the user's guide of the CAMx model (CAMx, 2014).

In the GNAQPMS-Hg model, dry deposition of Hg(0), Hg(II) and Hg(P) are all accounted for by adaption the parameterizations described above. Several physical properties (e.g. Henry's law constant, molecular weight, surface reactivity) of the Hg species are specified in order to calculate their deposition velocities. The Henry's Law constant for Hg(0) is set to be 0.11 M $\mathrm{atm}^{-1}$ (Lin and Pehkonen, 1999) with a temperature factor of -4970 K (Clever et al., 1985), and the surface reactivity is set to zero. Hg(II) represents $HgCl_2$ and $Hg(OH)_2$. Its Henry's Law constant is assumed to be the same as $HNO_3$ because they have similar solubility (Bullock and Brehme, 2002). Like $HNO_3$, Hg(II) has a strong tendency to stick to surfaces and its dry deposition occurs readily, so the surface resistance for Hg(II) in the dry deposition scheme is set to zero. The Hg(P) dry deposition velocity is set equal to that for sulfate, similar to that applied in the CMAQ-Hg and STEM-Hg model (Bullock and Brehme, 2002; Pan et al., 2008).

### S2.2 Wet deposition

In the model, wet deposition of the chemical species are calculated using an approach with medium complexity. In-cloud and below-cloud scavenging are included. The basic formulation implemented in the model is a scavenging approach in which the local rate of concentration change $\frac{\partial c}{\partial t}$ within or below a precipitating cloud depends on a scavenging coefficient $\Lambda$:

$$\frac{\partial c}{\partial t} = -\Lambda c$$

The scavenging coefficient is estimated differently for gases and particles, based on relationships described by Seinfeld and Pandis (1998). For gases, two components are calculated: 1) direct diffusive uptake of ambient gases into falling precipitation; and 2) accretion of cloud droplets that contain dissolved gases. For particles, there are also two components: 1) impaction of ambient particles into falling precipitation with an efficiency that is dependent upon particle size; and(2) accretion of cloud droplets that contain particle mass. Overall, the scavenging coefficient depends on an assumed scavenging efficiency, the total rainfall intensity (large-scale and convective precipitation), cloud water content and species solubility according to Henry's law, a mean cloud or rain droplet radius and rain droplet falling velocity. The large-scale and convective precipitation are not distinguished in this method. For species with low solubility (with a Henry's law constant of less than 100 M atm$^{-1}$), no wet deposition is calculated. More detail description of how to calculate the scavenging coefficients for gases and particles can be found in Seinfeld and Pandis (1998) or the user's guide of the CAMx model (CAMx, 2014). The physical properties (e.g. Henry's Law constant, surface reactivity, molecular diffusivity) of Hg species used in the wet deposition module are the same as those in the dry deposition module.

### S3 Mercury emissions

The AMAP 2000 anthropogenic emission inventory was used in the model simulation. However, the emissions over South Africa in this inventory were reported to be flawed (AMAP/UNEP, 2008). Here, we assessed the effects of these flawed emissions on the simulated results. As shown by Figure S7, we replaced the anthropogenic Hg emissions in South Africa by using the AMAP 2010 inventory and assessed this emission update on the simulated results. The emission amounts in South Africa decrease by about a factor of 4 (from 259 Mg to 64 Mg). After updating the emissions, the surface Hg concentrations in South Africa decrease by up to 1 ng $m^{-3}$, but have little changes elsewhere (the differences of concentrations are smaller than 0.01 ng $m^{-3}$ in most areas) as shown by Figure S8. The simulated TGM concentrations at Cape Point decrease from 1.77 ng $m^{-3}$ to 1.23 ng $m^{-3}$, more close to the observed values.

## S4 Model evaluation

### S4.1 Observational data

Due to limited public Hg observations, some model results were compared to observations with mismatched time periods. Actually, observations of wet deposition and precipitation in Europe and North America are from EMEP and MDN respectively, and the time periods are exactly the same with the simulation results. In contrast, no public Hg observation datasets are available in East Asia. So we used observations (collected from literatures) with mismatched time periods in East Asia.

All observations of Hg concentrations at land sites used in this study are averaged over time periods larger than 1 year. Analyses of long-term measurements show that trends in mean TGM during the last decade are small (of order $1\%a^{-1}$) or negligible at most background sites in the Northern Hemisphere (Temme et al., 2007; Wangberg et al., 2007). Therefore, the influences of the mismatch of time periods between model results and Hg concentration observations would not be large. Similar observational datasets (as shown in Table S2-S4) are also used by previous modeling studies (Selin et al., 2007, 2008; Holmes et al., 2010).

Observations from ship cruises are just used for initial comparison of simulated results over ocean following previous studies (Selin et al., 2007, 2008; Holmes et al., 2010). These observations are not used for quantitative model evaluation and not including in the calculation of statistical parameters of model performance.

Annual dry and wet deposition measurements in East Asia (Table S5) are all obtained from literatures. Considering that dry and wet deposition fluxes are affected by environmental factors (e.g. precipitation ) and they might differ from one year to another, so the influence of the mismatched time periods would be relatively larger. Again, no observations of Hg deposition are available at present. So there are no better choice.

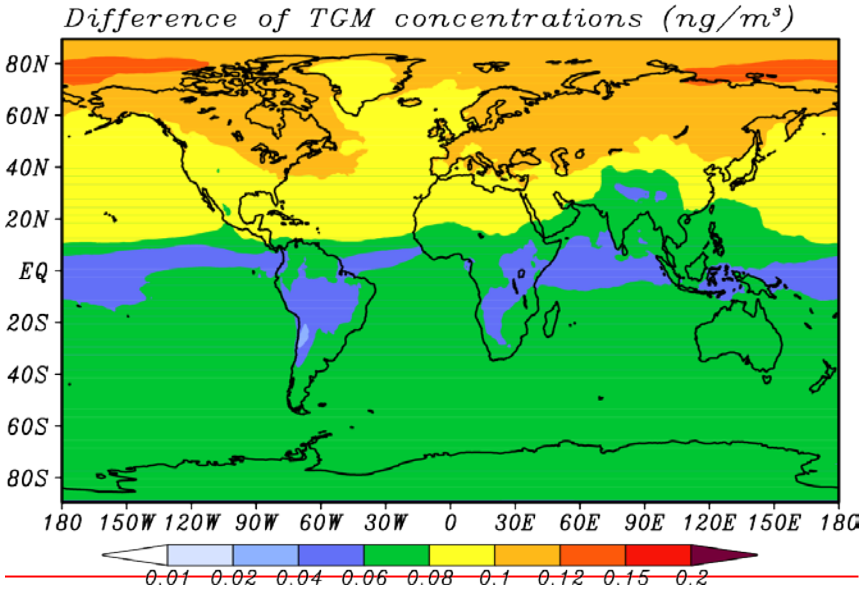
Overall, the influence of the mismatch of the time periods between model results and observations is relatively large for dry and wet deposition comparisons in East Asia but relatively small for other comparisons. Quantitative assessments of the influence are difficult and outside the scope of this study.

### S4.2 Diurnal and vertical variation

Fig. S12 compares the simulated and observed mean diurnal variations of TGM in St. Anicet, Delta, Mace Head, and Zeppelin. Detailed description of the observational sites can be found in Table S2. In St. Anicet, Mace Head, and Zeppelin, the TGM concentrations change little between daytime and nighttime and show no significant diurnal variations. While in Delta, the TGM concentrations are higher in daytime but lower in nighttime. These observed diurnal patterns are all well reproduced by the model. Fig. S13 shows the simulated averaged diurnal variation of surface TGM concentrations in Mt. Lulin (2862 m a.s.l.), Mt. Leigong (2178 m a.s.l.) and Mt. Changbai (741 m a.s.l.) in China. In Mt. Lulin, the simulated TGM concentrations exhibit a clear diurnal pattern, with higher concentrations in daytime but lower concentrations in nighttime. The daily maxima occurs in the afternoon. This simulated pattern is consistent with the observed results reported by Sheu et al. (2010). And they pointed out that this diurnal variation resulted from upslope movement of boundary layer air in daytime and subsidence of free troposphere air at night. The variation of TGM concentrations in Mt. Leigong is similar to that in Mt. Lulin and also agree with filed observations (Fu et al., 2010c). In Mt. Changbai, the diurnal variation of TGM shows different pattern. TGM peaks after sunrise and then decrease to the valley in the afternoon. Fu et al. (2012b) concluded that this variation was caused by regional transport.

Fig. S14 shows the vertical distribution of TGM concentrations derived from aircraft observations and model simulation over East Asia. The aircraft observations show two maxima, one near ground and the other at 6-7 km. The model reproduces the strong near ground TGM enhancement driven by continental outflow. However, the simulated TGM shows a monotonic decrease with altitude, and does not reproduce the observed 6-7 km enhancement. Friedli et al. (2004) pointed out that this high-altitude TGM enhancement was associated with lofted pollution from unknown sources or returning Asian plums after having circled the globe. Fig. S15 illustrates the simulated averaged vertical variation of TGM concentrations over the North Pacific Ocean (NPO) during April-May 2001. TGM concentrations over the south and north parts of NPO show similar vertical patterns. The highest concentrations are found at surface and then slowly decrease with altitude. At all levels, higher TGM concentrations occur over the north part of NPO which is related to long-range transport of Hg(0) from mainland in middle latitude. These simulated results agree with aircraft observations conducted in the INTEX-B filed experiment (Singh et al., 2009) and are also comparable to the simulated results from GEOS-Chem (Holmes et al., 2010).

The above comparisons confirm the model capability of simulating the diurnal and vertical variations of TGM.



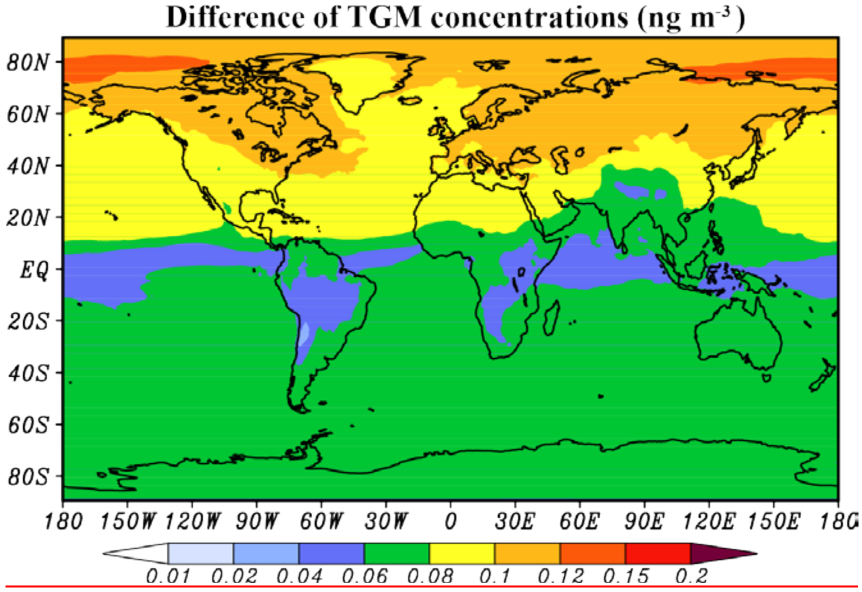


**Fig. S1.** Change in surface TGM concentrations (ng $m^{-3}$) by introducing bromine chemistry (positive value means the TGM concentrations decrease after added bromine chemistry).

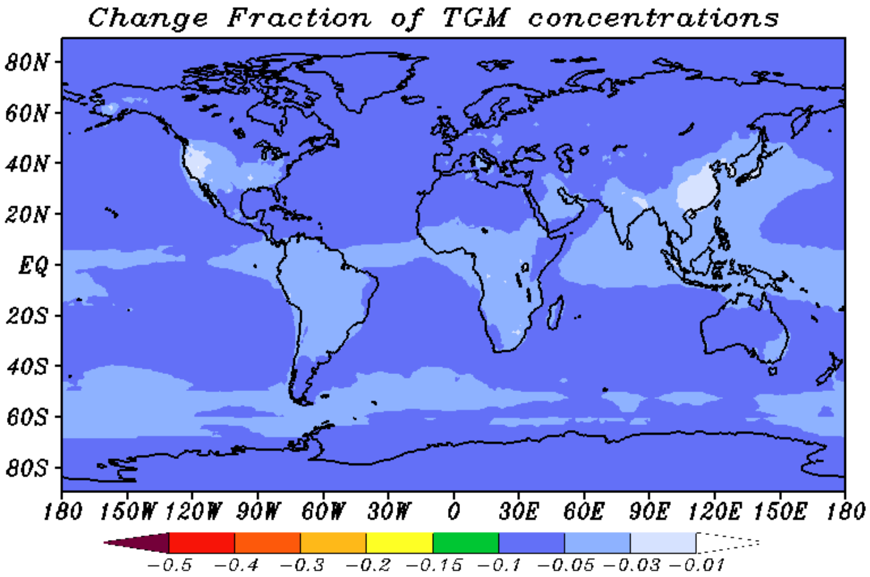


**Fig. S2.** Change fraction (unitless) of surface TGM concentrations by introducing the mechanism of gas-particle partitioning of Hg(II) (negative value means the TGM concentrations decrease after introducing the mechanism).

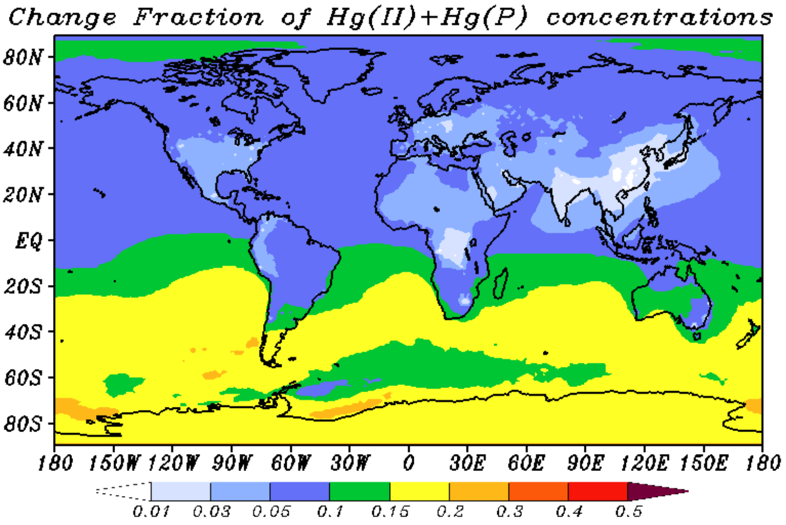
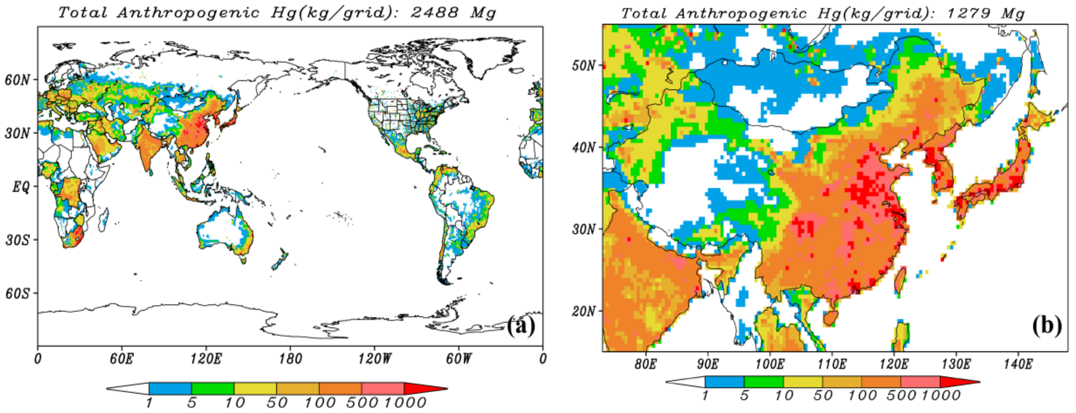


**Fig. S3.** Change fraction (unitless) of surface Hg(II)+Hg(P) concentrations by introducing the mechanism of gas-particle partitioning of Hg(II) (positive value means the Hg(II)+Hg(P) concentrations increase after introducing the mechanism).



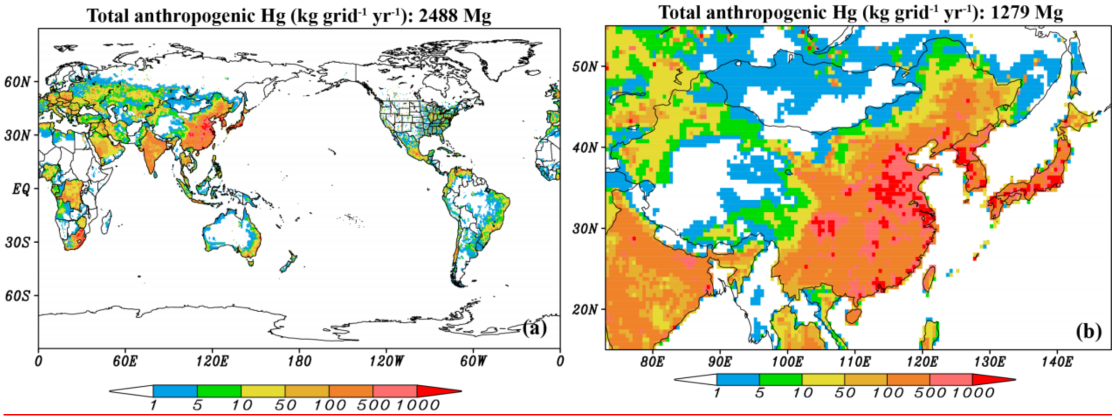
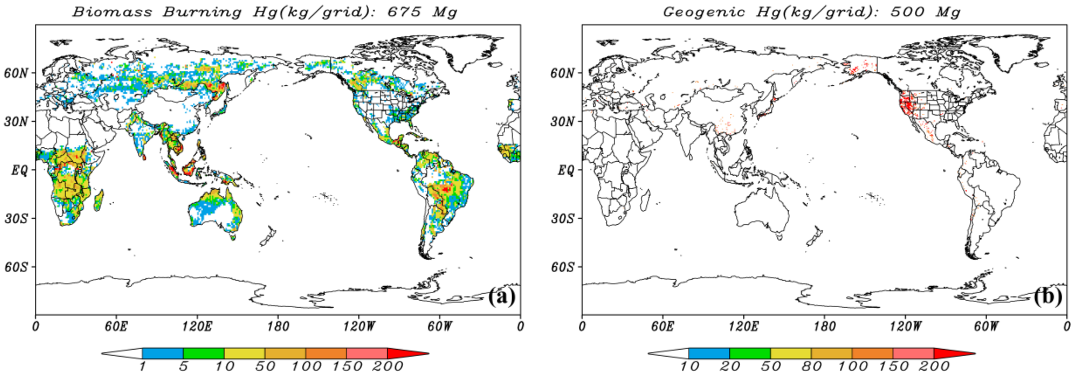


**Fig. S4.** Global (a) and East Asia (b) annual anthropogenic Hg emissions (kg ~~/~~grid$^{-1}$ yr$^{-1}$).



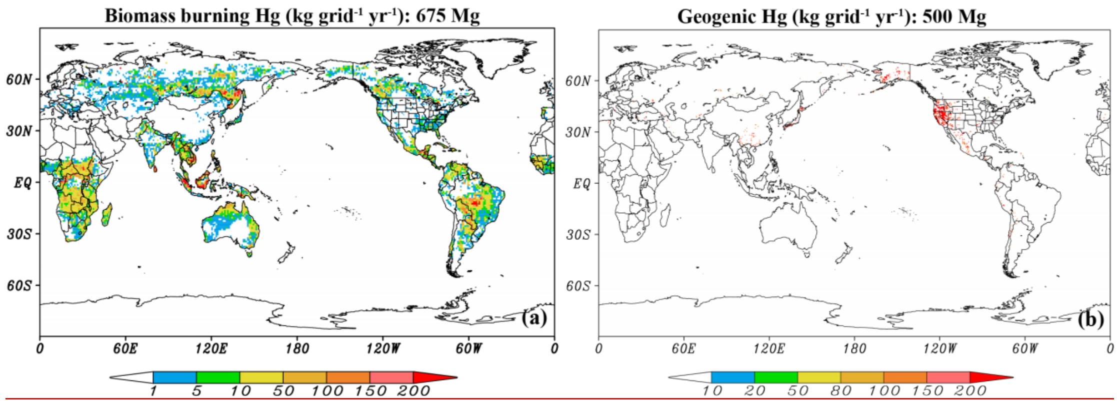


**Fig. S5.** Global annual biomass burning (a) and geogenic (b) Hg emissions (kg grid$^{-1}$ yr$^{-1}$~~kg/grid~~).

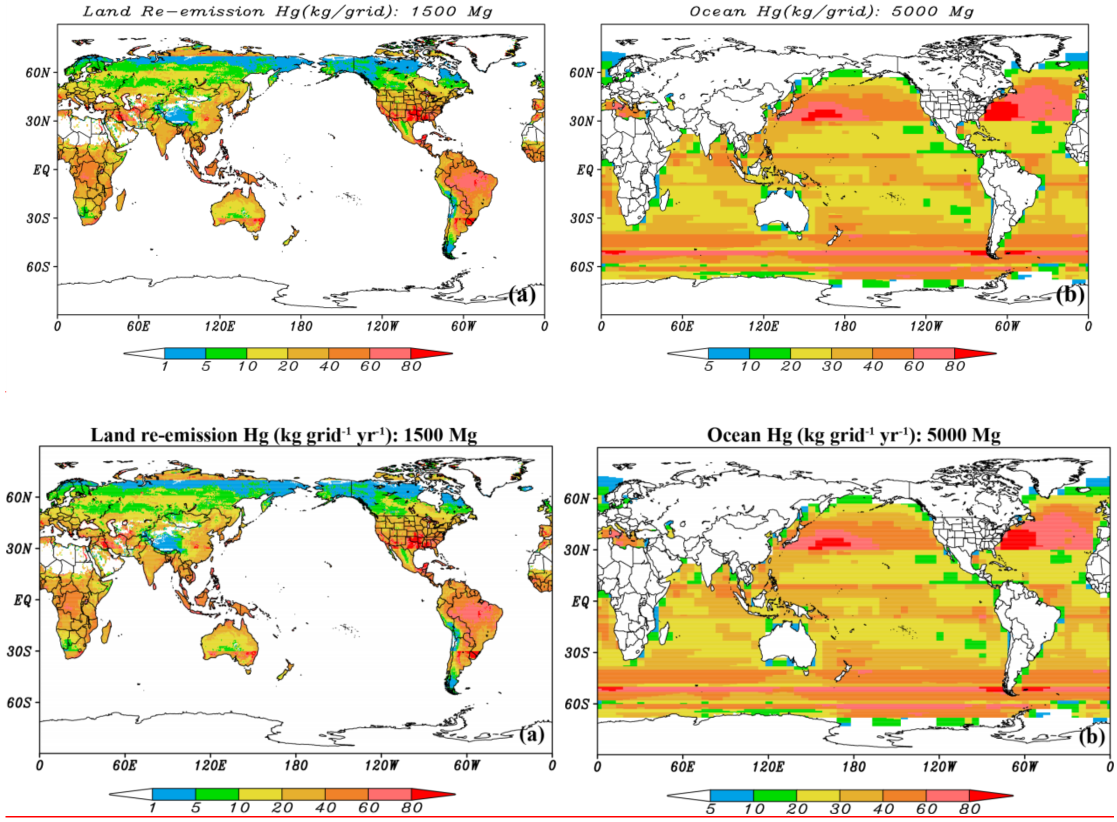


**Fig. S6.** Global annual land re-emission (a) and total ocean emissions (b) of Hg (kg grid$^{-1}$ yr$^{-1}$~~kg/grid~~).

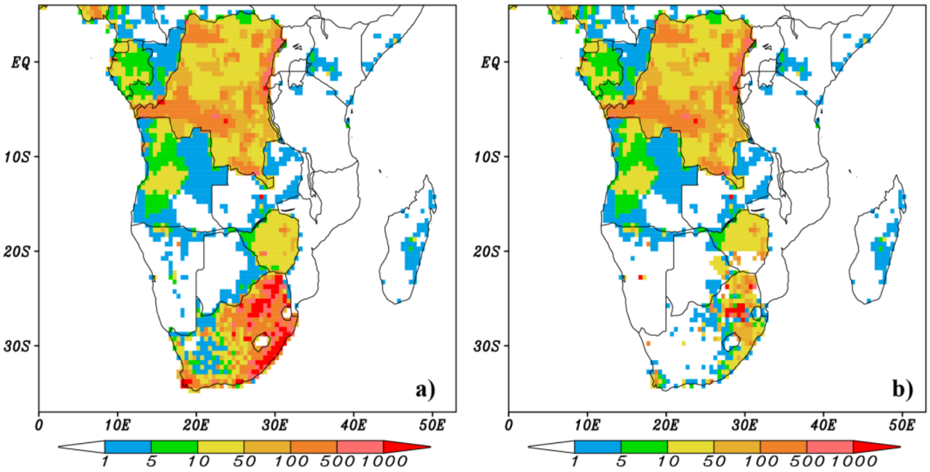


**Figure S7.** Annual anthropogenic Hg emissions (kg grid$^{-1}$ yr$^{-1}$~~kg/grid~~) in South Africa, a) the AMAP 2000 inventory and b) the AMAP 2010 inventory in South Africa (16-34$^{0}$E, 36-20$^{0}$S) + the AMAP 2000 inventory elsewhere.

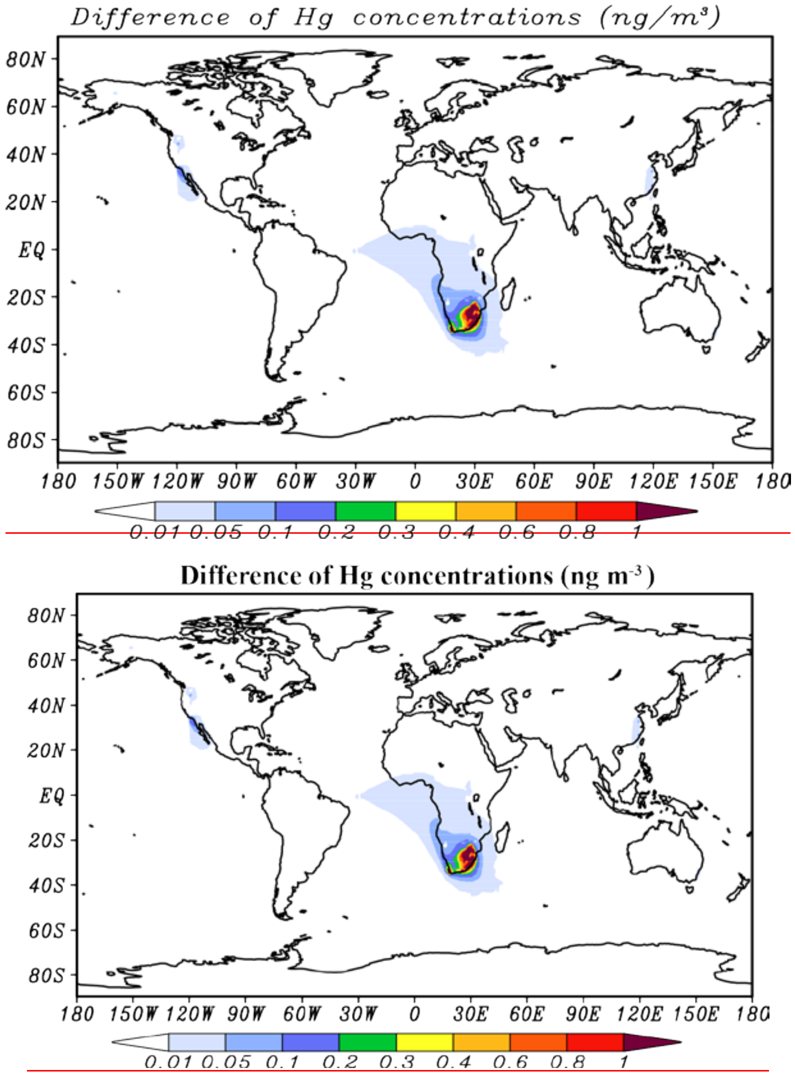


**Figure S8.** Difference of global surface Hg concentrations (ng m$^{-3}$) after updating the anthropogenic emissions in South Africa by using the AMAP 2010 inventory.

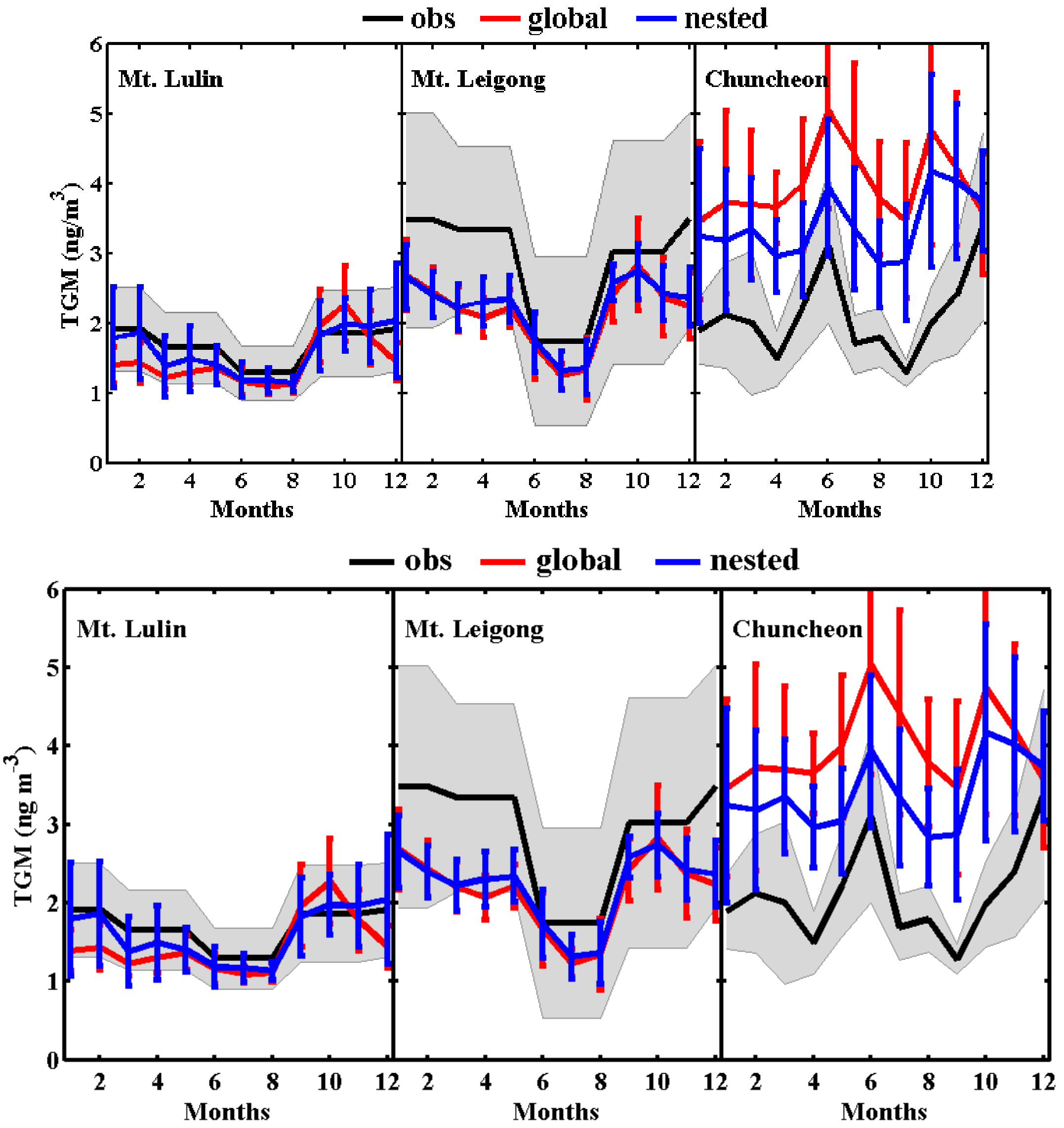


**Fig. S9.** Mean seasonal variation of TGM (ng m$^{-3}$) in Mt. Lulin and Mt. Leigong in China, and Chuncheon in Korea. Shaded areas and vertical bars show one standard deviation for observations and for model results.

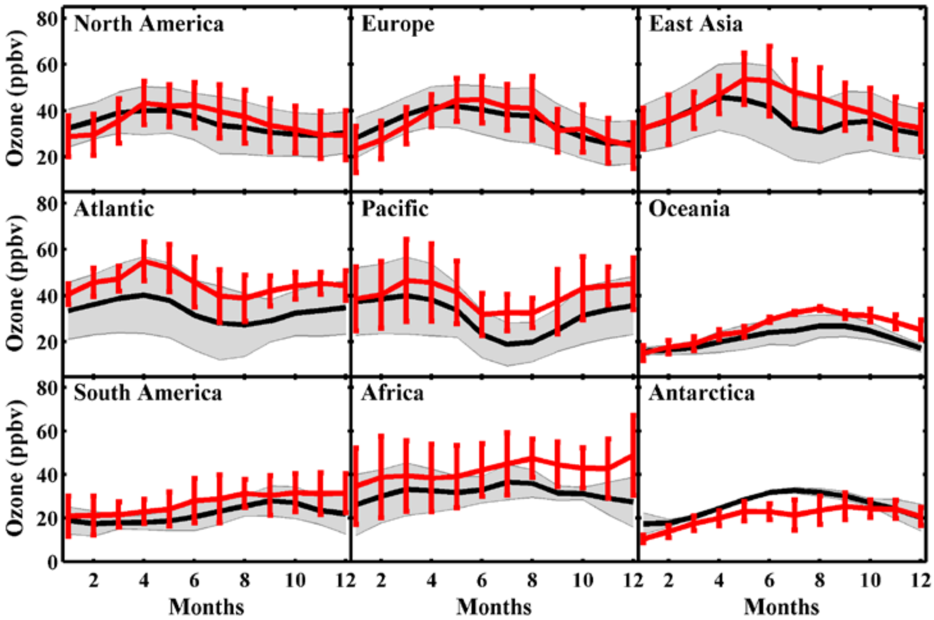


**Fig. S10.** Mean seasonal variation of surface ozone (ppbv) at 9 subregions in 2001. Gray shaded areas and red vertical bars show one standard deviation over the sites for observations and for model results. Observations are from the WDCGG (World Data Centre for Greenhouse Gases) and EANET (Acid Deposition Monitoring Network in East Asia) network.

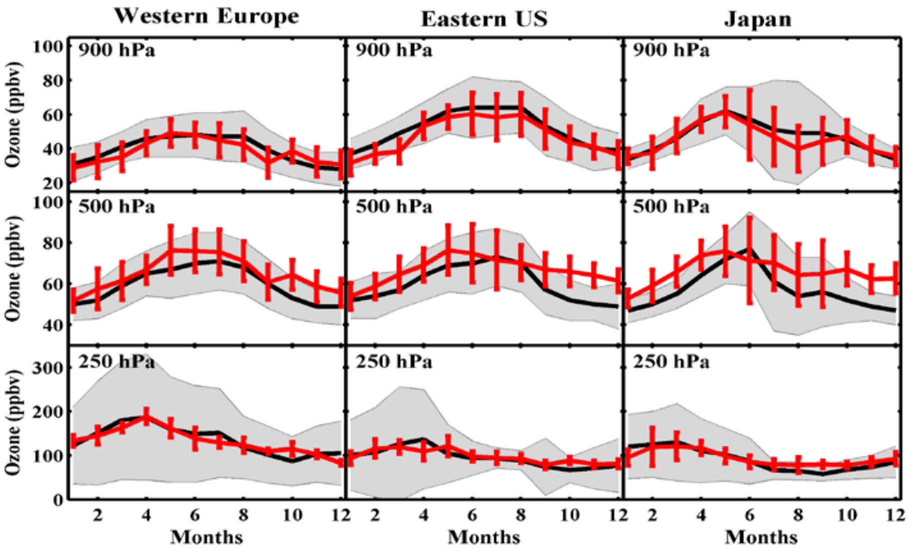
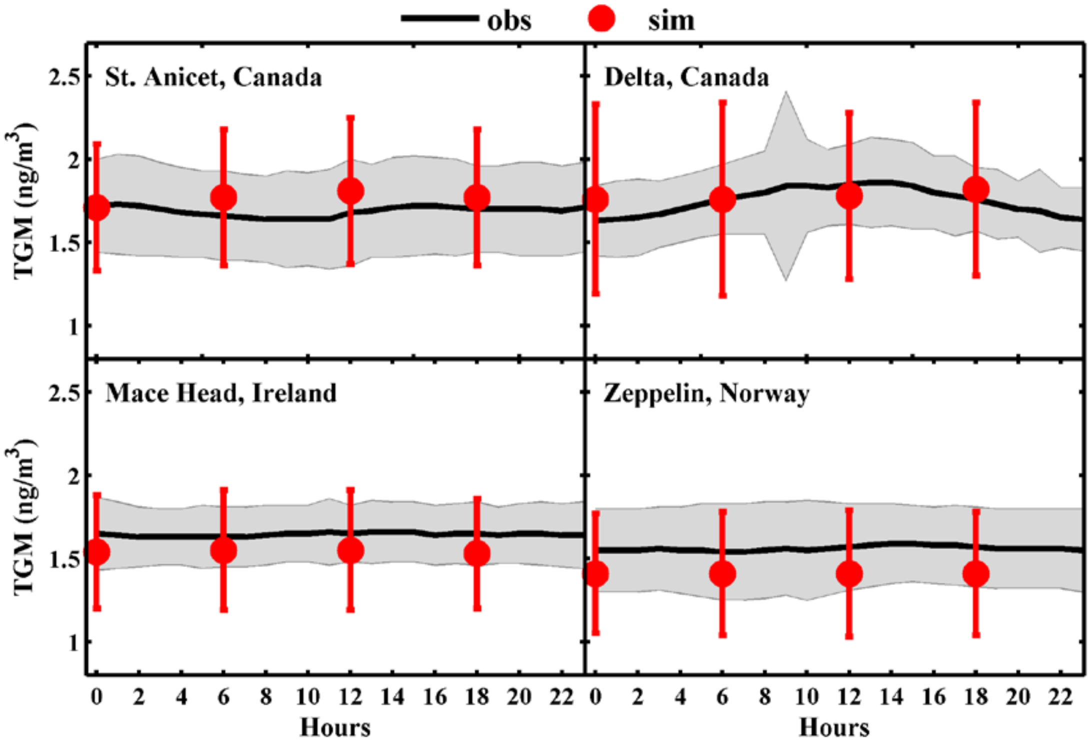


**Fig. S11.** Comparison of the ozone (ppbv) seasonal cycle between ozonesonde observations (black lines) and model results (red lines) in 900, 500 and 250 hPa in Western Europe, Eastern US and Japan. Gray shaded areas and red vertical bars show one standard deviation over the ozonesonde locations for observations and for model results. Observations are from Tilmes et al. (2012).



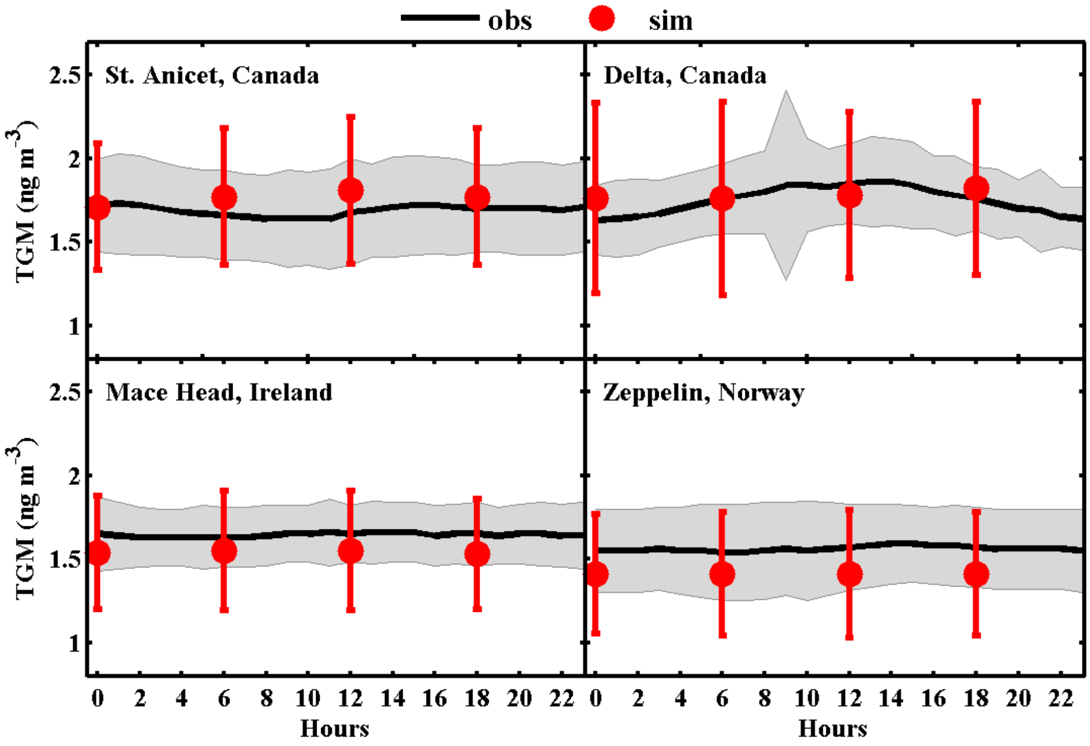


**Fig. S12.** Mean diurnal variation of TGM (ng m$^{-3}$) in St. Anicet, Delta, Mace Head, and Zeppelin in 2001. Gray shaded areas and red vertical bars show one standard deviation for observations and for model results. Note that the output frequency of the global simulation is 6 hours.

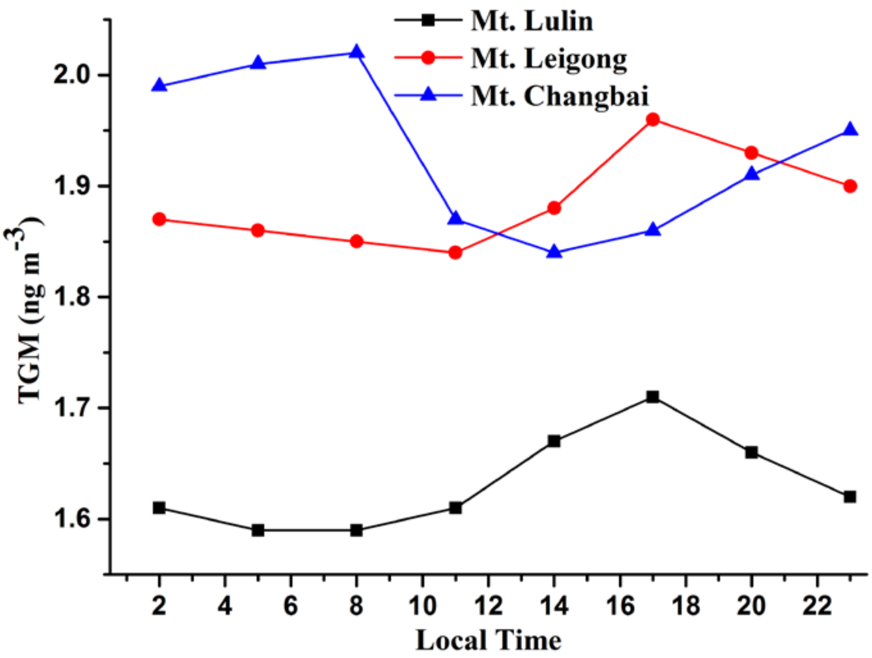


**Fig. S13.** Simulated averaged diurnal variation of surface TGM concentrations (ng m$^{-3}$) in Mt. Lulin,_–

Mt. Leigong and Mt. Changbai in 2001.

.

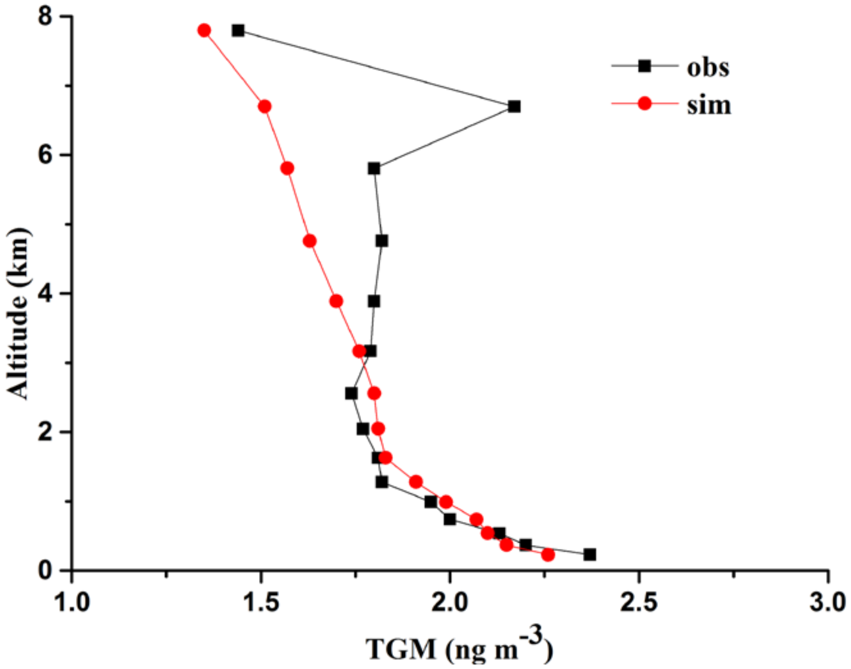


**Fig. S14.** Vertical variation of TGM concentrations (ng m$^{-3}$) over East Asia (24-42°N, 124-144°E) in April 2001. Observations are from the ACE-Asia aircraft campaign in April 2001 (Friedli et al., 2004). The observations are averaged according to each model layer height and depth.

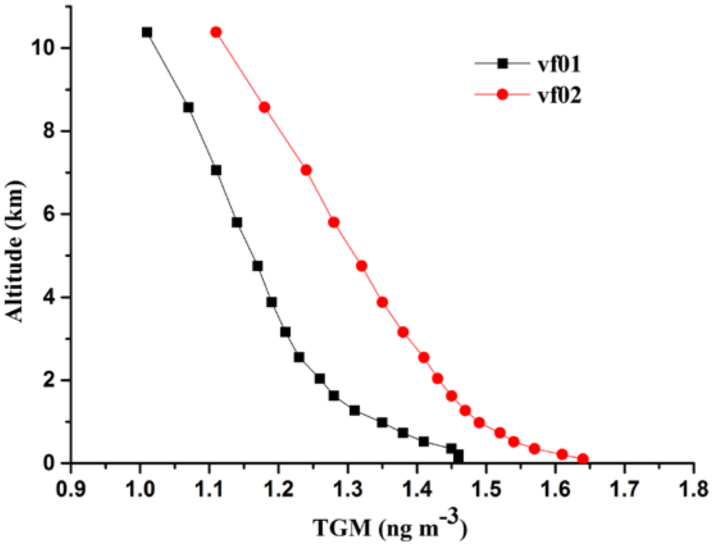
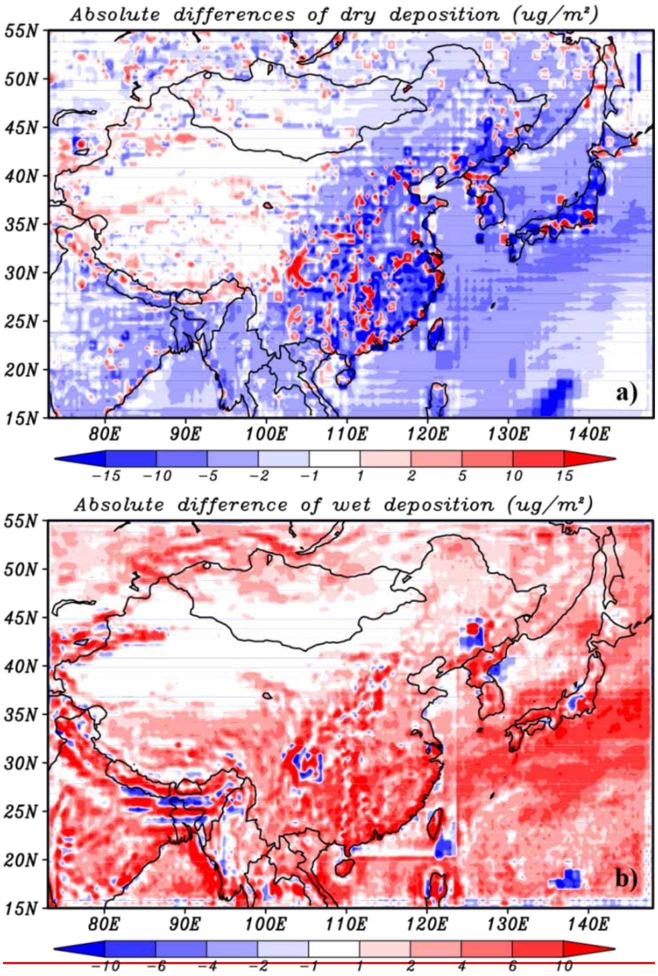
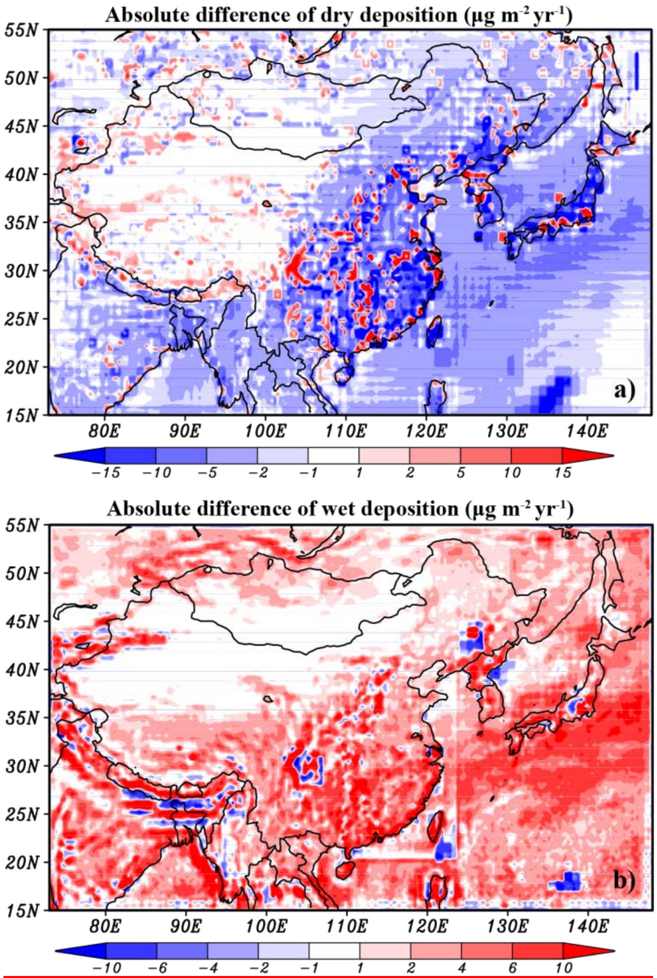


**Fig. S15.** Simulated averaged vertical variation of TGM concentrations (ng m$^{-3}$) over the North Pacific Ocean during April-May 2001. vf01 and vf02 stand for the vertical profile of TGM averaged over (19-25 °N, 120° E-120° W) and (30-55°N, 150°E-150°W) respectively.





Absolute difference of dry deposition (μg m$^{-2}$ yr$^{-1}$)

Absolute difference of wet deposition (μg m$^{-2}$ yr$^{-1}$)

**Figure S16.** Absolute differences of annual dry and wet deposition (μg m$^{-2}$ yr$^{-1}$) over East Asia between the global and nested simulations (Nested-Global).

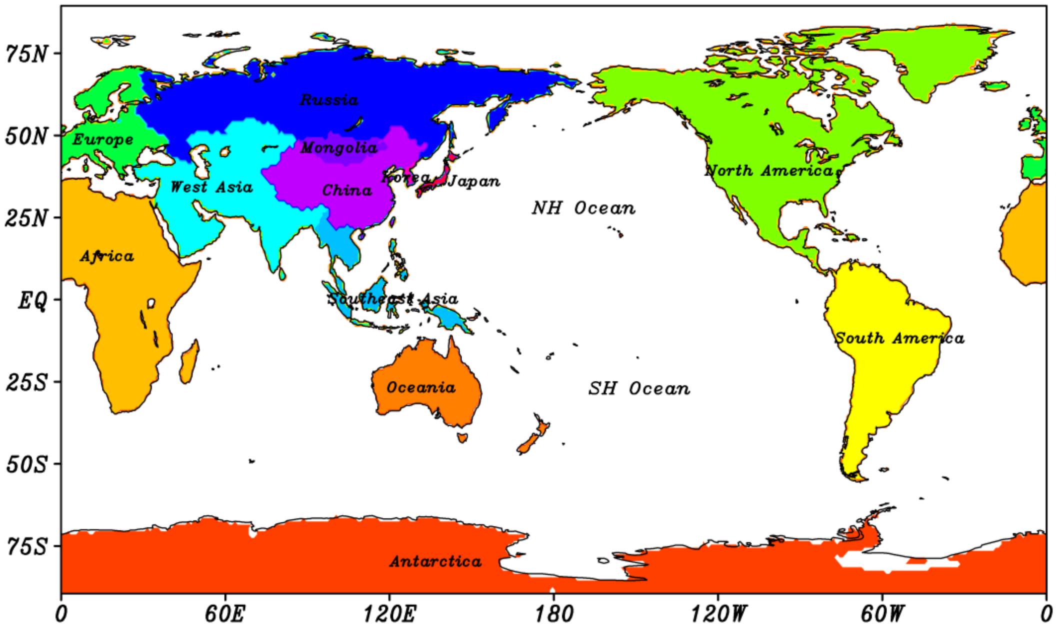


**Fig. S17.** 15 geographical areas used in this study, colors represent subregions over land while ocean is divided into two parts used the equator as boundary.

**Table S1.** Bromine reactions added in the model (T is the temperature in degrees Kelvin, and P is the pressure in atmospheres).

| NO. | Reaction | Rates |
|---|---|---|
| BR1 | $Hg(0)(g)+Br(g) \rightarrow HgBr(g)$ | $k1 = 3.6 \times 10^{-13} P \left(\frac{T}{298}\right)^{-1.86}$ $cm^3\ molec^{-1}\ s^{-1}$ |
| BR2 | $HgBr(g) \rightarrow Hg(0)(g)$ | $k2 = 3.9 \times 10^{9} exp\left(\frac{-8537}{T}\right)$ $s^{-1}$ |
| BR3 | $HgBr(g)+Br(g) \rightarrow HgBr_2(g)$ | $k3 = 2.5 \times 10^{-10} \left(\frac{T}{298}\right)^{-0.57}$ $cm^3\ molec^{-1}\ s^{-1}$ |
| BR4 | $HgBr(g)+OH(g) \rightarrow HgBrOH(g)$ | $k4 = 2.5 \times 10^{-10} \left(\frac{T}{298}\right)^{-0.57}$ $cm^3\ molec^{-1}\ s^{-1}$ |
| BR5 | $Hg(0)(g)+BrO(g) \rightarrow Hg(II)(g)$ | $k5 = 1.0 \times 10^{-15}$ $cm^3\ molec^{-1}\ s^{-1}$ |

**Table S2.** Long-term TGM/GEM measurements used for model evaluation (ng $m^{-3}$).

| **Site**[a] | **Years** | **Conc**[b] | **Reference** |
|---|---|---|---|
| Alert, Canada (83N, 62W)* | 1995-2002 | 1.55 | Environment Canada (2003) |
| FortChipewyan, Canada (59N, 111W) | 2000-2001 | 1.36 | Temme et al. (2007) |
| Kuujjuarapik, Canada (56N, 78W) * | 1999-2000 | 1.82# | Steffen et al. (2005) |
| Esther, Canada (52N, 110W) | 1997-1999 | 1.65 | Kellerhals et al. (2003) |
| Mingan, Canada (50N, 64W) | 1997-1999 | 1.62 | Kellerhals et al. (2003) |
| Bratts Lake, Canada (50N, 105W) | 2001-2005 | 1.53 | Temme et al. (2007) |
| Reifel Island, Canada (49N, 123W)* | 1997-1999 | 1.67 | Kellerhals et al. (2003) |
| Delta, Canada (49N, 123W) | 1999-2001 | 1.73 | Environment Canada (2003) |
| Burnt Island, Canada (46N, 83W) | 1997-1999 | 1.58 | Kellerhals et al. (2003) |
| St.Andrews, Canada (45N, 67W)* | 1997-1999,2001 | 1.42 | Environment Canada (2003) |
| St.Anicet, Canada (45N, 74W)* | 1997-1999,2001 | 1.64 | Kellerhals et al. (2003);<br>Poissant et al. (2005) |
| Kejimkujik, Canada (44N, 65W)* | 2001 | 1.45 | Environment Canada (2003) |
| Egbert, Canada (44N, 80W) | 1997-1999 | 1.67 | Kellerhals et al. (2003) |
| Pt.Petre, Canada (44N, 77W) | 1997-1999 | 1.78 | Kellerhals et al. (2003) |
| Cheeka Peak, USA (48N, 125W) * | 2001-2002 | 1.56 | Weiss-Penzias et al. (2003) |

| | | | |
|---|---|---|---|
| NewcombNY, USA (43N, 74W) [*] | 2006-2007 | 1.45 | Choi et al. (2008) |
| PacMonadnock, USA (43N, 72W) [*] | 2007 | 1.38[#] | Sigler et al. (2009) |
| RenoDRI, USA (40N, 120W)[*] | 2002-2005 | 2.10 | Stamenkovic et al. (2007) |
| AthensOH, USA (39N, 82W) [*] | 2004-2005 | 1.63[#] | Yatavelli et al. (2006) |
| PensacolaOLF, USA (31N, 87W) [*] | 2004-2006 | 1.34[#] | Edgerton et al. (2006) |
| Pallas, Finland (68N, 24E)[*] | 1998-2002 | 1.38 | EMEP (2005) |
| Zingst, Germany (55N, 13E)[*] | 2000 | 1.64 | EMEP (2005) |
| Neuglobsow, Germany (53N, 13E)[*] | 2004-2005 | 1.70 | EMEP (2009) |
| Langenbruegge, Germany (53N, 11E)[*] | 2002 | 1.98 | EMEP (2005) |
| Mace Head, Ireland (54N, 10W)[*] | 1995-2001 | 1.69 | Ebinghaus et al. (2002) |
| SanLucido, Italy (39N, 16E) | 2004-2005 | 1.80 | EMEP (2009) |
| Zeppelin, Norway (79N, 12E)[*] | 2000-2004 | 1.58 | EMEP (2005) |
| Andoya, Norway (69N, 16E)[*] | 2004 | 1.66 | EMEP (2009) |
| Birkenes, Norway (58N, 8E)[*] | 2005-2007 | 1.82 | EMEP (2009) |
| Lista, Norway (58N, 7E)[*] | 2000-2003 | 1.70 | EMEP (2005) |
| Amderma, Russia (70N, 62E) [*] | 2001-2003 | 1.66[#] | Steffen et al. (2005) |
| CaboDeCreus, Spain (42N, 3E)[*] | 2005 | 1.73 | EMEP (2009) |
| Rao, Sweden (57N, 12E)[*] | 2001 | 1.66 | EMEP (2005) |
| Rorvik, Sweden (57N, 12E)[*] | 2001-2002 | 1.66 | EMEP (2005) |
| Cape Point, South Africa (34S, 19E)[*] | 1998-2002,<br>2007-2008 | 1.22 | Baker et al. (2002);<br>Slemr et al. (2011) |
| Neumayer, Antarctica (70S, 8W)[*] | 2000 | 1.06 | Ebinghaus et al. (2002) |
| Changchun, China (44N, 125E) | 1999-2000 | 15.10 | Fang et al. (2004) |
| ChangbaiMt, China (42N, 129E) | 2005-2006 | 3.15 | Wan et al. (2009a) |
| Beijing, China (40N, 116E) | 2005 | 6.60[#] | Wang et al. (2007) |
| Chengshantou, China (37N, 123E) | 2007-2009 | 2.17[#] | Ci et al. (2011a) |

**Table S2.** Continued.

| **Site**[a] | **Years** | **Conc**[b] | **Reference** |
|---|---|---|---|
| Waliguan,China (36N, 101E) | 2007-2008 | 1.98 | Fu et al. (2012a) |
| Shanghai, China (31N, 121E) | 2008-2010 | 7.79 | Zhang et al. (2012) |
| GonggaMt, China (30N, 102E) | 2005-2006 | 3.89 | Fu et al. (2008a) |
| Chongqing, China (30N, 107E) | 2006-2007 | 6.74[#] | Yang et al. (2009) |
| Shangri-La, China (28N, 100E) | 2009-2010 | 2.59 | Zhang (2011) |
| Guiyang, China (27N, 107E) | 2001-2002 | 6.95 | Feng et al. (2004) |
| LeigongMt, China (26N, 108E) [*] | 2008-2009 | 3.03[#] | Fu et al. (2010c) |
| LulinMt, China (24N, 121E) [*] | 2006-2007 | 1.62[#] | Sheu et al. (2010) |
| Guangzhou, China (23N, 113E) | 2010-2011 | 4.86 | Liu et al. (2012) |
| Tokyo, Japan (36N, 140E) | 2000-2001 | 2.70 | Sakata and Marumoto (2002); |
| Chiba, Japan (36N, 140E) | 1991-1996 | 11.90 | Nakagawa and Hiromoto (1997) |
| Hayama, Japan (35N, 140E) | 1991-1996 | 13.20 | Nakagawa and Hiromoto (1997) |
| Chuncheon, Korea (38N, 127E)[*] | 2006-2009 | 2.11 | Holmes et al. (2010) |
| Seoul, Korea (37N, 127E) | 1997-2002 | 4.42 | Kim et al. (2005) |

|  |  |  |  |
|---|---|---|---|
| Kanghwa, Korea (37N, 126E) | 2008-2009 | 1.92 | Han et al. (2011) |
| An-Myun,Korea (37N, 126E) | 2005 | 4.27 | Nguyen et al. (2007) |
| Jeju Island, Korea (33N, 126E) | 2006-2007 | 3.58 | Nguyen et al. (2010) |

[a] Asterisk indicates that monthly mean observations are also available from the references.

[b] Pound sign indicates GEM measurements.

**Table S3.** Mercury measurements from ship cruises used for model evaluation.

| Cruise | Region | Date | Obs | Reference |
|---|---|---|---|---|
| Lamborg1999 | South and equatorial Atlantic Ocean | May to Jun, 1996 | TGM | Lamborg et al. (1999) |
| Temme2003 | Atlantic Ocean | Feb 2001 | TGM | Temme et al. (2003) |
| Laurier2003 | North Pacific Ocean | May to Jun, 2002 | TGM/RGM | Laurier et al. (2003) |
| Soerensen2010 | Notrh Atlantic, Indian Ocean, South Pacific | Aug 2006 to Apr 2007 | GEM | Soerensen et al. (2010) |
| Fu2010 | South China Sea | Aug 2008 | GEM | Fu et al. (2010a) |

| | | | | |
|---|---|---|---|---|
| Ci2011 | Yellow Sea | Jul 2010 | GEM | Ci et al. (2011b) |

**Table S4.** Long-term RGM and TPM measurements used for model evaluation (pg m$^{-3}$)$^{a}$.

| Site | RGM | TPM | Total | Period | Reference |
|---|---|---|---|---|---|
| St.Anicet, Canada (45N, 74W) | 3 | 26 | 29 | 2003 | Poissant et al. (2005) |
| Barrow, USA (71N, 157W) | 24 | NA | 24 | 1999-2001 | Landis et al. (2002) |
| Potsdam, USA (45N, 75W) | 4.2 | NA | 4.2 | 2002-2003 | Han et al. (2005) |
| Sterling, USA (43N, 77W) | 6 | NA | 6 | 2002-2003 | Han et al. (2005) |
| Stockton, USA (42N, 79W) | 5.7 | NA | 5.7 | 2002-2003 | Han et al. (2005) |
| Durham, USA (36N, 79W) | 16 | NA | 16 | 1999-2001 | Landis et al. (2002) |
| Baltimore, USA (32N, 77W) | 23 | NA | 23 | 1999-2001 | Landis et al. (2002) |
| Everglades, USA (26N, 81W) | 15 | NA | 15 | 1999-2001 | Landis et al. (2002) |
| Zingst, Germany (55N, 13E) | 25 | 22 | 47 | 1998-1999 | Munthe et al. (2003) |
| Neuglobsow, Germany (53N, 13E) | 18 | 25 | 43 | 1998-1999 | Munthe et al. (2003) |
| Mace Head, Ireland (54N, 10W) | 18 | 5 | 23 | 1998-1999 | Munthe et al. (2003) |
| Avspreten, Sweden (58N, 17E) | 8 | 9 | 17 | 1998-1999 | Munthe et al. (2003) |
| Rorvik, Sweden (57N, 12E) | 15 | 5 | 20 | 1998-1999 | Munthe et al. (2003) |
| Changchun, China (44N, 125E) | NA | 192.5 | 192.5 | 1999-2000 | Fang et al. (2004) |
| ChangbaiMt, China (42N, 129E) | 65 | 77 | 142 | 2005-2006 | Wan et al. (2009b) |
| Beijing, China (40N, 116E) | NA | 930 | 930 | 2003-2004 | Wang et al. (2006) |

| | | | | | |
|---|---|---|---|---|---|
| Waliguan,China (36N, 101E) | 7.4 | 19.4 | 26.8 | 2007-2008 | Fu et al. (2012a) |
| Hefei, China (32N, 117E) | NA | 330 | 330 | 2008-2009 | Wang (2010) |
| Shanghai, China (31N, 121E) | NA | 560 | 560 | 2004-2006 | Xiu et al. (2009) |
| GonggaMt, China (30N, 102E) | 6.2 | 30.7 | 36.9 | 2005-2006 | Fu et al. (2008b) |
| Chongqing, China (30N, 107E) | NA | 416 | 416 | 2005 | Wu (2006) |
| Shangri-La, China (28N, 100E) | 8.2 | 43.5 | 51.7 | 2009-2010 | Zhang (2011) |
| Guiyang, China (27N, 107E) | 35.7 | 368 | 403.7 | 2009 | Fu et al. (2011) |
| LulinMt, China (24N, 121E) | 12.1 | 2.3 | 14.4 | 2006-2007 | Sheu et al. (2010) |
| Seoul, Korea (38N, 127E) | 27.2 | 23.9 | 51.1 | 2005-2006 | Kim et al. (2009) |
| Tokyo, Japan (36N, 140E) | NA | 98 | 98 | 2000-2001 | Sakata and Marumoto (2002) |

[a] The sum of RGM and TPM is defined as total oxidized mercury and compared to the sum of Hg(II)+Hg(P) in the model.

**Table S5.** Long-term dry and wet depositions measurements in East Asia used for model evaluation (Units for deposition and precipitation are μg ~~ug~~ m$^{-2}$ yr$^{-1}$ and mm yr$^{-1}$).

| Site | Lat | Lon | Period | Dry | Wet | Prec | Reference |
|---|---|---|---|---|---|---|---|
| ChangbaiMt, China | 42.4 | 128.5 | 2005-2006 | 16.5 | 8.4 | 613 | Wan et al. (2009b) |
| Changchun, China | 43.8 | 125.4 | 1999-2000 | 131.8 | 108.0 | 567 | Fang et al. (2004) |
| Beijing, China | 40.1 | 116.3 | 2003 | 338.3 | NA | NA | Wang et al. (2006) |
| Shanghai, China | 31.4 | 121.4 | 2008-2009 | NA | 250.5 | 947 | Zhang et al. (2010) |
| Chongqin, China | 29.6 | 104.7 | 2005-2006 | 256.0 | 77.6 | 1403 | Wang et al. (2009) |
| GonggaMt, China | 29.6 | 102.2 | 2005-2007 | 66.4 | 26.1 | 1818 | Fu et al. (2010b) |
| Wujiang, China | 26.5 | 106.1 | 2006 | NA | 34.7 | 963 | Guo et al. (2008) |
| LeigongMt, China | 26.4 | 108.2 | 2005-2006,<br>2008-2009 | 44.0 | 16.2 | 1437 | Fu et al. (2010c)<br>Wang et al. (2009) |
| Bekkai, Japan | 43.4 | 145.1 | 2002-2003 | 4.4 | 5.8 | 1117 | Sakata and Marumoto (2005) |
| Hayakita, Japan | 42.7 | 141.6 | 2002-2003 | 5.2 | 7.1 | 882 | Sakata and Marumoto (2005) |
| Akita, Japan | 40.2 | 140.0 | 2002-2003 | 9.4 | 14.9 | 1576 | Sakata and Marumoto (2005) |

| | | | | | | | |
|---|---|---|---|---|---|---|---|
| Fukushima, Japan | 37.6 | 140.7 | 2002-2003 | 6.8 | 10.0 | 1599 | Sakata and Marumoto (2005) |
| Ishikawa, Japan | 37.2 | 136.9 | 2002-2003 | 6.6 | 14.2 | 2076 | Sakata and Marumoto (2005) |
| Tokyo, Japan | 35.6 | 139.6 | 2002-2003 | NA | 16.7 | 1912 | Sakata and Marumoto (2005) |
| Aichi, Japan | 35.0 | 137.5 | 2002-2003 | 13.2 | 13.1 | 1679 | Sakata and Marumoto (2005) |
| Hyogo, Japan | 34.8 | 134.8 | 2002-2003 | 8.2 | 14.0 | 1481 | Sakata and Marumoto (2005) |
| Hiroshima, Japan | 34.4 | 132.7 | 2002-2003 | 9.7 | 14.3 | 1624 | Sakata and Marumoto (2005) |
| Nagasaki, Japan | 33.3 | 129.7 | 2002-2003 | 8.3 | 17.7 | 2317 | Sakata and Marumoto (2005) |
| Korea | 35.9 | 127.8 | 2006-2008 | NA | 9.4 | 1068 | Ahn et al. (2011) |

**Table S6.** Hg budgets over East Asia (15-55°N, 75-145°E) in the global and nested simulations (Unit: Mg yr$^{-1}$).

| | global domain | nested domain | nested/global |
|---|---|---|---|
| Total Sources | 1461 | 1461 | 1.00 |
| anthropogenic | 979 | 979 | 1.00 |
| land | 269 | 269 | 1.00 |
| ocean | 213 | 213 | 1.00 |
| Total Sinks | 824 | 843 | 1.02 |
| Wet deposition | 182 | 278 | 1.53 |
| Dry deposition | 642 | 565 | 0.88 |
| TGM burden | 548 | 512 | 0.93 |

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