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Estimating mercury emission outflow from East Asia using CMAQ-Hg

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Abstract

East Asia contributes nearly 50% of the global anthropogenic mercury emissions into the atmosphere. Recently, there are concerns for the long-range transport of mercury from East Asia to North America, which may lead to enhanced dry and wet depositions in North America. In this study, we performed four monthly simulations (January, April, July and October in 2005) using CMAQ-Hg v4.6 in an East Asian model domain. Coupled with a mass balance analysis and a number of emission inventory scenarios, the chemical transport of atmospheric mercury, the seasonal mercury transport budgets and mercury emission outflow from the East Asian region were investigated. The total annual mercury deposition in the region for the modeling year is estimated to be 821 Mg, with 396 Mg contributed by wet deposition and 425 Mg contributed by dry deposition. Regional mercury transport budgets show strong seasonal variability, with a net removal of RGM (7~5 Mg mo^{-1}) and PHg (13~21 Mg mo^{-1}), and a net export of GEM (60~130 Mg mo^{-1}) from the study domain. The annual outflow caused by the East Asian emission is estimated to be in the range of 1369~1671 Mg yr^{-1}, primarily in the form of GEM. This represents about 75% of the total mercury emissions (anthropogenic and natural) in the region. The emission outflow from this source region would contribute to 20~30% of mercury deposition in areas remote from anthropogenic emission sources.

1 Introduction

Mercury is a global pollutant subject to long-range transport, due to the long atmospheric lifetime of gaseous elemental mercury (GEM, 0.5–2 years) (Selin et al., 2007; Schroeder and Munthe, 1998; Lin and Pehkonen, 1999; Shia et al., 1999; Lindberg et al., 2007). On the other hand, it can be quickly removed from the atmosphere via wet and dry deposition at its divalent oxidation state, either in the forms of reactive gaseous mercury (RGM) or particulate mercury (PHg) (Lindberg et al., 2002; Schroeder et al.,

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1998; Lin and Pehkonen, 1999; Stratton et al., 2001). Once deposited, its methylation and bio-accumulation in the food chain pose a threat to human health, particularly for the sensitive sub-populations (Rolfhus et al., 2003; Mason et al., 1995, 2006; Miller et al., 2007).

East Asia is the largest mercury source region in the world. It contributes to about 50% of all anthropogenic emissions to the atmosphere (Pacyna et al., 2006). Recently, there were a number of studies that reported experimental and modeling evidences of the long-range transport of mercury from East Asia to North America. For example, observational analysis using total mercury to carbon monoxide concentration ratio showed that the emission plumes from East Asia can be identified in the west coast of North America (Jaffe et al., 2005; Weiss-Penzias et al., 2006, 2007). Global model simulations estimated that the long-range transport contributes to 5~36% of total (e.g., dry and wet) deposition in North America depending on the locations, with an average of 16% (Jaffe and Strode, 2008; Seigneur et al., 2004). On the other hand, the impact of regional emission uncertainties (Wu et al., 2006; Shetty et al., 2008; Streets et al., 2005) on the source-receptor attribution estimates has not been addressed; and a detailed, quantitative assessment on the fate of atmospheric mercury in the region has not been made.

To answer the questions on how mercury emissions from East Asia may affect mercury concentration and deposition in other regions, a better understanding of the transport, transformation and deposition in the region is needed. However, few efforts have been made to address this issue, although preliminary measurement and modeling analyses have suggested that mercury emissions transporting out of the region may be significant (Pan et al., 2006, 2008; Friedli et al., 2004; Weiss-Penzias et al., 2007; Kim et al., 2009). One approach to study the regional emission outflow of air pollutants is to construct the mass budget of the pollutants of interest (Lamborg et al., 1995; Moussiopoulos et al., 2004). Coupled with a comprehensive modeling analysis, the sources, sinks, and the associated chemical transport pathways can be understood quantitatively.

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In this study, the mercury model of the USEPA Community Multi-scale Air Quality modeling system (CMAQ-Hg) (Bullock and Brehme, 2002; Byun and Schere, 2006) was applied to simulate the emissions, transport, and deposition of atmospheric mercury in a model domain covering the East Asian region. The model results were incorporated in constructing the mass budget of mercury for estimating the seasonal and annual mercury outflow caused by the emissions in the region. The annual outflow was estimated under three emission inventory (EI) scenarios to understand the impact of emission uncertainties. The seasonal trend of mercury chemical transport was investigated, and its implications were discussed. This work, to our knowledge, is the first modelling assessment effort on the regional chemical transport of atmospheric mercury in the region, and a part of the modeling efforts of the USEPA's Intercontinental transport and Climatic effects of Air Pollutants (ICAP) Program to understand the effect of emissions outside of the US to regional air quality via long-range transport.

2 Methods

2.1 Model domain and input data

2.1.1 Model domain

The ICAP East Asian domain is in a Lambert conformal projection centered at 34° N and 110° E. The domain contains 97×164 horizontal grid cells at a spatial resolution of 36 km with 14 vertical layers. The domain covers China and other parts of Asia, including Bhutan, Myanmar, Northeastern India, Bangladesh, Nepal, northern Laos, Vietnam, Japan, Taiwan, North and South Korea, and southern Mongolia. The study domain is shown in the subsequent visualization figures in the Results and discussion section.

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2.1.2 Meteorological data and modeling periods

Hourly meteorological fields were used for model simulations. To study the seasonal trend of mercury chemical transport, the simulations were performed for four seasonal months (January, April, July and October) in 2005. The meteorological data were prepared by the ICAP program using a Meso-scale Meteorological Model (MM5 version 3.7) (Grell et al., 1994). The quality-assured MM5 outputs were processed to the CMAQ-ready format using Meteorology-Chemistry Interface Processor (MCIP version 3.3) as described by Byun and Ching (1999). In the MCIP processing, the dry deposition velocities (V_{dep}) of GEM and RGM were calculated by using M3DRY deposition scheme (Pleim and Byun, 2004). The V_{dep} of sulfate aerosols were used as the surrogate for PHg (Bullock and Brehme, 2002).

2.1.3 Emission inventory

Anthropogenic mercury emission inventory in China was based on the work of Streets and coworkers (Streets et al., 2005; Wu et al., 2006) for the year 2001. The EI outside of China were based on the work by Pacyna and coworkers (Pacyna et al., 2006) for the year 2000. We recognized that the base years of the inventory data were not consistent with the modeling period. However, the EI represented the most updated data at the time when this study was conducted. The emission speciation followed the recommendations of Streets et al. (2005), with 56% as GEM, 32% as RGM, and 12% as PHg. Natural and re-emissions of GEM were processed following the approaches by Shetty et al. (2008).

To understand the impact of emission uncertainty on the outflow estimates, three EI scenarios were considered. The total mercury emission in the study domain for the three scenarios is summarized in Table 1. The base case included both anthropogenic and natural emissions as estimated by the above published works (denoted as “Base Case”). Since the anthropogenic emission has been thought to be underestimated (Shetty et al., 2008; Friedli et al., 2004; Weiss-Penzias et al., 2007), an

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inferred, scaled-up emission from inverse modeling results was also considered (Pan et al., 2007) (denoted as "Inferred" case). To understand the relative importance of anthropogenic to natural source contribution, a case considering only natural and re-emission was also performed (denoted as "Natural Only" case). The spatial distribution of the gridded Base-Case EI is shown in Fig. 1. The higher emission in the month of July is due to the higher surface temperature and solar radiation that drive a greater emission from natural processes. The EI for other criteria pollutants was based on the ICAP emission, which was originally based on NASA's study on the Transport and Chemical Evolution over the Pacific (Carmichael et al., 2003) with continuous updates for the 2005 modeling year.

2.1.4 Boundary and initial conditions

Boundary and initial conditions (BC/ICs) were re-projected from the outputs of a global 3-D chemical transport model, GEOS-Chem CTM (Selin et al., 2007) into the map projection of the study domain. To understand the effect of BCs on the estimated outflow caused by mercury emissions in the region, simulations using "background" BC/ICs were also performed (details see Sect. 2.3). The background BC/ICs assumed a GEM concentration of 1.2 ng m^{-3} and zero concentration of RGM and PHg.

2.2 Chemical transport models

The CMAQ-Hg version 4.6 was used for all simulations. The model components, science updates, and model uncertainty issues have been discussed in details earlier (Bullock and Brehme, 2002; Lin et al., 2006b, 2007; Pongprueksa et al., 2008). The Carbon Bond mechanism (CB05) was used as the gas-phase chemical mechanism to generate the concentrations of photochemical oxidants. The Rosenbrock solver (ROS3 in CMAQ CTM) was used as the chemical solver because of its flexibility (not mechanism specific). A global mass-conserving scheme (YAMO) was used for vertical and horizontal advection calculation, and the K-theory eddy diffusivity scheme

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was used for the vertical diffusion (documentation for these schemes is available at http://www.cmascenter.org).

2.3 Calculation of regional mercury budget

Mercury emission outflow from the domain is estimated by performing chemical transport budget calculations using the CMAQ-Hg simulation results for each modeling month. A schematic for the calculation of mercury transport budget is shown in Fig. 2. The change of mercury mass within the domain boundary over a simulation period is influenced by the mercury mass entering and leaving the domain boundary, the emissions from anthropogenic and natural sources, and the deposition controlled by the chemical and physical processes in the atmosphere (e.g., chemistry, scavenging, mixing, etc.). The net change within the domain boundary can be estimated as the difference between mercury masses at the beginning and at the end of the modeling period:

$$CM = FM - IM = I - O + E - D \quad (1)$$

where CM is the change of mercury mass, FM is the mercury mass in the domain at the end of the modeling period (final mass), IM is the mercury mass in the domain at the beginning of the modeling period (initial mass), I is the incoming mercury mass over the modeling period, O is the outgoing mercury mass over the modeling period, E is the emission input within the domain boundary over the modeling period, and D is the removal of mercury mass by deposition in the domain over the modeling period. All the terms are in Mg. Since the I and O terms represent the atmospheric transport into and out of the domain, and the transport budget (TB) can be defined as:

$$TB = I - O = FM - IM - E + D\,. \quad (2)$$

The transport budgets for GEM, RGM and PHg were calculated for each of the modeling months. A positive value of transport budget indicates a net removal of mercury

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mass in the domain (what's coming in is greater than what's going out); while a negative value indicates a net export of mercury from the domain.

The mercury outflow caused (or enhanced) by the mercury emissions within the domain (*OF*) can be considered as the difference in transport budget between when there is emission input and when there is no emission input, i.e.,

$$OF = TB_i - TB_o \quad (3)$$

where TB_i is the transport budget corresponding to one of the emission scenarios shown in Table 1, TB_o is the transport budget under zero emission input. Equation (3) estimates the actual mercury emission outflow from the domain, which is independent of the BC/ICs used in the simulations. To verify that the estimated mercury outflow is not influenced by BC/ICs, the outflow was estimated using two different sets of BC/ICs as described in Sect. 2.1.4. The estimated outflows (*OF*) were found to be identical for both BC/IC cases.

2.4 Data analysis

The CMAQ-Hg model outputs are in network common data format (netCDF). A suite of netCDF file operators developed by Zender and Mangalarn (2007) were used for the analysis of the CMAQ-Hg outputs. The Package for Analysis and Visualization of Environmental data (PAVE) version 2.3 (available at http://www.cmascenter.org) was used for data visualization.

3 Results and discussion

3.1 Simulated mercury concentration in East Asia and model verification

The spatial distribution of atmospheric mercury concentrations in the East Asian region was analyzed using the CMAQ-Hg model outputs of the base-case simulations (Table 1 and Fig. 1) and GEOS-Chem BC/ICs. The results are shown in Fig. 3 for each

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model month in terms of total average surface mercury concentration (ng m^{-3}). There are two important features in Fig. 3. One is that the average surface concentration resembles the spatial pattern of natural emission (Shetty et al., 2008) with a signature of large point source emissions. Mercury emissions from large point sources account for about 45% of mercury anthropogenic emissions in China. The hotspots caused by large point source emissions, including the elevated concentrations in the provinces of Liaoning, Hebei, Guangdong, Guizhou, Gansu, can be seen in the month of January (Fig. 3a) when the natural emission is the weakest. Most of the emissions are from coal combustion and the smelting processes of zinc and lead (Streets et al., 2005). In contrast to area and natural sources that release mercury in the surface layer only, emissions from point sources have higher temperatures and are released at higher altitudes. Therefore, they have a greater potential to enter the free troposphere for long-range transport. The other feature is that there is a strong concentration gradient from the East Asian continent to the Pacific Ocean, suggesting that circumpolar westerlies transport the mercury emissions from the source region into the Pacific.

The simulated concentrations of atmospheric mercury in the study domain range from 1.1 to 9.3 ng m^{-3}, with 85–99% constituted by GEM, depending on the locations. These results are consistent with the global model predictions (Seigneur et al., 2004; Selin et al., 2007; Strode et al., 2008). The fraction of RGM and PHg is typically greater at locations near large point sources due to the anthropogenic emission speciation (nearly 50% of mercury is emitted as RGM and PHg), and decreases rapidly away from emission sources because of their relatively shorter atmospheric lifetime. The simulated surface concentrations were compared to the observed concentrations in the East Asian region (Liu et al., 2002; Wang et al., 2006, 2007; Fang et al., 2001, 2004; Feng et al., 2004a; Yang et al., 2009; Xiu et al., 2009; Fu et al., 2008a,b; Wan et al., 2009a,b; Kim et al., 2009; Nguyen et al., 2007; Chand et al., 2008). We recognized that the timeframe of measurements reported in the literature is not synchronized with the model year and emission inventory base years in this study. However, based on the fact that the air mercury concentration did not change significantly over the past

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few years (Choi et al., 2009; Kim et al., 2009; Wan et al., 2009a,b) in the region, the model verification in terms of the magnitude of speciated mercury concentrations yields valuable insights on the model performance. The comparison is summarized in Table 2.

As seen in Table 2, the model predicted concentrations are generally consistent with observations. The simulated GEM concentrations at remote sites agree excellently with field observations. However, the model under-predicted GEM concentrations in most urban areas in China by missing the peak observed concentrations. This is because a regional model such as CMAQ-Hg is incapable to simulate those high concentrations measured at the ground stations due to the model assumption of instantaneous dilution of emitted plumes in the relatively coarse grids (36-km resolution). The uncertainties of mercury emissions, which have been thought to be under-estimated or miss certain emission sources (Pan et al., 2006; Weiss-Penzias et al., 2007; Friedli et al., 2004; Feng et al., 2004a, 2005), may also be important reasons in the under-prediction of GEM. Model predictions of RGM and PHg agree reasonably well with the observations made in China, but over-predict those observed in South Korea and at Cape Hedo, Japan (Table 2). Process analysis of model results revealed that the over-prediction of RGM and PHg in South Korea may be due to the uncertainty in the emission speciation. The somewhat over-predicted RGM and PHg at Cape Hedo was caused by the oxidation of GEM downwind of the source regions in China, and possibly the under-predicted dry deposition of RGM and PHg over water surface. The simulated NOx and VOCs concentration leaving the continent exhibits sufficiently strong photochemical activity to cause the oxidation, and the lower V_{dep} of PHg compared to that of RGM leads to the greater simulation PHg concentrations at the location.

3.2 Model-predicted mercury deposition in East Asia

The spatial distribution of dry and wet deposition of atmospheric mercury of the base-case simulations with GEOS-Chem BC/ICs are shown in Figs. 4 and 5 in terms of monthly cumulated deposition fluxes (normalized to $\mu g\,m^{-2}\,yr^{-1}$). The spatial distribu-

tion of dry deposition resembles the footprint of the source locations of anthropogenic emissions (Fig. 4). The greatest deposition occurs at the immediate proximity of the emission sources, mainly in the major urban areas of China, Korea, and Japan. The simulated dry deposition is typically in the range from 10 to 150 $\mu g\,m^{-2}\,yr^{-1}$, with values exceeding 300 $\mu g\,m^{-2}\,yr^{-1}$ near large point sources. The month of April appeared to have a slightly larger deposition compared to other months, a result caused by a combination of relatively low planetary boundary layer height and chemical oxidation of GEM (more discussion on the domain-wide deposition in Sect. 3.3).

The spatial distribution of wet deposition is highly correlated with the locations where precipitation occurs and has a very high seasonal variability (Fig. 5). The wet deposition of mercury typically ranges from 5 to 100 $\mu g\,m^{-2}\,yr^{-1}$ in the study domain. The total wet deposition is comparable but somewhat smaller than the total dry deposition (Table 3). This is consistent with the model assessments performed for North America that 30~50% of mercury deposition is through the wet processes (Lin et al., 2007). Among the four modeling months, the month of July has the greatest wet deposition because of the relatively greater precipitation and more significant chemical oxidation of GEM. The high wet deposition at the south border in July and October is caused by the high RGM concentration from the BCs in the south boundary in both months.

The scarcity of the observations for dry and wet mercury depositions limits the evaluation of model performance for the simulated mercury deposition. In China, wet deposition of 34.7 $\mu g\,m^{-2}\,yr^{-1}$ was reported in a rural area (Wujiang River Basin) in Guizhou in 2006 (Guo et al., 2008). Also, wet deposition of 152.4 $\mu g\,m^{-2}\,yr^{-1}$ and dry deposition of 165.8 $\mu g\,m^{-2}\,yr^{-1}$ in the urban area of Changchun from July 1999 to July 2000 were also reported (Feng et al., 2004b). The dry and wet deposition of mercury predicted by the model is at the same order of magnitude with the reported values (15~120 $\mu g\,m^{-2}\,yr^{-1}$ of wet deposition and 30~130 $\mu g\,m^{-2}\,yr^{-1}$ of dry deposition for the four months in Wujiang River Basin; and 35~260 $\mu g\,m^{-2}\,yr^{-1}$ of wet deposition and 75~480 $\mu g\,m^{-2}\,yr^{-1}$ of dry deposition for the four months in Changchun, Jilin).

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3.3 Regional mercury budget and outflow caused by the East Asian emissions

The regional mercury mass budgets of emission (E), deposition (D), and the total mercury mass at the beginning and the end of each modeling month (IM and FM) as defined in Eq. (1) were calculated for GEM, RGM and PHg from the CMAQ-Hg model data. These values, as well as their transport budgets estimated according to Eq. (2), are summarized in Table 3. From Table 3, it is clear that dry deposition is the primary removal mechanism for RGM and that wet deposition is the main removal mechanism for PHg, although the dry and wet removals are of the same order of magnitude on the four-month combined basis. GEM is primarily removed through dry deposition due to its low solubility in the aqueous phase and vegetation uptakes (Lin and Pehkonen, 1999; Lin et al., 2006a). The dry deposition of GEM has a very large seasonal variability, with the greatest removal in the month of July due to the decreased deposition resistance in the summer month. Assuming that the net deposition is three times the four-month sum, we estimated that a total annual mercury deposition in the East Asian domain for the modeling year 2005 is 821 Mg, with 396 Mg contributed by wet deposition and 425 Mg contributed by dry deposition.

For the four modeling months, the transport budget (Eq. 2) of GEM is consistently negative ($-128 \sim -62\,\mathrm{Mg\,mo^{-1}}$), while the transport budgets of RGM ($7 \sim 15\,\mathrm{Mg\,mo^{-1}}$) and PHg ($13 \sim 21\,\mathrm{Mg\,mo^{-1}}$) are consistently positive (Table 3). These indicate that there is a net mass of GEM transported out of the East Asian region and a net removal of RGM and PHg in the region, consistent with earlier findings by a global model (Strode et al., 2008). The GEM mass leaving the study domain shows a strong seasonal variability and is the greatest in the month of July ($128\,\mathrm{Mg\,mo^{-1}}$), mainly caused by the increased GEM emission from the natural processes. This is more than twice as much as the mass transporting out of the domain in the month of January ($62\,\mathrm{Mg\,mo^{-1}}$). The removal of RGM and PHg shows a weaker seasonal variability ($20 \sim 34\,\mathrm{Mg\,mo^{-1}}$ combined) compared to that of GEM export. The variability is mainly due to the wet deposition. The relatively smaller dry deposition variability is caused by the offset of

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boundary layer mixing with chemical oxidation of GEM. For example, although January has weaker GEM oxidation that leads to lower dry deposition, the shallower mixing layer height in the month causes greater RGM/PHg deposition because of less emission dilution. Summing up the transport budgets of GEM, RGM and PHg, there is an overall "export" of mercury from the East Asian region. The greatest transport quantity occurs in the month of July (99 Mg mo^{-1}), more than twice as much in the month of January (42 Mg mo^{-1}). On an annual basis, it is estimated that 835 Mg of mercury is transported out of the study domain, primarily in the form of GEM.

In the absence of mercury emission input, the mercury mass entering the model domain from the boundaries is readily removed due to chemical oxidation of GEM followed by dry and wet deposition. The model-estimated transport budget with zero mercury emission in January, April, July and October of the modeling year is 34, 48, 50, 45 Mg, respectively (all net removal). If mercury emission is considered in the model simulations, the transport budget changed from net removal to net export. This suggests that the mercury emissions within the domain not only offset the removal of incoming mercury mass from the domain boundaries, but also results in additional mercury mass leaving the domain.

The mercury outflow caused by the mercury emissions in the study domain under the three emission scenarios (Table 1) was estimated using Eq. (3). The results are shown in Fig. 6, which also exhibits a strong seasonal variability. Since the anthropogenic emission inventory in the "Base Case" has been thought to be underestimated, the estimated outflow in this case can be considered a lower limit of the emission outflow. The "Inferred" case used the scaled-up emission inventory (Pan et al., 2007), therefore the estimated outflow can be considered an upper limit. Assuming that the annual outflow is three times the four-month sum, the estimated East Asian mercury outflow caused by emissions is in the range of 1369~1671 Mg yr^{-1} in the modeling year 2005. With the emission scenario where no anthropogenic emission was considered ("Natural Only" case), the mercury outflow caused by natural emission amounts to 805 Mg yr^{-1}.

The difference in the estimated annual emission outflow between the "Base/Inferred"

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case and the "Natural Only" case is the outflow caused by anthropogenic mercury emission in the region. For the Base case, the estimated anthropogenic emission outflow is 564 Mg yr^{-1}; while the outflow for the Inferred case is 866 Mg yr^{-1}. These correspond to 68% (Base) and 62% (Inferred) of the anthropogenic emissions in the study domain. Overall, we estimated that about 75% of mercury emissions in the East Asian region result in being transported out of domain (76% for the Base case; 74% for the Inferred case; 83% for the Natural Only case).

3.4 Implications on intercontinental and trans-boundary transport

Trans-Pacific and trans-boundary mercury transport events have been reported in a number of recent studies (Jaffe et al., 2005; Jaffe and Strode, 2008; Strode et al., 2008; Kim et al., 2005, 2009). Our earlier modeling assessment of the trans-Pacific transport of mercury showed that the direct transport of the Asian mercury plumes to North America is insignificant because the plumes have been much diluted during the process of long-range transport (Lin et al., 2006a). However, the mercury input into the atmosphere from the mercury emission outflow of the study region is significant. Based on the assessment in this study, the estimated annual emission outflow from the region is in range of 1369 to 1671 Mg yr^{-1}, mainly in the form of GEM. Considering the current global mercury burden of 5000~6000 Mg in the atmosphere and the life-time of atmospheric mercury at about one year (Mason and Sheu, 2002; Selin et al., 2007), the estimated emission outflow represents about 20~30% of the global burden of atmospheric mercury. Since the dry deposition of mercury is linear with respect to the concentration of GEM (Pongprueksa et al., 2008), the input to the global mercury pool from the East Asian emission outflow would contribute to about 20~30% of the deposition in areas remote from mercury emission sources. This estimate seems to be consistent with the results of the global model estimate of 18~26% (Strode et al., 2008).

The work by Kim et al. (2009) suggested that the elevated mercury concentration observed in Seoul, South Korea was caused by the anthropogenic plumes emitted in

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industrial areas of China in 25 of the 70 observed elevated concentration episodes at a ground station (34 episodes caused by local events) (Kim et al., 2009 and the references cited therein). Interestingly, based on the monthly averaging concentration and deposition results from the CMAQ-Hg simulations performed in this study, we did not observe consistent transport events from China to Korea. This is evidenced by two observations: (1) the simulated surface concentration fields do not exhibit a gradient from the industrial areas (e.g., Liaoning province) in China to the Korean region in any of the modeling months for the Base (Fig. 3) and the Inferred cases, and (2) the observed GEM concentration at the Changbaisan site (at the border of China and Korea peninsula, 3.58 ng m^{-3} mean surface concentration) (Wan et al., 2009a,b) is not significantly higher than the observed average surface concentrations in Seoul, South Korea (3.22 ng m^{-3} mean surface concentration) (Kim et al., 2009). The aircraft observations of mercury emission outflow during ACE-Asia campaign also showed that high mercury concentrations were observed at higher altitudes (>3 km above sea level) instead of near ground (Pan et al., 2006). These results imply that, even though the trans-boundary mercury transport events may occur, the transport events would be episodic. The impact of such episodic transport events on dry and wet mercury depositions at the receptor sites, which is the primary concern of mercury pollution, requires further modeling assessment.

4 Conclusions

In this study, we investigated the regional chemical transport of atmospheric mercury in the East Asian region and the mercury emission outflow from the region using a comprehensive modeling approach coupled with regional mass balance analysis. This study marks the first regional modeling assessment in the region that accounts for nearly 50% of the global anthropogenic mercury emissions.

The simulated base-case mean monthly concentrations resemble the spatial distribution of GEM emissions from natural sources, with concentration hotspots caused by

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large point sources. The concentration gradient from the Asian continent to the Pacific suggests significant mercury outflows. In the region, the dry deposition is mainly contributed by RGM while the wet deposition is contributed by both RGM and PHg. The annual total mercury deposition in the East Asian region for the modeling year (2005) is estimated to be 821 Mg, with 396 Mg contributed by wet deposition and 425 Mg by dry deposition.

Regional mercury mass budgets show strong seasonal variability, with a net removal of RGM (7~5 Mg mo^{-1}) and PHg (13~21 Mg mo^{-1}) in the study domain, and a net export of GEM (60~130 Mg mo^{-1}) from the domain for all four modeling months. The estimated annual emission outflow from the East Asian region in the modeling year is in the range of 1369~1671 Mg yr^{-1}, primarily in the form of GEM. The emission outflow represents about 75% of total mercury emissions (anthropogenic plus natural). About 65% of anthropogenic mercury emissions result in outflows, mainly into the Pacific.

The outflow caused by mercury emissions from the East Asian region contributes significantly to the global background concentration of mercury in the atmosphere. Since mercury deposition in regions remote from mercury emission source locations is linear to the ambient concentration of GEM, the mercury outflow can contribute considerably to deposition in areas remote from the anthropogenic source regions. We estimate that the mercury outflow from East Asia would contribute to 20~30% of mercury deposition in other regions. Major uncertainties of this assessment include mercury chemical mechanisms and mercury speciation of the anthropogenic emission estimates. Further understanding of mercury chemistry and emission processes will greatly reduce the uncertainties.

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Table 1. Mercury emission scenarios considered in the study domain (Mg yr^{-1}).

Scenarios	GEM	RGM	PHg	Domain Total	Anthrop. Emission
Base Case[a]	1249	275	89	1793	826
Inferred[b]	1669	434	155	2258	1390
Natural Only[c]	968	0	0	968	0

[a] Anthropogenic emissions from Wu et al. (2006) and Pacyna et al. (2006), plus natural emission.
[b] Anthrop. emission from the scaled-up values from inverse modeling (Pan et al., 2007), plus natural emission.
[c] Natural emission based on the method by Shetty et al. (2008).

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Table 2. Comparison of the base-case model results with the field observations in the East Asian region.

Station	Location	Category	Observations			Date/	Model Results			Ref.[a]
			GEM(ng/m^3)	RGM(pg/m^3)	PHg(pg/m^3)	Period	GEM(ng/m^3)	RGM(pg/m^3)	PHg(pg/m^3)	
Waliguan, China	100.9 E 36.3 N	Remote	1.7±1.1 0.6±0.1			Dec-05 Aug-05	1.46(1.16~3.18)			1
Yangzi River Delta, China	120.7 E 30.8 N	Suburban	5.4±4.1			Sep-05	2.49(1.36~8.06)			1
Guangzhou, China	113.3 E 23.2 N	Urban	13.5±7.1			Jan-05	3.15(1.46~9.93)			1
Beijing, China	116.4 E 39.9 N	Urban	8.3±3.6 6.5±5.2 4.9±3.3 6.7±3.5		180~3510	Jan-05 Apr-05 Jul-05 Oct-05	2.38(1.35~4.80) 2.50(1.28~6.68) 3.36(1.76~7.75) 2.88(1.38~6.83)		751(17.1~2357)	1, 2
Beijing, China	116.7 E 40.0 N	Rural	1.8~4.6			Feb-98	2.67(1.28~9.17)			3
Changchun, China	125.3 E 43.8 N	Urban Suburban	18.4(4.7~79.6) 11.7(2.3~25.6)		22~1984	Jul-09~ Jul-00	1.84(1.20~4.9)		258(0.02~695)	4, 5
Guiyang, China	106.7 E 26.6 N	Urban	8.4±4.9			Nov-01~ Nov-02	2.83(1.20~5.65)			6
Chongqing, China	106.5 E 29.5 N	Urban	6.74±0.37			Aug-06~ Sep-07	2.81(1.37~5.22)			7
Shanghai, China	121.4 E 31.1 N	Urban			70~1450	Jul-04~ Apr-06			230(0.01~1960)	8
Mt. Gongga, China	102.7 E 29.9 N	Remote	4.0(0.5~21.0)	6.2	30.7	May-05~ Apr-06	2.20(1.20~8.28)	31.4(0.03~116)	75.8(0.03~482)	9,10
Changbaisan, China	128.3 E 42.2 N	Remote	3.58±1.78	65.0	77.0	Aug-05~ Jul-06	1.57(1.19~2.80)	25.3(0.02~118)	129(0.01~488)	11,12
Seoul, Korea	127.0 E 37.5 N	Urban	3.22±2.1	27.2±19.3	23.9±19.6	Feb-05~ Feb-06	3.68(1.36~8.17)	532(0.01~1766)	403(0.03~1641)	13
An-Myum Island, Korea	126.3 E 36.5 N	Rural	4.6±2.2			Dec-04~ Apr-06	1.60(1.20~5.54)			14
Cape Hedo, Japan	128.2 E 26.8 N	Remote	2.04±0.38	4.5±5.4	3.0±2.5	Mar-04~ May-04	1.46(1.20~2.32)	20.6(0.02~94.8)	52(0.02~274)	15

[a]: 1: (Wang et al., 2007); 2: (Wang et al., 2006); 3: (Liu et al., 2002); 4: (Fang et al., 2004); 5: (Fang et al., 2001); 6: (Feng et al., 2004a); 7: (Yang et al., 2009); 8: (Xiu et al., 2009); 9: (Fu et al., 2008a); 10: (Fu et al., 2008b); 11: (Wan et al., 2009b); 12: (Wan et al., 2009a); 13: (Kim et al., 2009); 14: (Nguyen et al., 2007); 15: (Chand et al., 2008).

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Table 3. Mercury mass budget in the study domain (Mg mo^{-1}).

Model Month	January			April			July			October		
Species	GEM	RGM	PHg	GEM	RGM	PHg	GEM	RGM	PHg	GEM	RGM	PHg
Initial Hg Mass	366.2	14.4	22.3	370.6	17.4	22.7	404.0	12.2	11.7	399.7	11.6	12.3
Final Hg Mass	369.3	14.2	21.3	400.3	15.3	18.9	439.6	11.7	10.6	375.8	14.0	18.3
Emissions	67.7	22.8	7.5	114.7	22.0	7.3	181.2	22.8	7.5	112.6	22.8	7.5
Wet deposition	0.003	7.6	18.7	0.005	10.8	25.2	0.016	16.9	23.2	0.008	13.1	16.6
Dry deposition	2.5	22.2	3.1	9.3	26.4	6.4	17.6	18.0	2.4	9.0	22.4	2.3
Transport Budget	−62.1	6.8	13.3	−75.7	13.1	20.5	−128.0	11.6	17.0	−127.5	15.1	17.4

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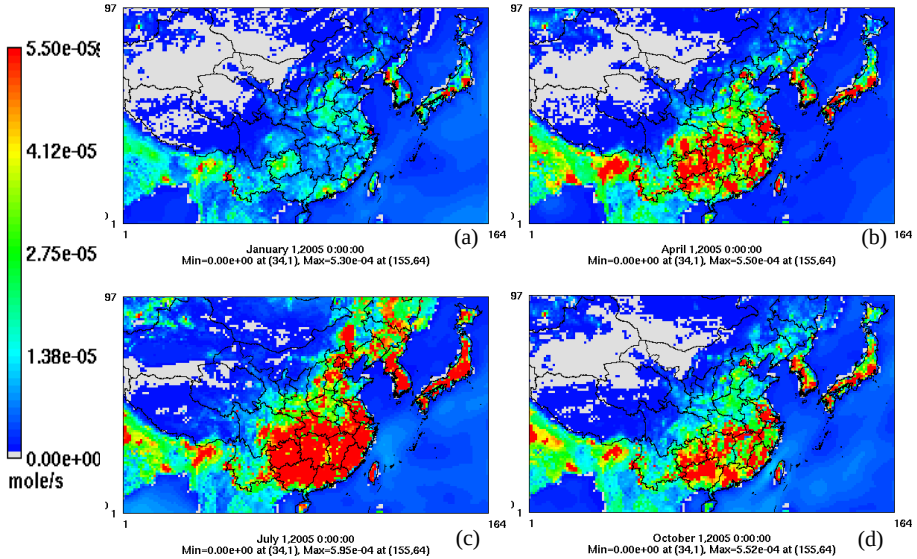

Fig. 1. Spatial distribution of base-case mercury emission in the study domain.

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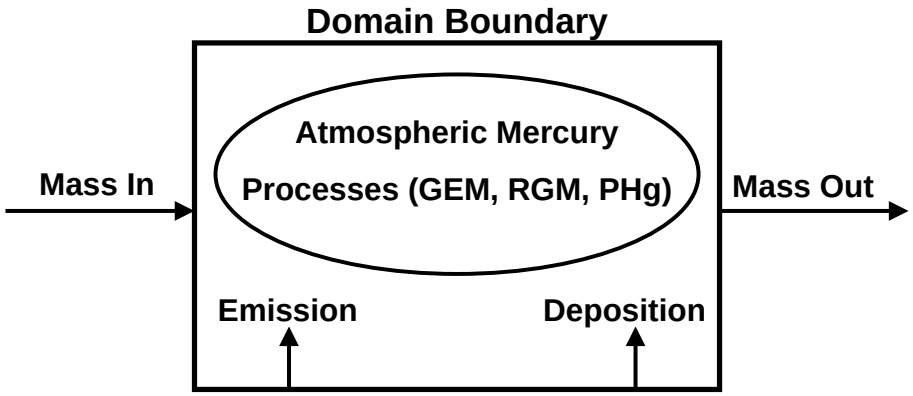

Fig. 2. Schematics of regional mercury budget calculation.

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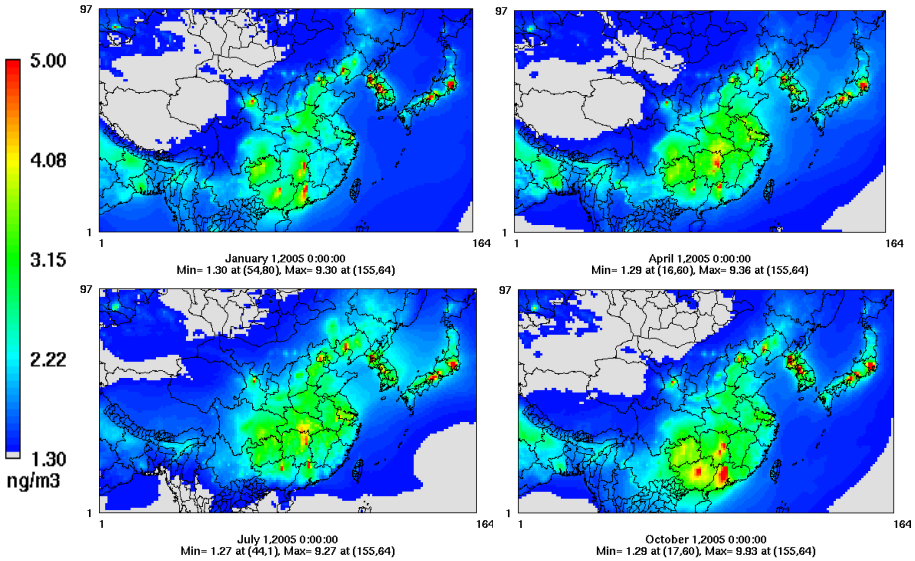

Fig. 3. Spatial distribution of monthly average surface concentration (ng m^{-3}) in the base-case simulation with GEOS-Chem BC/ICs.

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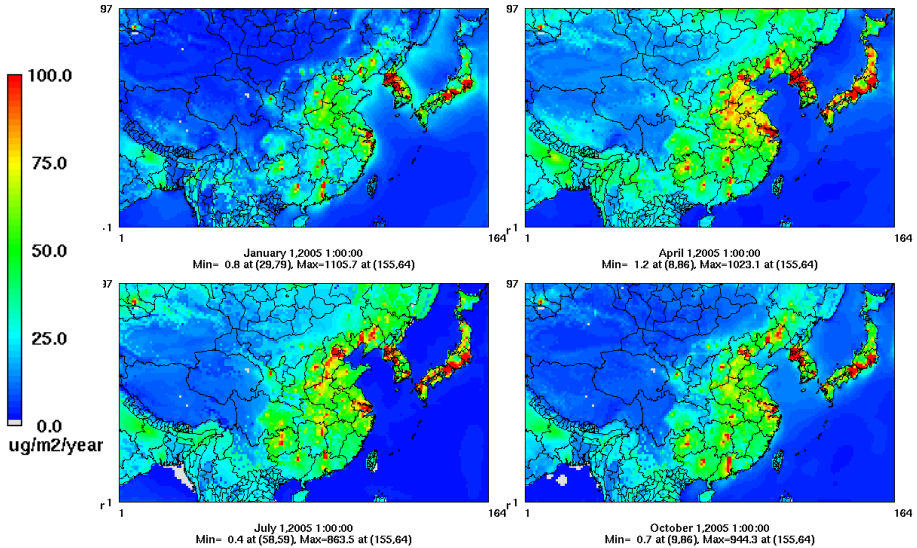

Fig. 4. Spatial distribution of monthly accumulated dry deposition (normalized to annual deposition, $\mu g\, m^{-2}\, yr^{-1}$) in the base-case simulation with GEOS-Chem BC/ICs.

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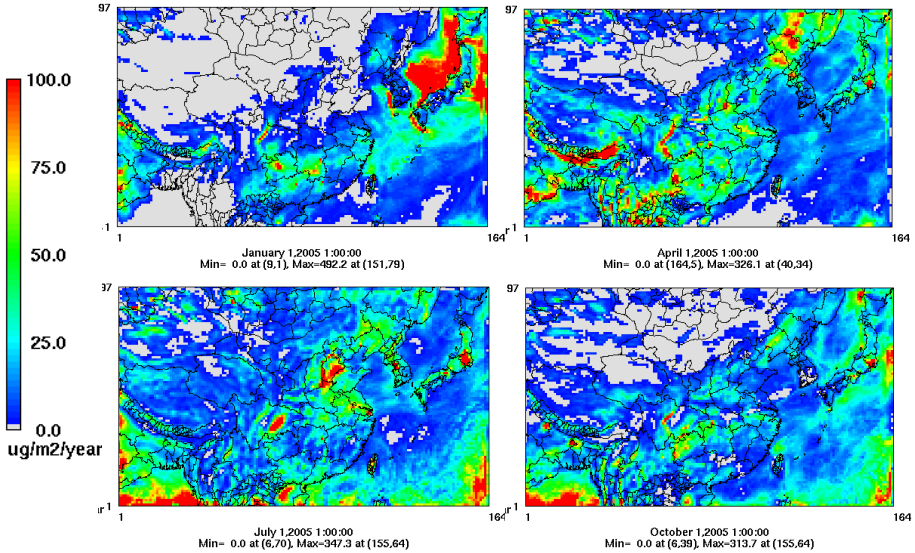

Fig. 5. Spatial distribution of monthly accumulated wet deposition (normalized to annual deposition, $\mu g\, m^{-2}\, yr^{-1}$) in the base-case simulation with GEOS-Chem BC/ICs.

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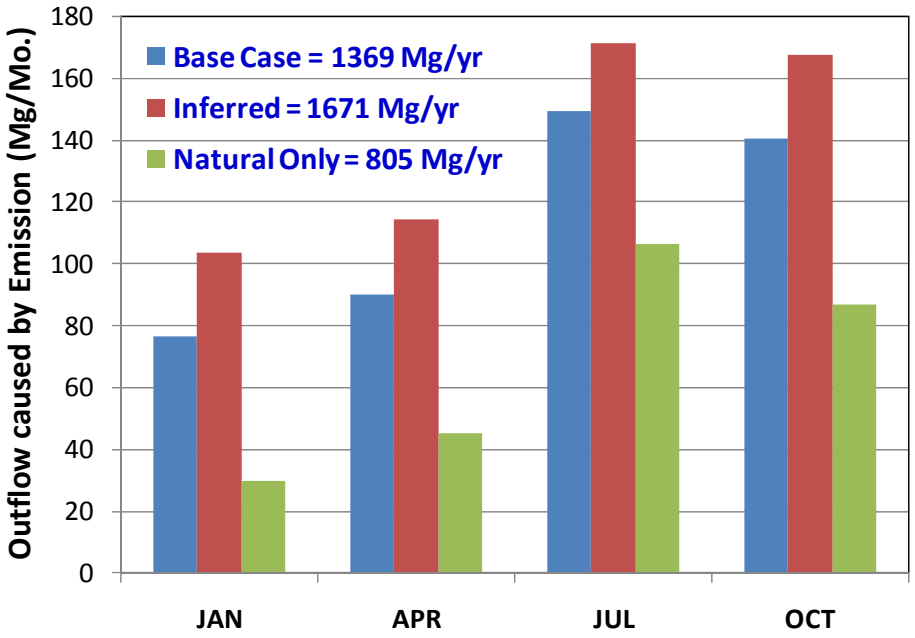

Fig. 6. Estimated mercury outflow caused by the emissions in East Asia under various emission scenarios.

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