



Intercontinental  
transport of  
atmospheric mercury

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# Intercontinental transport and deposition patterns of atmospheric mercury from anthropogenic emissions

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## Abstract

Global policies that regulate anthropogenic mercury emissions to the environment require quantitative and comprehensive source–receptor relationships for mercury emissions, transport and deposition among major continental regions. In this study, we use the GEOS-Chem model to establish source–receptor relationships among eleven major continental regions worldwide. Source–receptor relationships for surface mercury concentrations (SMC) show that some regions (e.g. East Asia, the Indian subcontinent and Europe) should be responsible for their local surface Hg(II) and Hg(P) concentrations because of near-field transport and deposition contributions from their local anthropogenic emissions (up to 64 % and 71 % for Hg(II) and Hg(P), respectively, over East Asia). We define region of primary influence (RPI) and region of secondary influence (RSI) to establish intercontinental influence patterns. Results indicate that East Asia is SMC RPI for almost all other regions, while Europe, Russia and the Indian subcontinent also make some contributions to SMC over some receptor regions because they are dominant RSI source regions. Source–receptor relationships for mercury deposition show that approximately 16 % and 17 % of dry and wet deposition, respectively, over North America originate from East Asia, indicating that trans-pacific transport of East Asian emissions is the major foreign source of mercury deposition in North America. Europe, Southeast Asia and the Indian subcontinent are also important mercury deposition sources for some receptor regions because they are dominant RSI. We also quantify seasonal variation on mercury deposition contributions over other regions from East Asia. Results show that mercury deposition (including dry and wet) contributions from East Asia over the Northern Hemisphere receptor regions (e.g. North America, Europe, Russia, Middle East and Middle Asia) vary seasonally, with the maximum values in summer and minimum values in winter. The opposite seasonal pattern occurs on mercury dry deposition contributions over Southeast Asia and the Indian subcontinent.

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## 1 Introduction

Mercury, known as a global pollutant, can be transported across continents and oceans. The long atmospheric lifetime (about 1 yr) of elemental Hg(0), which makes up approximately 95–99% of atmospheric mercury, contributes to the long distance transport (Schroeder and Munthe, 1998). The remaining mercury consists of gaseous soluble Hg(II) and nonvolatile particulate Hg(P), which are the major contributors to dry and wet deposition of atmospheric mercury (Corbitt et al., 2011). Once deposited into ecosystems, mercury bioaccumulation and methylation in food webs may occur and adversely affect human health, especially infants, who could suffer immune system suppression or neurodevelopmental delays (Rolfhus et al., 2003; Mergler et al., 2007; Selin et al., 2010). Because of anthropogenic emissions from human activities, atmospheric mercury deposition to continents and oceans has increased threefold over the past several centuries (Schuster et al., 2002; Roos-Barracough et al., 2002; Fitzgerald et al., 2005). Growing concerns about increased environmental mercury have promoted the launch of the United Nations Environment Programme (UNEP) Global Legally Binding Treaty on Mercury (Corbitt et al., 2011). Global policies that regulate anthropogenic mercury emissions to the environment require an understanding of source–receptor relationships for mercury emissions, transport and deposition among major continental regions worldwide.

Previous studies have presented some source–receptor relationships on regional and intercontinental scales. Trans-pacific transport of mercury from East Asia, attributed to circumpolar westerlies in the mid-latitudes, contributes to mercury deposition in North America (Seigneur et al., 2004; Selin and Jacob, 2008; Strode et al., 2008). Jaffe and Strode (2008) demonstrated that most Hg(II) and Hg(P) from Asian emissions were removed in Asia, but most Hg(0) from Asian emissions was transported long distance and Asian anthropogenic sources contributed to 7–20% (average 16%) of mercury deposition in North America. Zhang et al. (2012) indicated that 10–22% and 13–20% of mercury wet and dry deposition, respectively, in the US originated

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from North American anthropogenic sources. Lin et al. (2010) investigated mercury emission outflow from East Asia using a chemical transport model and coupling with mass balance analyses, and showed that 75 % of mercury emissions from East Asia were transported outside the region and contributed to 20–30 % of mercury deposition at remote receptors. They also claimed that global anthropogenic sources accounted for 75 % of mercury deposition in East Asia, with 25 % from natural sources. Philip et al. (2007) estimated that mercury deposition in North America was 335 Mg in 2002, and the net outflow to the global pool was 21 Mg. Li Pan et al. (2010) showed that mercury mass outflow (approximately  $681\text{--}714\text{ Mg a}^{-1}$ ) constituted 70 % of mercury emissions from East Asia, with the highest outflow during spring and early summer. Although previous studies have shown some source–receptor relationships among some regions, establishing quantitative and comprehensive influence patterns among major continental regions is needed. Previous studies also focused mainly on selected continental regions (e.g. East Asia, North America and Europe), with little attention focused on other regions which were also critical (e.g. India, Southeast Asia and Russia). Also, mercury emissions from other regions could also influence mercury concentrations and deposition in East Asia, which draws little research attention.

Quantitative and comprehensive influence patterns among major continental regions for some air pollutants (e.g. aerosols,  $\text{O}_3$  and nitrogen deposition) have been established in previous studies (Chin et al., 2007; Liu and Mauzerall, 2007; Liu et al., 2009a, b). Chin et al. (2007) used a global model to estimate the impact of dust aerosols from source regions on surface aerosol concentrations on regional and hemispheric scales, and identified influence patterns among Asia, North America, Europe and Africa. They demonstrated that African and European dust could be transported eastward, where it merged with Asian dust and was subsequently transported across North Pacific to western North America. Liu et al. (2009a, b) described a method for tagging tracers and evaluated intercontinental transport of fine aerosols using the definition of region of primary influence (RPI). Subsequently, they estimated global premature mortality resulting from intercontinental transport of fine aerosols. Relative to quan-

titative and comprehensive influence patterns for some other air pollutants, a quantitative and comprehensive understanding of influence patterns for mercury is needed.

In this study, we use the GEOS-Chem global chemical transport model to evaluate intercontinental transport and deposition patterns of atmospheric mercury from anthropogenic emissions. This study aims to (1) simulate the global distributions of mercury concentrations and deposition and estimate the global budget of atmospheric mercury under the average conditions over the last decade; (2) establish quantitative and comprehensive source–receptor relationships for mercury emissions, transport and deposition among major continental regions worldwide.

## 2 Methods

### 2.1 Model description

We use the GEOS-Chem global mercury model version 9-01-03 (<http://acmg.seas.harvard.edu/geos/>), including a 3-D atmosphere model coupled to 2-D ocean and terrestrial reservoirs (Selin et al., 2008; Holmes et al., 2010; Soerensen et al., 2010; Amos et al., 2012). Simulations are conducted with  $2^\circ \times 2.5^\circ$  horizontal resolution and 47 vertical hybrid eta levels from the surface to 0.01 hPa. The simulations are driven by assimilated meteorological fields from the NASA Goddard Earth Observing System (GEOS-5) and conducted for 2004–2011, with the year 2004 used for initialization and the years 2005–2011 for analysis. As such, all results presented here are 7 yr averages, which can approximately represent average global atmospheric mercury conditions over the last decade. Three inorganic mercury forms, including elemental Hg(0), gaseous soluble Hg(II) and nonvolatile particulate Hg(P), are tracked by the atmospheric simulations.

In GEOS-Chem model version 9-01-03, we have two alternative options to mercury model (the mercury + OH/O<sub>3</sub> model from Selin et al., 2007, or the mercury + Br model from Holmes et al., 2010) and we choose the mercury + Br model. With oxidation of Hg(0) by Br atoms and photoreduction of Hg(II) in cloud droplets, the mercury + Br

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the inventory from Pacyna et al. (2010), and demonstrated significant improvements of their in-plume reduction simulations while comparing with the mercury concentrations observed at 19 surface sites and wet deposition observed at MDN sites. Here, we use the same principles as Amos et al. (2012) and Zhang et al. (2012). The improved ability of GEOS-Chem to reproduce the observations in previous studies (Amos et al., 2012; Soerensen et al., 2012; Zhang et al., 2012) results in the confidence in global simulations and establishment of source–receptor relationships in this study.

### 2.3 Tagged regional tracers

Hg(0), Hg(II) and Hg(P) (which is emitted as Hg(II)) are tagged by defined regions when they are emitted. To establish quantitative and comprehensive source–receptor relationships for mercury emissions, transport and deposition on intercontinental scales, eleven continental regions are defined, as shown in Fig. 1: North America (NA), South America (SA), Europe (EU, excluding the portion of Russia in the European domain), Russia (RU), Africa (AF), the Indian subcontinent (IN), East Asia (EA), Southeast Asia (SE), Australia (AU), Middle East (ME) and Middle Asia (MA). In addition, a tracer (denoted as “Nature”) is used to represent the emissions from natural sources (e.g. oceanic emissions, land reemissions and primary natural emissions (Selin et al., 2007)) and untagged regions in Fig. 1. Figure 2 compares the magnitudes of Hg(0), Hg(II) and Hg(P) anthropogenic emissions from the eleven continental regions. Globally, mercury emissions from EA are the dominant anthropogenic source in the world, and the Hg(0), Hg(II) and Hg(P) emissions account for 48 %, 48 % and 47 % of the global values respectively. The procedure of tagging tracers does not perturb the physical and chemical processes of the model. To evaluate the tagging procedure, we run a base case simulation that is a completed global simulation with no tagging procedure. The sum of concentrations or fluxes originating from tagged tracers (including the eleven continental tagged regions and “Nature”) is compared with those obtained under the base case simulation over a given continental region. Generally, the differences in most regions are less than 1 %, with up to 3 % in a few regions. We attribute the large discrepancy

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The zonal mean interhemispheric ratio at the surface is 1.4 for Hg(0) concentrations, similar to the value of 1.2 found for total gaseous mercury concentrations by Selin et al. (2007). Furthermore, a strong concentration gradient from East Asia to the Pacific Ocean is evident in Fig. 3a, suggesting the possibility of trans-pacific transport of mercury, similar to the findings from Jaffe and Strode (2008) and Lin et al. (2010). Because of accumulation in the upper troposphere and stratosphere (Selin et al., 2007), Hg(II) and Hg(P) concentrations at the surface are far lower than Hg(0) concentrations. High surface Hg(II) concentrations occur in polar regions, likely because of high Br atoms concentrations and Hg(0) oxidation rates in polar regions when using the mercury + Br model (Holmes et al., 2010; Parrella et al., 2012).

Figure 3d reveals that mercury dry deposition over continents also resembles the spatial emission pattern from the GEIA emission inventory. East Asia, the Indian subcontinent, central Europe and southeast US are major deposition regions. Mercury dry deposition over oceans is largely from the uptake of Hg(II) by sea-salt aerosol, and mostly occurs in the Southern Hemisphere. The total fluxes over continents are approximately equal to those over oceans. Hotspots for mercury wet deposition are East Asia, the Indian subcontinent, East Africa, the north and southwest Pacific and the Gulf of Guinea. Unlike dry deposition, high fluxes occur over some oceans for mercury wet deposition and total fluxes over oceans are two-and-a-half times more than those over continents. On a global scale, mercury wet deposition that comprises 42 % of total mercury deposition is comparable but somewhat smaller than dry deposition. This is in agreement with values of 30–50 % found in some regional model studies (Lin et al., 2007, 2010). Total mercury deposition fluxes range from  $1 \mu\text{g m}^{-2} \text{a}^{-1}$  over some areas in the Antarctic to  $77 \mu\text{g m}^{-2} \text{a}^{-1}$  over some areas in East Asia.

### 3.2 Global atmospheric mercury budget

Figure 4 shows the global budget of atmospheric mercury derived from this study. Our global budget is similar to those presented in Holmes et al. (2010), Soerensen et al. (2010) and Amos et al. (2012). Anthropogenic emissions in this study are

1930  $\text{Mga}^{-1}$  and total emissions from land are 3730  $\text{Mga}^{-1}$ . It should be noted that we improve the proportion of Hg(0) anthropogenic emissions and merge Hg(P) emissions with Hg(II), based on the findings from Zhang et al. (2012) and Amos et al., (2012), so Hg(0), Hg(II) and Hg(P) anthropogenic emissions in this study are different than those reported by Holmes et al. (2010) and Soerensen et al. (2010). Global atmospheric burdens are 3600 Mg Hg(0), 640 Mg Hg(II) and 400 Mg Hg(P). The flux of Hg(0) to Hg(II) by Br oxidation is 7100  $\text{Mga}^{-1}$ , which is similar to the value of 8000  $\text{Mga}^{-1}$  from Holmes et al. (2010) and Driscoll et al. (2013). However, because we adopt the in-cloud reduction rate constant from Amos et al. (2012), which is half that used by Holmes et al. (2010), the photoreduction flux of Hg(II) to Hg(0) is 1900  $\text{Mga}^{-1}$ , nearly half that from Holmes et al. (2010) and Driscoll et al. (2013). From the amendment of Hg(II) partitioning between gas and particle phases, approximately 480 Mg Hg(II) converts to Hg(P) each year. The amendment to the mechanism of Hg(0) across the air-sea interface (discussed in Sect. 2.1) results in Hg(0) dry deposition to ocean being only 50  $\text{Mga}^{-1}$ , which is significantly different than earlier global model predictions (Selin et al., 2008; Holmes et al., 2010; Corbitt et al., 2011). However, net Hg(0) ocean evasion is consistent with other studies (3050  $\text{Mga}^{-1}$  in this study, 2900  $\text{Mga}^{-1}$  from Amos et al., 2012, 3100  $\text{Mga}^{-1}$  from Amos et al., 2013, 2950  $\text{Mga}^{-1}$  from Soerensen et al., 2010, and 3000  $\text{Mga}^{-1}$  from Driscoll et al., 2013). Globally, mercury dry deposition is 2330  $\text{Mga}^{-1}$  (58 % Hg(0), 40 % Hg(II) and 2 % Hg(P)), and wet deposition is 2900  $\text{Mga}^{-1}$  (85 % Hg(II) and 15 % Hg(P)). Meanwhile, 1600 Mg Hg(II) is taken up by sea-salt aerosols and deposits to the ocean each year. The fluxes of mercury deposition and uptake by sea-salt aerosols are similar to those in Amos et al. (2012). To sum all emissions and deposition (deposition is assumed negative here) up, we estimate that nearly 1000 Mg mercury is transported from land to the ocean and is sequestered by the ocean each year.

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### 3.3 Source–receptor relationships for surface mercury concentrations

#### 3.3.1 Contributions from local and foreign sources

The simulations are conducted under three scenarios: “Total”, “Local” and “Background”. “Total” refers to concentrations or deposition fluxes over a specific receptor region resulting from all global source emissions (exactly the same as the base case simulation discussed in Sect. 2.3). “Local” refers to those resulting from local anthropogenic emissions. “Background” denotes those owing to all sources except anthropogenic emissions from the receptor region, which consists of two categories: “Others” and “Nature”. “Others” includes anthropogenic emissions from the ten regions other than the receptor region and “Nature” refers to emissions from all global natural sources and untagged regions (the same as the “Nature” tracer discussed in Sect. 2.3).

The average area-weighted (A-W) concentration from Liu et al. (2009a) is used in this study to quantify the average concentration over a specific region and is defined in Eq. (1). Then, the SMC over different regions are comparable.

$$C_{aw} = \frac{\sum_{i=1}^n C_i \cdot S_i}{\sum_{i=1}^n S_i} \quad (1)$$

In Eq. (1),  $C_{aw}$  is the A-W SMC over a receptor region R. The total number of grid boxes covered by region R is  $n$ ,  $C_i$  is the SMC in grid box  $i$  in region R and  $S_i$  is the area of grid box  $i$  in region R. We also define the percent contributions of  $C_{aw}$  (POC) using ratios of “Local” or “Background” to “Total” for each receptor region.

Table 1 summarizes the A-W SMC over each receptor region from each source category (including “Total”, “Local” and “Background”), with all POCs for each receptor region. It indicates that A-W surface Hg(0) concentrations over the ten regions other than EA are mainly attributed to global natural sources (> 50 %). For EA, local anthropogenic emissions are the predominant sources and comprise 41 % of the total contributions. Thus, EA is the only region whose contributions from local anthropogenic emissions

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exceed foreign ones. Global anthropogenic emissions and transformation dominate A-W surface Hg(II) concentrations over EU, IN and EA (> 50 %), while in other regions they are dictated by global natural sources. Local emissions also play a dominant role in EU, IN and EA, accounting for 52 %, 53 % and 64 %, respectively. For A-W surface Hg(P) concentrations, regions that are dominated by global anthropogenic emissions and transformation (> 50 %) include NA, EU, RU, IN and EA. Similarly to Hg(II), local emissions play a dominant role in these regions, especially in EU (66 %) and EA (71 %).

Natural sources include oceanic emissions, land reemissions and primary natural emissions, which account for two-thirds of the total emissions (Pirrone et al., 2010; 72 % in this study). Besides, natural sources mainly emit Hg(0) (Selin et al., 2007, 2008). Therefore it is not surprising there are large contributions from global natural sources to surface Hg(0) concentrations over all regions. Hg(II) and Hg(P) show near-field transport and deposition, owing to their sufficient water solubility. Some regions (e.g. EA, IN and EU), where large local anthropogenic emissions occur, should be responsible for their local surface Hg(II) and Hg(P) concentrations.

### 3.3.2 Region of primary influence and region of secondary influence

It is useful for global environmental policymakers to understand which foreign region's anthropogenic emissions have significant effects on a specific receptor region's SMC. To make this clear, we adopt the definition of region of primary influence (RPI) from Liu et al. (2009a). Because of high anthropogenic mercury emissions from EA (Fig. 2), EA is the SMC RPI for most regions in our simulations. Therefore, we define one source region as the region of secondary influence (RSI) on a given receptor region so that we can identify other influence patterns outside of those from EA. Calculations of RPI and RSI in this section are based on POC (defined in Sect. 3.3.1), where here POC is calculated using ratios of SMC originating from each foreign region to SMC originating from "Total" over a given receptor region. When all POCs are added together for a given

receptor region, an equivalent value to the POC calculated using ratios of “Others” to “Total” for the receptor region in Table 1 is obtained.

Figure 5 illustrates intercontinental influence patterns based on the RPI and RSI for SMC. EA is the SMC (Hg(0), Hg(II) and Hg(P)) RPI for the other ten regions, except that RU is the surface Hg(P) concentrations RPI for EU. EU, IN and RU are the surface Hg(0), Hg(II) and Hg(P) concentrations RPIs, respectively, for EA. Their contributions are all extremely low ( $0 < \text{POC} < 5$ ), indicating that local emissions influence EA significantly. For surface Hg(0) concentrations, there are three significant ( $\text{POC} > 15$ ) RPI relationships ( $\text{EA} \rightarrow \text{RU}$ ,  $\text{EA} \rightarrow \text{SE}$  and  $\text{EA} \rightarrow \text{NA}$ ), compared with only one significant RPI for surface Hg(II) concentrations ( $\text{EA} \rightarrow \text{MA}$ ) and no significant RPI for surface Hg(P) concentrations. This indicates that Hg(0) is transported far from source regions, while Hg(II) and Hg(P) show local emissions and transport, which is consistent with the results in Sect. 3.3.1.

EU is the surface Hg(0) concentrations RSI for NA, AF, ME and RU and is the dominant RSI source region for Hg(0).  $\text{RU} \rightarrow \text{EU}$  and  $\text{RU} \rightarrow \text{MA}$  are moderate RSI relationships ( $5 < \text{POC} < 10$ ) regarding surface Hg(II) concentrations. For Hg(P), IN is the dominant RSI source region, which is RSI for EA, SE, AF and ME. NA, AF, ME, MA and AU are the primary receptor regions because they are not RPI or RSI for any region. Overall, EA is responsible for SMC over almost all regions if contributions from their local anthropogenic emissions are not accounted for. However, EU, RU and IN also make some contributions to SMC over the primary receptor regions (e.g. NA, AF, ME, MA and AU).

### 3.4 Source–receptor relationships for mercury deposition

#### 3.4.1 Contributions from local and foreign sources

Similarly to Sect. 3.3, the categories of “Total”, “Local” and “Background” (including “Others” and “Nature”) are used to identify intercontinental influence patterns for mercury deposition. For each region, we add the deposition fluxes in all grid boxes to obtain

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a total deposition. Here we define the percent contributions of deposition fluxes (POF) using ratios of “Local” or “Background” to “Total” for each receptor region.

Table 2 summarizes the mercury deposition fluxes (including dry and wet) over each receptor region from each source category (including “Total”, “Local” and “Background”), with all POFs for each receptor region. It shows that global natural sources dominate mercury dry deposition over all regions (> 50 %) except IN and EA. For the nine regions that are mainly affected by global natural sources, contributions from foreign anthropogenic emissions all exceed local anthropogenic emissions, with only EU having nearly equal values. However, mercury dry deposition over IN and EA mainly originate from global anthropogenic emissions (> 50 %), with the primary contributions being from local emissions. Differences between local and foreign emissions are 14 % and 44 %, respectively. Similarly to dry deposition, mercury wet deposition over all regions except EA are dictated by global natural sources (> 50 %). For the ten other regions, contributions from foreign anthropogenic emissions all exceed local anthropogenic emissions, with only IN having nearly equal values. EA is the only region whose contributions from anthropogenic sources are greater than natural sources and the difference between local and foreign emissions is 32 %.

Lin et al. (2010) demonstrated that annual average mercury dry and wet deposition were 425 Mg and 396 Mg, respectively, in East Asia, higher than those in this study. Contributions from anthropogenic sources to total mercury deposition (dry + wet) account for 75 % based on the results from Lin et al. (2010), which is also higher than those in this study (62 % and 54 % for dry and wet deposition, respectively). We attribute the discrepancies to the large area of East Asia (including parts of India and Southeast Asia) defined and the different anthropogenic emissions inventory used by Lin et al. (2010). Philip et al. (2007) estimated  $335 \text{ Mg a}^{-1}$  for total mercury deposition in North America, which was lower than that in this study ( $437.4 \text{ Mg a}^{-1}$ ). The small area of North America (excluding Alaska and the portion of northern Canada) defined in their study likely accounts for this discrepancy.

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Comparisons between mercury dry and wet deposition in Table 2 indicate that contributions from global natural sources to mercury dry deposition are approximately equal to those for wet deposition over each region, while contributions from local anthropogenic emissions to dry deposition exceed those for wet deposition. Conversely, dry deposition is lower than wet deposition for contributions from foreign anthropogenic emissions over each region. Overall, wet deposition occurs farther from source regions than dry deposition that mainly shows near-field occurrence. We attribute this to close relationships between mercury wet deposition and the formation of clouds and precipitation.

### 3.4.2 Region of primary influence and region of secondary influence

RPI and RSI are also used to understand source–receptor relationships for mercury deposition among the eleven regions. Similarly to POC in Sect. 3.3.2, POF is used to calculate RPI and RSI in this section. The influence patterns are shown in Fig. 6. EA is the mercury dry and wet deposition RPI for the other ten regions and IN is EA's RPI. It should be noted that EA is the significant mercury deposition (including dry and wet) RPI for NA, RU and MA (POF > 15). EA is the only significant dry deposition RPI for SE and the only significant wet deposition RPI for EU. For NA, approximately 16% and 17% of dry and wet deposition, respectively, originate from EA, consistent with 7–20% (average 16%) from Jaffe and Strode (2008). It also indicates that trans-pacific transport of EA emissions is the major foreign source of mercury deposition in NA, which is in agreement with previous findings (Seigneur et al., 2004; Selin and Jacob, 2008; Strode et al., 2008). Compared with NA, the dry and wet deposition contributions from EA are 17% and 18%, respectively, for RU and 15% and 16%, respectively, for MA.

For mercury dry deposition, EU and SE both have three RSI relationships for other receptor regions and IN is the mercury wet deposition RSI for AF, SE, NA and ME, making it the dominant RSI source region for wet deposition. EU, SE and IN are also important mercury deposition source regions for some receptor regions (e.g. AU, AF,



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maximum values in summer and minimum values in winter. When low contributions originate from EA, contributions from local emissions dominate mercury deposition in these regions. In summer, high concentrations of oxidant (Br atoms) in the Northern Hemisphere result in active mercury chemical reactions, contributing to more deposition (Holmes et al., 2006; Holmes et al., 2010; Parrella et al., 2012). However, mercury dry deposition contributions over SE and IN from EA have the opposite seasonal pattern. For SE, the monsoon climate in EA may contribute to this difference. Also, the Qinghai-Tibet Plateau may affect the seasonal pattern over IN. More studies should be conducted about the impact of the monsoon climate in EA on the transport of atmospheric mercury from EA to SE and the impact of the Qinghai-Tibet Plateau on the transport of atmospheric mercury between EA and IN.

#### 4 Conclusions

With growing concerns about the influence of intercontinental transport and deposition of air pollutants, a quantitative and comprehensive understanding of influence patterns for mercury is needed. In this study, we tag emissions of Hg(0), Hg(II) and Hg(P) emitted from eleven continental regions and use the GEOS-Chem model (v9-01-03) and GEIA emission inventory to evaluate the intercontinental transport and deposition patterns of atmospheric mercury from anthropogenic emissions. Compared with previous GEOS-Chem versions (e.g. v8-03-02 and v9-01-02), we obtain  $1900 \text{ Mg a}^{-1}$  for Hg(II) photoreduction in clouds. Approximately 480 Mg Hg(II) converts to Hg(P) through partitioning and nearly 1000 Mg mercury is transported from land to the ocean and is sequestered by the ocean each year.

For each continental region, contributions from local and foreign sources are quantified and intercontinental influence patterns are established for area-weighted (A-W) surface mercury concentrations (SMC) using the definition of region of primary influence (RPI) and region of secondary influence (RSI). Global natural sources are the dominant sources to A-W surface Hg(0) concentrations over all regions except EA.

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For EA, local anthropogenic emissions are the dominant sources and comprise 41 % of total contributions. Results show that some regions (e.g. EA, IN and EU) should be responsible for their local surface Hg(II) and Hg(P) concentrations because of high contributions from local anthropogenic emissions. EA is the SMC RPI for all other regions, except that RU is the surface Hg(P) concentrations RPI for EU. For surface Hg(0) concentrations, there are three significant RPI relationships (EA → RU, EA → SE and EA → NA), with only one for Hg(II) (EA → MA) and none for Hg(P). EU is the dominant RSI source region for Hg(0), with IN the dominant RSI for Hg(P). RU → EU and RU → MA are moderate RSI relationships regarding Hg(II). Generally, EA is responsible for SMC over almost every region, while EU, RU and IN also make some contributions to SMC over other receptor regions (e.g. NA, AF, ME, MA and AU).

Similarly to SMC, the contributions from local and foreign sources are quantified and intercontinental influence patterns are established for mercury deposition. Global natural sources are the main contributors for dry deposition over all regions except IN and EA, and for wet deposition over all regions except EA. Dry deposition over IN and EA are dictated by anthropogenic emissions, with differences between local and foreign emissions of 14 % and 44 %, respectively. Difference between local and foreign emissions is 32 % for the contributions from anthropogenic sources to wet deposition over EA. EA is the mercury deposition RPI for the other ten regions, being the significant dry deposition RPI for NA, RU, MA and SE, and wet deposition RPI for NA, RU, MA and EU. Approximately 16 % and 17 % of dry and wet deposition, respectively, over NA originate from EA, indicating that trans-pacific transport of EA emissions is the major foreign source of mercury deposition in NA. EU and SE both have three dry deposition RSI relationships for other receptor regions, while IN is the dominant wet deposition RSI source region. EU, SE and IN are also responsible for some receptor regions' mercury deposition.

Seasonal variation on mercury deposition contributions over other regions from EA is quantified. Mercury deposition (including dry and wet) contributions from EA over the Northern Hemisphere receptor regions (e.g. NA, EU, RU, ME and MA) vary seasonally,

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with the maximum values in summer and minimum values in winter. However, the opposite seasonal pattern occurs on mercury dry deposition contributions over SE and IN from EA. Generally, international efforts that strengthen bilateral cooperation between receptor regions and their RPI or RSI (especially EA) to reduce mercury emissions are necessary to address global mercury pollution, and policymakers should also be aware of temporal patterns affecting receptor regions from source regions.

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**Table 1.** Contributions from local and foreign sources to annual average A-W SMC (including Hg(0), Hg(II), Hg(P)) over each receptor region.

Sources	Forms	Unit	Receptors										
			NA	SA	EU	RU	AF	IN	EA	SE	AU	ME	MA
Total <sup>a</sup>	Hg(0)	ng m <sup>-3</sup>	<b>1.45</b>	<b>1.14</b>	<b>1.59</b>	<b>1.53</b>	<b>1.19</b>	<b>1.41</b>	<b>1.97</b>	<b>1.29</b>	<b>0.97</b>	<b>1.32</b>	<b>1.48</b>
	Hg(II)	pg m <sup>-3</sup>	13.71	9.75	11.35	12.48	14.79	43.05	29.26	5.28	18.38	21.44	16.56
	Hg(P)	pg m <sup>-3</sup>	2.36	0.86	9.93	8.80	12.66	18.28	48.58	0.80	3.47	18.89	15.87
Local <sup>b</sup>	Hg(0)	ng m <sup>-3</sup>	<b>0.08</b>	<b>0.08</b>	<b>0.18</b>	<b>0.11</b>	<b>0.04</b>	<b>0.24</b>	<b>0.81</b>	<b>0.11</b>	<b>0.02</b>	<b>0.05</b>	<b>0.07</b>
		%	5	7	11	7	3	17	41	9	2	4	5
	Hg(II)	pg m <sup>-3</sup>	2.70	0.99	5.90	3.05	1.25	22.79	18.63	1.31	1.06	2.09	2.13
		%	20	10	52	24	8	53	64	25	6	10	13
	Hg(P)	pg m <sup>-3</sup>	0.96	0.07	6.59	4.05	0.52	8.81	34.72	0.15	0.14	1.65	2.40
		%	41	9	66	46	4	48	71	18	4	9	15
Background <sup>c</sup> (Others)	Hg(0)	ng m <sup>-3</sup>	<b>0.39</b>	<b>0.18</b>	<b>0.41</b>	<b>0.44</b>	<b>0.28</b>	<b>0.31</b>	<b>0.22</b>	<b>0.32</b>	<b>0.15</b>	<b>0.39</b>	<b>0.46</b>
		%	27	16	26	29	24	22	11	25	16	30	31
	Hg(II)	pg m <sup>-3</sup>	3.31	1.86	2.10	3.18	3.62	5.74	2.27	1.13	3.74	6.18	5.36
		%	24	19	18	25	24	13	8	21	20	29	32
	Hg(P)	pg m <sup>-3</sup>	0.42	0.17	1.52	1.89	3.49	2.67	2.87	0.24	0.71	5.40	5.03
		%	18	20	15	22	28	15	6	30	20	29	32
Background <sup>c</sup> (Nature)	Hg(0)	ng m <sup>-3</sup>	<b>0.98</b>	<b>0.86</b>	<b>1.01</b>	<b>0.99</b>	<b>0.86</b>	<b>0.85</b>	<b>0.95</b>	<b>0.85</b>	<b>0.77</b>	<b>0.88</b>	<b>0.95</b>
		%	68	76	64	65	72	60	48	66	80	67	64
	Hg(II)	pg m <sup>-3</sup>	7.67	6.78	3.37	6.23	9.80	14.42	8.31	2.81	13.35	13.10	9.10
		%	56	70	30	50	66	33	28	53	73	61	55
	Hg(P)	pg m <sup>-3</sup>	0.98	0.60	1.82	2.83	8.57	6.74	10.91	0.41	2.57	11.77	8.41
		%	42	70	18	32	68	37	22	51	74	62	53

<sup>a</sup>“Total” refers to average A-W SMC over a receptor region resulting from all global source emissions.

<sup>b</sup>“Local” refers to average A-W SMC over a receptor region resulting from local anthropogenic emissions.

<sup>c</sup>“Background” denotes average A-W SMC over a receptor region resulting from all sources except anthropogenic emissions from the receptor region, including anthropogenic emissions from the other ten regions (“Others”) and emissions from all global natural sources and untagged regions (“Nature”).

The percent contributions (POC) that are defined using ratios of “Local” or “Background” to “Total” are quantified.

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**Table 2.** Contributions from local and foreign sources to dry and wet deposition over each receptor region.

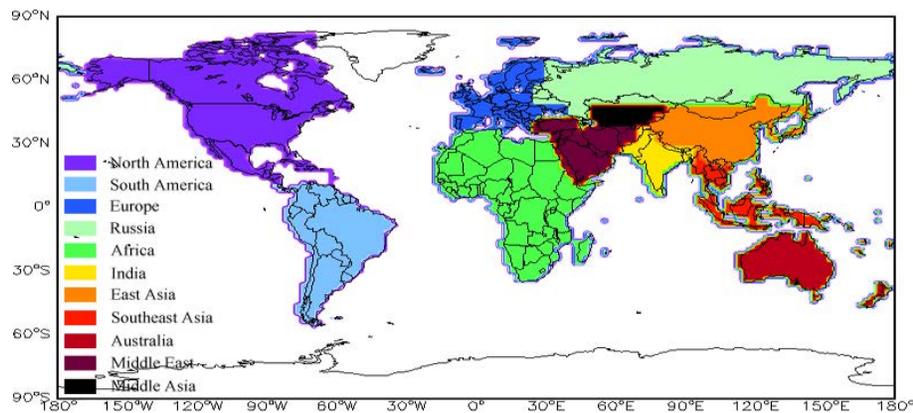
Sources	Forms	Unit	Receptors										
			NA	SA	EU	RU	AF	IN	EA	SE	AU	ME	MA
Total	Dry	Mga <sup>-1</sup>	<b>292.7</b>	<b>328.3</b>	<b>95.3</b>	<b>203.0</b>	<b>388.8</b>	<b>82.3</b>	<b>266.6</b>	<b>91.8</b>	<b>109.4</b>	<b>77.2</b>	<b>24.2</b>
	Wet	Mga <sup>-1</sup>	<b>144.7</b>	<b>158.4</b>	<b>43.2</b>	<b>97.5</b>	<b>187.7</b>	<b>57.4</b>	<b>113.1</b>	<b>68.4</b>	<b>66.4</b>	<b>37.6</b>	<b>10.5</b>
Local	Dry	Mga <sup>-1</sup>	<b>32.2</b>	<b>23.1</b>	<b>21.2</b>	<b>18.5</b>	<b>17.7</b>	<b>26.5</b>	<b>141.7</b>	<b>13.1</b>	<b>5.3</b>	<b>4.7</b>	<b>1.3</b>
	Dry	%	11	7	22	9	5	32	53	14	5	6	6
	Wet	Mga <sup>-1</sup>	<b>8.5</b>	<b>6.1</b>	<b>5.1</b>	<b>5.4</b>	<b>5.0</b>	<b>11.8</b>	<b>48.2</b>	<b>4.0</b>	<b>0.9</b>	<b>1.2</b>	<b>0.2</b>
	Wet	%	6	4	12	6	3	21	43	6	1	3	2
Background (Others)	Dry	Mga <sup>-1</sup>	<b>77.2</b>	<b>56.5</b>	<b>23.4</b>	<b>60.5</b>	<b>89.6</b>	<b>15.1</b>	<b>23.5</b>	<b>23.1</b>	<b>19.2</b>	<b>22.9</b>	<b>7.8</b>
	Dry	%	26	17	25	30	23	18	9	25	18	30	32
	Wet	Mga <sup>-1</sup>	<b>41.3</b>	<b>34.2</b>	<b>12.1</b>	<b>31.3</b>	<b>48.1</b>	<b>12.6</b>	<b>12.5</b>	<b>16.4</b>	<b>14.7</b>	<b>10.9</b>	<b>3.3</b>
	Wet	%	29	22	28	32	26	22	11	24	22	29	31
Background (Nature)	Dry	Mga <sup>-1</sup>	<b>183.9</b>	<b>244.6</b>	<b>51.6</b>	<b>125.4</b>	<b>278.2</b>	<b>40.3</b>	<b>101.1</b>	<b>54.9</b>	<b>83.1</b>	<b>49.4</b>	<b>15.1</b>
	Dry	%	63	74	54	62	72	49	38	60	76	64	62
	Wet	Mga <sup>-1</sup>	<b>94.4</b>	<b>116.1</b>	<b>26.0</b>	<b>60.8</b>	<b>133.1</b>	<b>32.7</b>	<b>52.1</b>	<b>47.3</b>	<b>49.7</b>	<b>25.3</b>	<b>6.9</b>
	Wet	%	65	73	60	62	71	57	46	69	75	67	66

The categories are defined same as Table 1.

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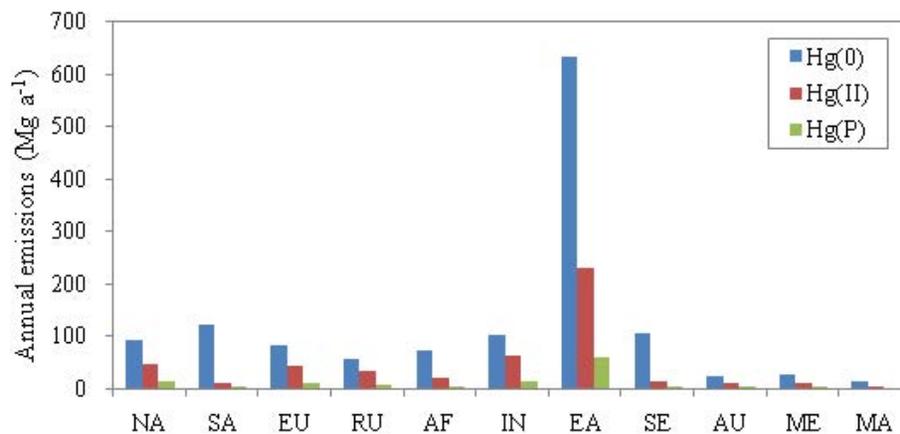
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**Fig. 1.** Eleven continental regions tagged in our GEOS-Chem simulations.

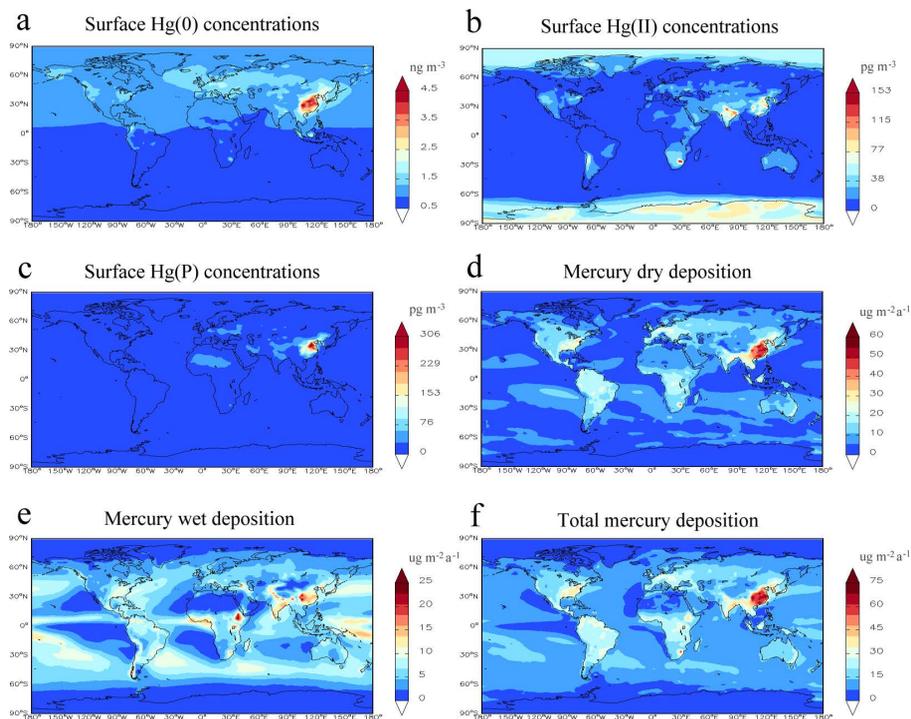
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**Fig. 2.** Tagged regional anthropogenic mercury emissions.

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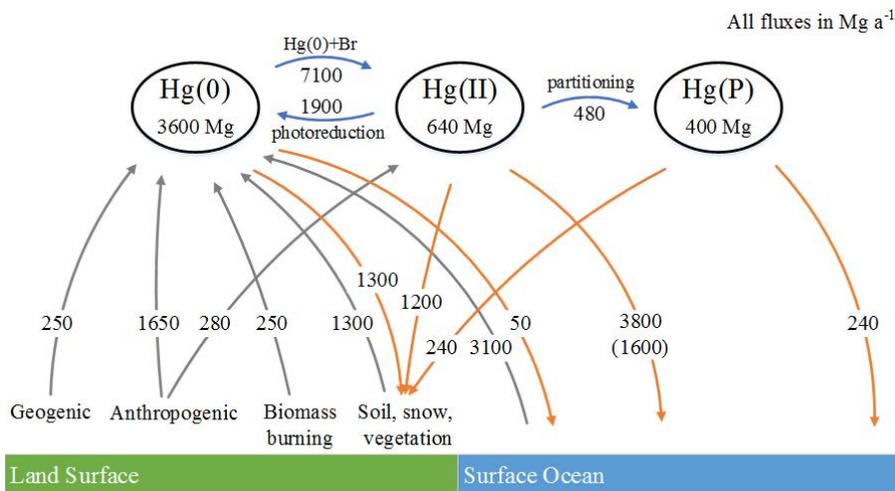


**Fig. 3.** Global spatial distributions of annual average SMC and mercury deposition: **(a)** surface Hg(0) concentrations; **(b)** surface Hg(II) concentrations; **(c)** surface Hg(P) concentrations; **(d)** mercury dry deposition (Hg(0) dry deposition + Hg(II) dry deposition + Hg(P) dry deposition); **(e)** mercury wet deposition (Hg(II) wet deposition + Hg(P) wet deposition); **(f)** Total mercury deposition (dry deposition + wet deposition).

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**Fig. 4.** Global budget of atmospheric mercury derived from this study. The fluxes in parentheses indicate uptake of Hg(II) by sea-salt aerosol.

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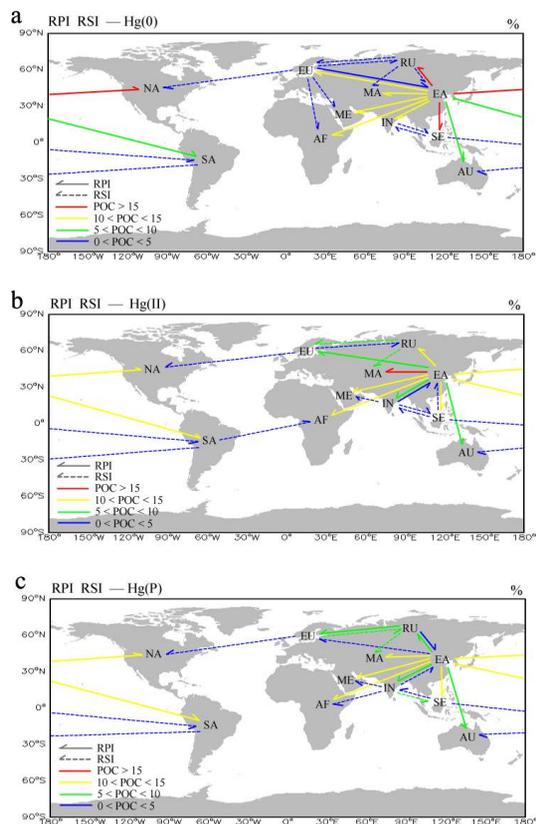
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**Fig. 5.** Intercontinental influence patterns based on the RPI (region of primary influence) and RSI (region of secondary influence) for SMC: **(a)** Hg(0); **(b)** Hg(II); **(c)** Hg(P). Arrows point in the direction of influence from RPI or RSI to a receptor region. Colors indicate the magnitude of POC contributed from RPI or RSI to a receptor region.

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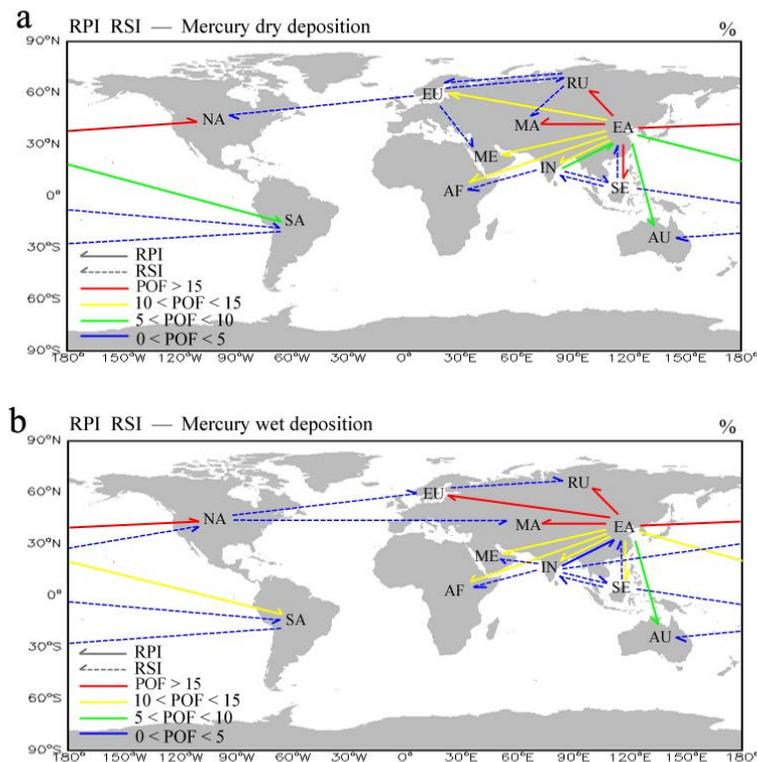
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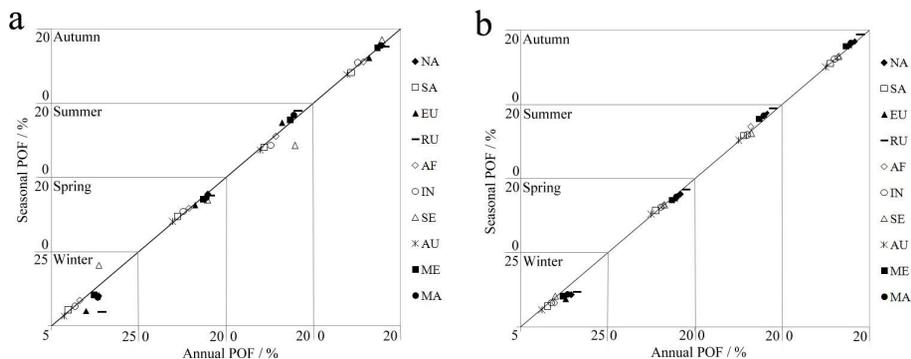
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**Fig. 6.** Intercontinental influence patterns based on the RPI (region of primary influence) and RSI (region of secondary influence) for mercury deposition: **(a)** mercury dry deposition; **(b)** mercury wet deposition. The arrows and colors are defined same as Fig. 5.

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**Fig. 7.** Seasonal variation on mercury deposition contributions over other regions from East Asia (EA): **(a)** mercury dry deposition; **(b)** mercury wet deposition. Four period zones are defined (winter, spring, summer and autumn) and each period zone has its own ranges of POF values. The line is for equivalence between seasonal POF and annual POF.