



Intercontinental
transport of
atmospheric mercury

L. Chen et al.

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Intercontinental transport and deposition patterns of atmospheric mercury from anthropogenic emissions

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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.

Intercontinental transport of atmospheric mercury

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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

Intercontinental transport of atmospheric mercury

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Intercontinental
transport of
atmospheric mercury**

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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, 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-

aerosol loading, to more than 90 % in cold air and high aerosol loading. With these developments, model simulations of mercury wet deposition and Hg(P) concentrations at observation sites in the US were improved (Amos et al., 2012; Soerensen et al., 2012).

2.2 Emissions

5 The Global Emission Inventory Activity (GEIA) anthropogenic mercury emission inventory for the year 2005 (Pacyna et al., 2010) is used in this study. This inventory includes Hg(0), Hg(II), and Hg(P) at 1320, 480 and 130 Mg a⁻¹, respectively, with a horizontal resolution of 1° × 1° and no seasonal variation. Combustion of fossil fuels (primarily coal) accounts for 46 % of the total anthropogenic emissions, making it the largest emission
10 source from human activities. The inventory includes artisanal/small-scale gold mining that is not included in previous inventories (Pacyna et al., 2005; Wilson et al., 2005), but is responsible for 18 % of anthropogenic emissions. The inventory indicates that Asian sources release approximately two-thirds of global anthropogenic mercury emissions, with China as the largest source region worldwide. Although the US and India are the
15 second and third highest contributors, respectively, their emissions combined are only 30 % those of China.

According to the findings from Amos et al. (2012) and Zhang et al. (2012), we adjust the mercury emission speciation. Amos et al. (2012) assumed that Hg(P) was emitted as semi-volatile Hg(II) rather than as refractory, and merged it with Hg(II) emissions.
20 The adjustment considerably improved their model simulations of Hg(P) at observation sites. Although its chemical mechanism has not been identified (Lohman et al., 2006), in-plume reduction of Hg(II) emitted from power plants has been confirmed in previous model studies (Seigneur et al., 2003, 2006; Lohman et al., 2006; Vijayaraghavan et al., 2008; Kos et al., 2011; Zhang et al., 2012). In addition, Edgerton et al. (2006) observed that Hg(0) accounted for about 84 % of total mercury emissions from power
25 plant plumes at three surface sites. Therefore, Zhang et al. (2012) substituted an 86.5 : 9.9 : 3.6 (Hg(0) : Hg(II) : Hg(P)) speciation for the 50 : 40 : 10 speciation used for fossil fuel combustion, which comprised 46 % of the total anthropogenic emissions in

Intercontinental transport of atmospheric mercury

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



(3%) to nonlinear calculations in some processes of the model, such as convection, diffusion, partitioning and chemical reactions. The small nonlinear calculations do not affect the implications of our tagging procedure and the analysis of our results.

3 Results and discussion

5 Surface mercury concentrations (SMC) (including Hg(0), Hg(II) and Hg(P)) and mercury deposition (including dry and wet deposition) are discussed in this section. The surface layer in GEOS-Chem is at the bottom of the troposphere, which averages 120 m high and is where most human activities occur. The SMC and mercury deposition fluxes are averaged from 2005 to 2011 to approximately represent the average conditions over the last decade.

10 First, we run a base case global simulation with no tagging procedure to obtain global distributions and budget for model evaluation and then we quantify and compare source–receptor relationships for SMC and mercury deposition among the eleven continental regions with the tagging procedure.

15 3.1 Global distributions of mercury concentrations and deposition

The global spatial distributions of annual average SMC are shown in Fig. 3a–c. Figure 3d–f illustrates the global spatial distributions of annual average mercury dry deposition, wet deposition and total deposition (dry + wet), respectively. The surface Hg(0) concentrations resemble the spatial emission pattern from the GEIA emission inventory, with a footprint of dominant anthropogenic source regions (e.g. East Asia, the Indian subcontinent and Europe). The model-predicted surface Hg(0) concentrations in this study range from 0.9 to 4.3 ng m⁻³, consistent with the results from Holmes et al. (2010). Figure 3a reveals a considerably stronger interhemispheric gradient of surface Hg(0) concentrations, which has also been shown from cruise data (Lamborg et al., 2002; Temme et al., 2003) and previous model predictions (Selin et al., 2007).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Intercontinental transport of atmospheric mercury

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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

**Intercontinental
transport of
atmospheric mercury**

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

1930 Mga^{-1} and total emissions from land are 3730 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 Mga^{-1} , which is similar to the value of 8000 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 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 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 Mga^{-1} in this study, 2900 Mga^{-1} from Amos et al., 2012, 3100 Mga^{-1} from Amos et al., 2013, 2950 Mga^{-1} from Soerensen et al., 2010, and 3000 Mga^{-1} from Driscoll et al., 2013). Globally, mercury dry deposition is 2330 Mga^{-1} (58 % Hg(0), 40 % Hg(II) and 2 % Hg(P)), and wet deposition is 2900 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.

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

Intercontinental transport of atmospheric mercury

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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 Mg a^{-1} for total mercury deposition in North America, which was lower than that in this study (437.4 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.

Intercontinental
transport of
atmospheric mercury

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures



Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Intercontinental transport of atmospheric mercury

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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,

ME and MA) except EA. The two RSI relationships (NA → EU and NA → MA) indicate that NA mainly acts as a primary receptor region for SMC and mercury dry deposition, but it acts as a secondary source region for mercury wet deposition.

3.5 Seasonal variation on mercury deposition contributions over other regions from East Asia

As the largest source region, EA strongly affects the spatial distribution of global atmospheric mercury because of its substantial anthropogenic emissions. However, temporal variation of influence patterns on other regions from EA is also important to understand. To understand the temporal variation of influence patterns on the ten receptor regions from EA, we calculate seasonal POFs of mercury dry and wet deposition for each receptor region. Combined with their annual POFs from EA, we quantify seasonal patterns among them (Fig. 7).

Figure 7a illustrates that mercury dry deposition contributions over the ten receptor regions from EA have two seasonal patterns. First, NA, EU, RU, ME and MA (black filled marks) have low values (seasonal POF < annual POF) in winter and high values (seasonal POF > annual POF) in summer. RU is a typical region whose wintertime POF is only half that of its summertime POF. Second, other regions, such as SE and IN, have low values in summer and high values in other seasons (SE in winter and IN in spring). SE is a typical region whose POF in winter is nearly three times that of its summertime value. Seasonal variation on dry deposition contributions over the remaining regions is not apparent. Figure 7b shows that seasonal variation on mercury wet deposition contributions over NA, EU, RU, ME and MA (black filled marks) from EA is similar to the results for dry deposition. Slightly higher values in autumn are the only difference. Compared with dry deposition, seasonal variation on wet deposition contributions over the remaining regions (no filled marks) is not apparent, including SE and IN.

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, with the

Intercontinental transport of atmospheric mercury

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



**Intercontinental
transport of
atmospheric mercury**

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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 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.

Intercontinental transport of atmospheric mercury

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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,

**Intercontinental
transport of
atmospheric mercury**

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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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**Intercontinental
transport of
atmospheric mercury**

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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**Intercontinental
transport of
atmospheric mercury**

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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Intercontinental transport of atmospheric mercury

L. Chen et al.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


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Intercontinental transport of atmospheric mercury

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

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**Intercontinental
transport of
atmospheric mercury**

L. Chen et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



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- 25

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 ^a	Hg(0)	ng m ⁻³	1.45	1.14	1.59	1.53	1.19	1.41	1.97	1.29	0.97	1.32	1.48
	Hg(II)	pg m ⁻³	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 ⁻³	2.36	0.86	9.93	8.80	12.66	18.28	48.58	0.80	3.47	18.89	15.87
Local ^b	Hg(0)	ng m ⁻³	0.08	0.08	0.18	0.11	0.04	0.24	0.81	0.11	0.02	0.05	0.07
		%	5	7	11	7	3	17	41	9	2	4	5
	Hg(II)	pg m ⁻³	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 ⁻³	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 ^c (Others)	Hg(0)	ng m ⁻³	0.39	0.18	0.41	0.44	0.28	0.31	0.22	0.32	0.15	0.39	0.46
		%	27	16	26	29	24	22	11	25	16	30	31
	Hg(II)	pg m ⁻³	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 ⁻³	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 ^c (Nature)	Hg(0)	ng m ⁻³	0.98	0.86	1.01	0.99	0.86	0.85	0.95	0.85	0.77	0.88	0.95
		%	68	76	64	65	72	60	48	66	80	67	64
	Hg(II)	pg m ⁻³	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 ⁻³	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

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

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

^c“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.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Intercontinental transport of atmospheric mercury

L. Chen et al.

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

The categories are defined same as Table 1.

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


Intercontinental transport of atmospheric mercury

L. Chen et al.

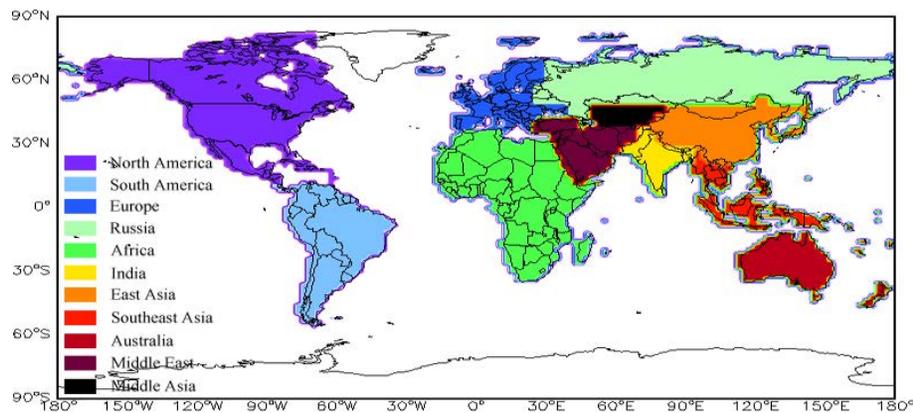
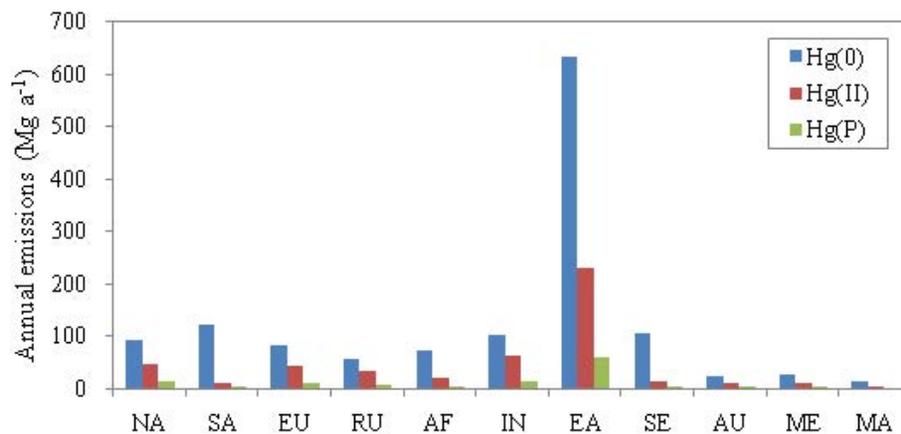
[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[⏪](#)[⏩](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)

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

**Intercontinental
transport of
atmospheric mercury**

L. Chen et al.

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

Intercontinental transport of atmospheric mercury

L. Chen et al.

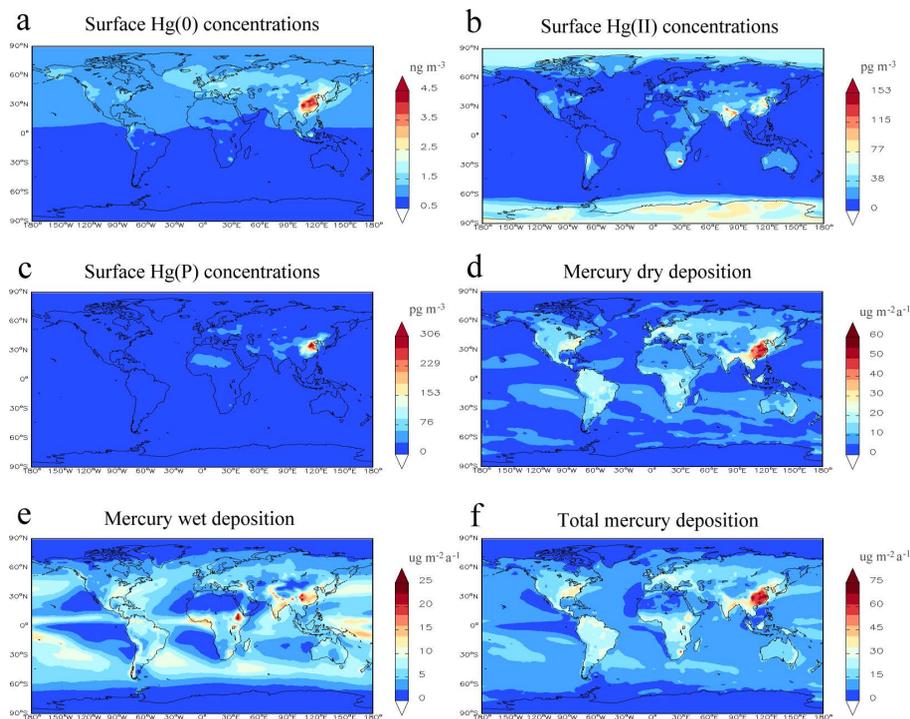


Fig. 3. Global spatial distributions of annual average SMC and mercury deposition: **(a)** surface Hg(0) concentrations; **(b)** surface Hg(II) concentrations; **(c)** surface Hg(P) concentrations; **(d)** mercury dry deposition (Hg(0) dry deposition + Hg(II) dry deposition + Hg(P) dry deposition); **(e)**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

[Title Page](#)
[Abstract](#)
[Introduction](#)
[Conclusions](#)
[References](#)
[Tables](#)
[Figures](#)
[◀](#)
[▶](#)
[◀](#)
[▶](#)
[Back](#)
[Close](#)
[Full Screen / Esc](#)
[Printer-friendly Version](#)
[Interactive Discussion](#)


Intercontinental transport of atmospheric mercury

L. Chen et al.

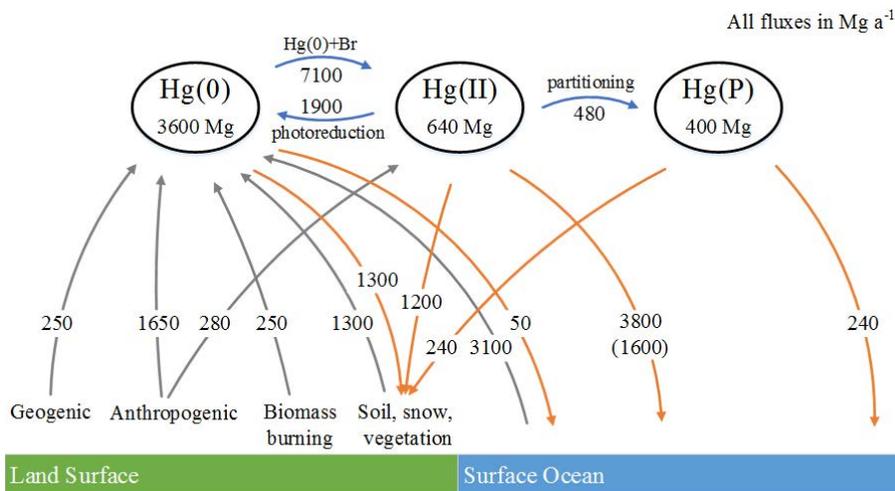


Fig. 4. Global budget of atmospheric mercury derived from this study. The fluxes in parentheses indicate uptake of Hg(II) by sea-salt aerosol.

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion



Intercontinental transport of atmospheric mercury

L. Chen et al.

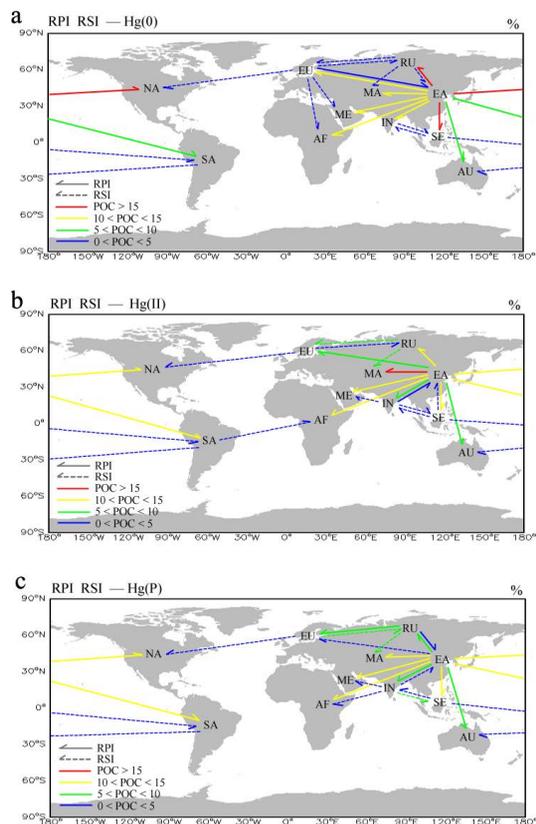


Fig. 5. Intercontinental influence patterns based on the RPI (region of primary influence) and RSI (region of secondary influence) for SMC: **(a)** Hg(0); **(b)** Hg(II); **(c)** Hg(P). Arrows point in the direction of influence from RPI or RSI to a receptor region. Colors indicate the magnitude of POC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

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

Intercontinental transport of atmospheric mercury

L. Chen et al.

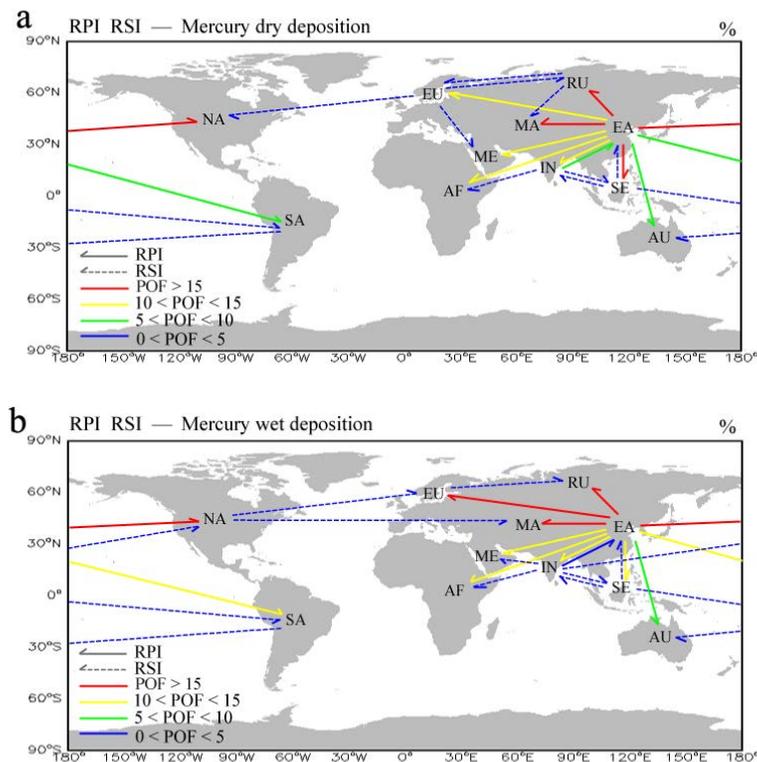


Fig. 6. Intercontinental influence patterns based on the RPI (region of primary influence) and RSI (region of secondary influence) for mercury deposition: **(a)** mercury dry deposition; **(b)** mercury wet deposition. The arrows and colors are defined same as Fig. 5.

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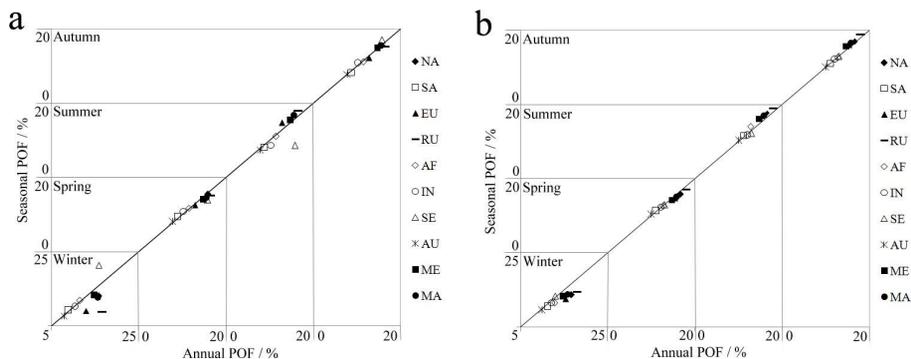


Fig. 7. Seasonal variation on mercury deposition contributions over other regions from East Asia (EA): **(a)** mercury dry deposition; **(b)** mercury wet deposition. Four period zones are defined (winter, spring, summer and autumn) and each period zone has its own ranges of POF values. The line is for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

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Printer-friendly Version

Interactive Discussion

