Reply to Comments from Reviewer #1

We thank the editor and reviewers' comments which help us to improve the manuscript. We have carefully revised our manuscript following the reviewers' comments. A point-to-point response is given below. The reviewers' comments are in black and our replies are in blue.

To reviewer

Comment 1:

The authors mention that mercury species were measured but only GEM data are presented and their trend calculated. What was the contribution of GOM and PBM? Could they contribute to the trend? This is important to discuss because the regional atmospheric Hg emissions in Section 2.5 are probably not only those of GEM but of total mercury. What was the seasonal variation of GOM and PBM? Could it provide some additional evidence for the reasons of GEM seasonal variation?

Response:

The average GOM and PBM concentrations during the studied period were 14.81 ±13.21 pg m^{-3} and 20.10 ±34.02 pg m^{-3}, which accounted for 0.68% and 0.92% in total Hg, respectively. Therefore, the contribution of GOM and PBM to total Hg trend was supposed to be negligible. The downward trend of atmospheric Hg was dominated by the GEM concentration.

It is true that anthropogenic Hg emission inventories included GEM, GOM and PBM emissions. However, the residence time of GOM and PBM is shorter than that of GEM, generally several days to a few weeks for GOM and PBM and 0.5 – 2 year for GEM (Schroeder and Munthe, 1998). In addition, the concentrations of GOM and PBM were affected by emissions, weather condition and depositon processes simultaneously. Therefore, the GEM concentrations in the air of a background site are primarily impacted by GEM emissions. Thus, the regional atmospheric Hg emissions in Section 2.5 are GEM emissions instead of total Hg emissions. We have added sentences to make this point clear in Section 2.5.

Figure R1 showed the seasonal variation of GOM and PBM from March 2014 to February 2015. Considering that the concentrations of GOM and PBM were affected by emissions, weather condition and depositon processes simultaneously, we need more researches to

determine the dominant impact factors. So it is hard to get some additonal evidence for the reason of GEM seasonal variation from the seasonal variation of GOM and PBM.

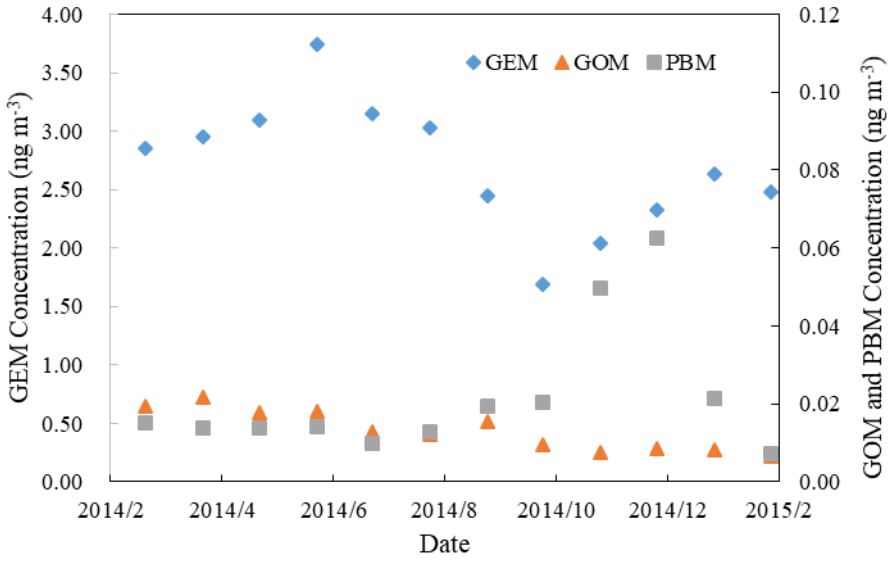


Figure R1. Monthly variation of GEM, GOM and PBM at Chongming from March 2014 to February 2015

The calculation process of GEM emissions is revised as below.

"Regional atmospheric GEM emissions by month are calculated by using both the technology-based emission factor methods and transformed normal distribution function method. Detailed introduction of these two methods and the speciation profile of the emitted Hg for each sector are described in our previous study (Wu et al., 2016)."

See the revised manuscript, line 203-206

Comment 2 :

Section 3.1: Averages and their standard deviations should always be stated with the number of measurements because only then statistical tests for significance of differences can be made. In line 194 the authors claim that GEM concentrations in 2014 were significantly higher than… - at which significance level? Line 199: the annual decrease rate should be given with its uncertainty and number of months.

Response:

Both the standard deviation and the number of measurements have been added in the text for

statistical tests of significance of differences.

The GEM concentrations in 2014 were significantly higher than the background concentration of Northern Hemisphere at the significance level with p value less than 0.01.

"The GEM concentrations in 2014 were higher (*t* test, $p<0.01$) than the Northern Hemisphere back-ground concentration (about 1.5 ng m^{-3}) (Sprovieri et al., 2010) and those measured in other remote and rural locations in China (Zhang H et al., 2015; Fu et al., 2008a; Fu et al., 2009)."

See revised manuscript, line 228 - 231.

The annual decrease rate has been given with its uncertainty and the number of months. In addition, the number of valid data to calculate the monthly average is listed in Table S3.

"During this period, monthly GEM concentrations showed a significant decrease with a rate of -0.60 ± 0.08 ng m^{-3} yr^{-1} (R^2=0.64, p<0.01 significance level, n = 32) (Figure 2a)."

See revised manuscript, line 233 - 234.

Table S3. The number of valid data during sampling period

Year	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
2014			5914	6125	6493	5568	4634	6255	6491	7106	7578	5564
2015	5227	4532	5216	3392	4072	4797	7591	6538	3434	2223	4363	8833
2016			1370	8293	7476	5884	5424	5641	3561	4544	6292	4589

See revised manuscript, supporting information Table S3.

Comment 3:

Figure 3: It is not clear how the points in Figure 3 were calculated? Were the data detrended before the averaging? In view of the strong downward trend they should be. What is the standard deviation or standard error of the monthly means – please show them as vertical bars in Fig. 3. Are the differences between the months statistically significant? This is a precondition for the discussion of the seasonal variation.

Response:

The GEM concentrations in the same month but different years were averaged to get monthly

average during sampling period. The monthly GEM concentrations were detrended before the average (Figure 2). The standard deviations of the monthly means have been added.

"According to the decomposition result (Figure 2c), we observed strong seasonal cycle at Chongming. The GEM concentrations were highest in July and lowest in September, so GEM concentrations in the same month from different years were averaged to understand the detrended seasonal circle (Figure 3). The error bars in the Figure 3 meant the standard deviation of the monthly average."

See revised manuscript, line 270 –274.

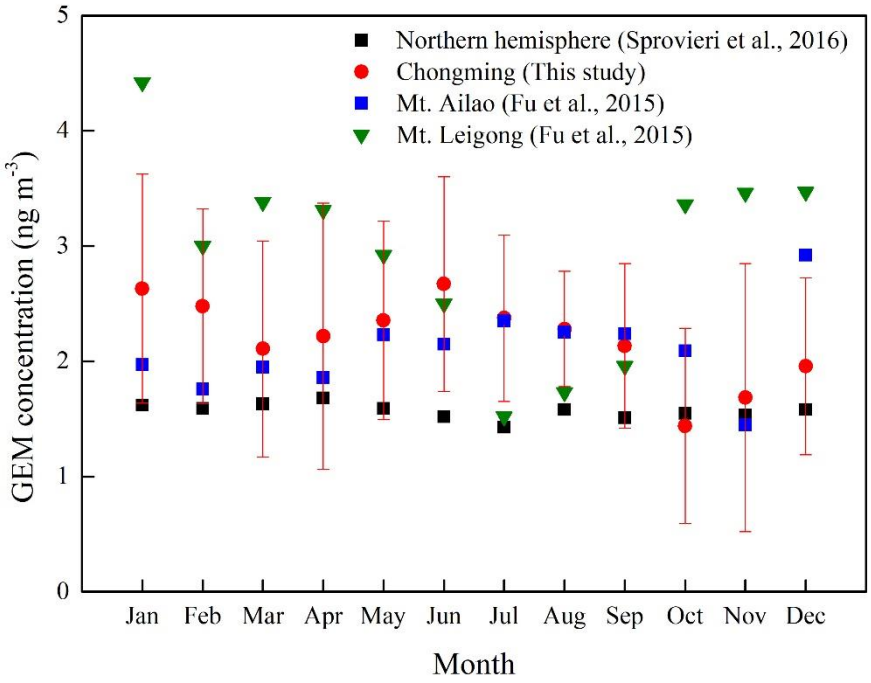


Figure 3. Monthly variations of GEM concentration at remote sites in China

See revised manuscript at Figure 3.

The difference between month are statistically significant (F test, $p<0.001$). In addition, the observed GEM concentration signal was decomposed (signal = trend + seasonal + random, example here: https://anomaly.io/seasonal-trend-decomposition-in-r/). By using this method, we also observed very strong detrended seasonal cycle where GEM peak was observed in July

and the GEM trough was in September.

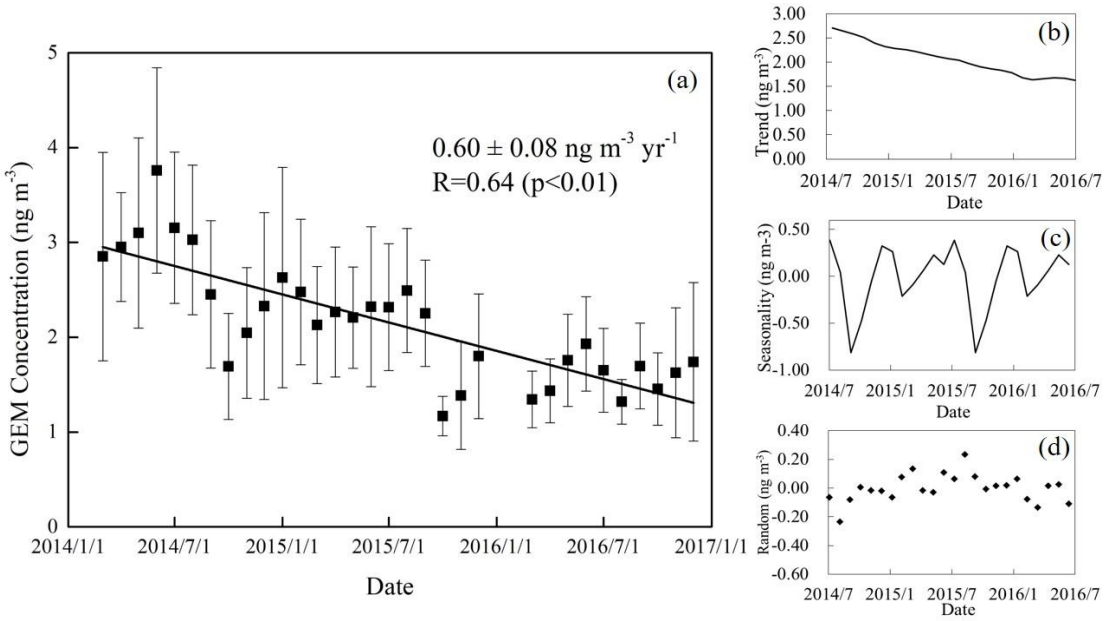


Figure 2. Monthly average GEM concentrations during the studied period (a) observed monthly GEM concentrations (b) GEM trend after decomposition (c) GEM seasonality after decomposition (d) GEM random after decomposition

Note: The observed concentrations during July 2015-April 2016 were TGM concentrations indeed due to the problems of Tekran 1130/1135. However, the GOM concentrations at Chongming island accounted for less than 1% of TGM. Thus, the GEM concentrations were approximated to TGM concentrations during July 2015-April 2016.

Comment 4:

4.1 Figure 4 and its capture: This figure needs substantial revision to illustrate the point the authors make and to make it understandable for the readers. Negative emissions are deposition fluxes and should be named as such. Thus “natural emissions” in spring, autumn and winter are in fact deposition fluxes. Net fluxes are needed to illustrate the point made by the authors but they are not shown. The capture should also state that it is about the emissions and depositions in the YRD region?

Response:

The natural emissions in the manuscript are defined as the followings.

Nature emissions=bi-directional Hg flux × studied period × studied area.

Therefore, emissions and flux are different concepts in this study. We use "natural emissions" instead of "natural flux" to correspond to "anthropogenic emissions". It should be pointed out that the natural emission is a concept of net natural emission, which reflected a net effect of two competing processes (Zhang, 2009): total natural Hg emissions and total Hg deposition. When the value is positive, it means the net effect is Hg emissions to air. Otherwise, Hg deposites. We have made this concept clear in both text and Figure 4.

"The GEM emissions from natural sources E_N are calculated as followings.

$$E_N = \sum_i F_i \times A_i \times t \quad (6)$$

Where F_i is a bi-directional Hg flux of canopy i, ng km^{-2} yr^{-1}; A is the studied area, km^{-2}; t is the studied year, yr. The bi-directional Hg flux was obtained from the study of Wang et al. (2016) directly. It should be pointed out that the natural emission is a concept of net emission in this manuscript, which reflected a net effect of two competing processes (Zhang, 2009): total Hg natural emissions and total Hg deposition. The total natural emissions included primary natural release and re-emission of legacy Hg stored in the terrestrial and water surface (Wang et al., 2016). When the value is positive, it means the net effect is Hg emissions to air. Otherwise, Hg deposited."

See revised manuscript at line 216 – line 224.

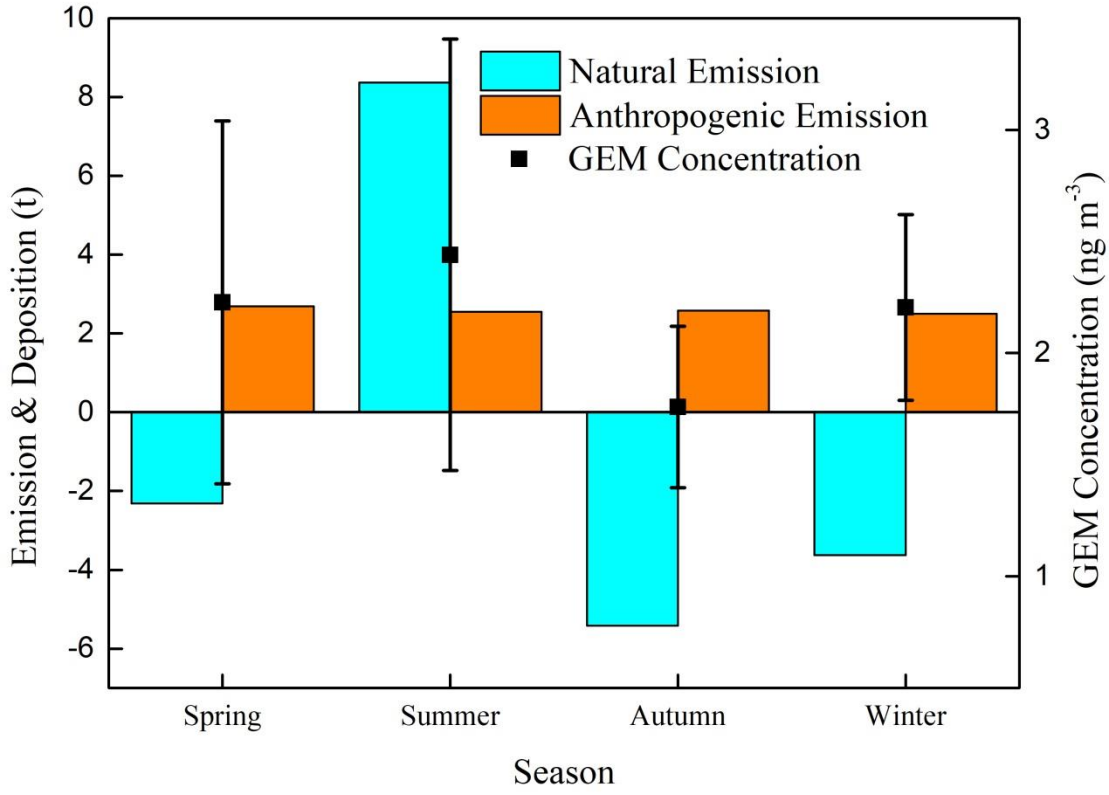


Figure 4. Seasonal cycle of GEM concentrations and natural emissions during 2014-2016. The error bars represent the standard deviation of seasonal average. Positive values of natural emissions represent Hg emitted to air. Otherwise, Hg deposited.

See the revised manuscript Figure 4.

4.2 This will rise a problem: trajectory analysis in section 3.4 shows large influence of transport from the NW provinces of China outside of the YRD region. How does this transport influence the seasonal variation?

Response:

Trajectory outside of the YRD region showed similar seasonal variation as those passes through YRD region. The original definition of "NW" "SW" and "EAST" did not clearly distinguish the pathway of trajectory. Thus, the original "NW" actually contains the Jiangsu province of YRD region. In our revised manuscript, the trajectories were grouped into 3 clusters: NCP, SW-YRD and ABROAD. The NCP mainly passed through north China plain and around regions but not via the YRD regions; the SW-YRD passed through the YRD regions before arriving to Chongming island; the ABROAD mainly originated from the East China Seas, South Korea,

Japan and Northeast Asia continent, and then arrived to our monitoring sites directly without passing the mainland China. From Figure S3, we can see that the NCP cluster and ABROAD cluster showed similar seasonal variation as cluster SW-YRD. High GEM concentrations were observed in summer.

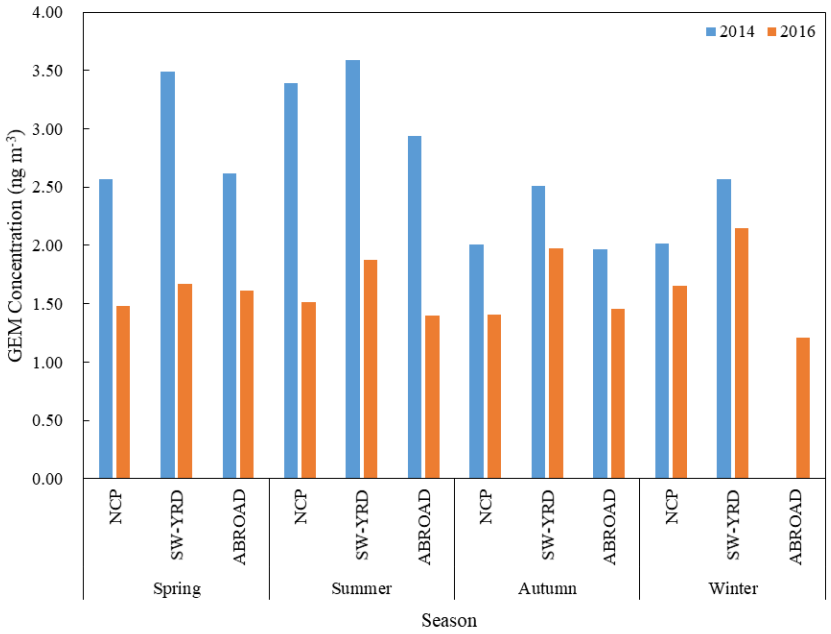


Figure S3. The seasonality of GEM concentration in the NCP, SW-YRD and ABORD region (No trajectory transport though ABROAD in winter of 2014)

We also revised the manuscript as below.

"Transport also overall enhanced the observed seasonal variation of GEM concentrations at Chongming Island. According to the statistics of backward trajectories in section 3.4, the GEM concentrations in the air mass which did not pass via the YRD regions also showed high GEM concentration in warm season in 2014 (Figure S3).**"**

See the revised manuscript at line 309 - 312

Comment 5:

Line 243-245: "The annual emissions from both natural source and anthropogenic source… was 0.75 and 10.3 t, respectively" – the reader may think that natural emissions make less than 10% of the anthropogenic ones and cannot thus be responsible for the seasonal variation. One has to look in Fig. 4 to find out that the "natural emissions" are a sum of natural emissions in one season and "natural" deposition fluxes in three seasons. That provokes a question: how is

anthropogenically emitted mercury removed from the atmosphere if there are only "natural" deposition fluxes? Please use the correct terminology and separate the natural and anthropogenic emissions from the deposition fluxes of both.

Response:

Sorry for the misunderstanding. To avoid confusion, we deleted the sentence of "The annual emissions from both natural source and anthropogenic source… was 0.75 and 10.3 t, respectively" in the revised manuscript. The impact of anthropogenic emissions and natural emissions were discussed separately. The anthropogenic emissions were in the range of 2.5 – 2.7 t while natural emissions varied from -5.4 – 8.4 t in different season. Thus, one important conclusion of our study is that the seasonal GEM cycle was dominated by the natural emissions.

"Source emission is one significant factor on GEM concentrations in the air. The GEM concentrations at a remote site are generally regarded under the impact of regional emissions. Therefore, the emissions in the YRD regions (Anhui, Zhejiang, Jiangsu, and Shanghai) were calculated. However, the anthropogenic emissions were in the range of 2.5-2.7 t, which is almost unchanged. Compared to the anthropogenic emissions, we observed almost synchronized trends between natural emissions and air Hg concentrations in Figure 4."

See the revised manuscript at line 291 - 306

It is difficult to distinguish whether the deposited Hg is from natural sources or anthropogenic sources. Therefore, the bi-directional Hg flux scheme contained total Hg deposition flux (both so called "natural" and "anthropogenic" deposition fluxes) (Zhang et al., 2009). And the natural Hg emissions in this study have considered the removal of anthropogenic Hg emissions. We have clearly defined the concept of "natural emissions" in this study. The natural emission is a concept of net natural emission, which reflected a net effect of total natural Hg emissions and total Hg deposition amount. Therefore, the data of natural emissions in the four seasons contains both emissions and deposition. The positive value in summer means that net effect is Hg emissions to air. In other three seasons, Hg deposited. Detailed revision is described as follows.

"The GEM emissions from natural sources E_N are calculated as followings.

$$E_N = \sum_i F_i \times A_i \times t$$

where F_i is a bi-directional Hg flux of canopy i, ng km^{-2} yr^{-1}; A is the studied area, km^{-2}; t is the studied year, yr. The bi-directional Hg flux was obtained from the study of Wang et al. (2016) directly. It should be pointed out that the natural emission is a concept of net emission in this manuscript, which reflected a net effect of two competing processes (Zhang, 2009): total Hg natural emissions and total Hg deposition. The total natural emissions included primary natural release and re-emission of legacy Hg stored in the terrestrial and water surface (Wang et al., 2016). When the value of E_N is positive, it means the net effect is Hg emissions to air. Otherwise, Hg deposited."

See the revised manuscript at line 215 - 223

Comment 6:

The results of PCA analysis and Table 2: The authors attribute the factor 2 to "exchange of PBL with free troposphere" but do not explain why. Last row in the table 2 called "variance explain" lists for 2016 exchange of PBL with the free troposphere 75.735 which together with 50.625 for "combustion" makes more than 100. As such the units of "variance explain" cannot be percent. What are the numbers in this row and does it make sense to present them with three valid numbers after decimal point?

Response:

We have added explanation as follows.

"Considering the low loading of CO and high loading of O_3, the factor 2 can be viewed as a sign of the transport of air mass from stratosphere (Fishman and Seiler, 1983; Jaffe, 2010). The air mass from stratosphere will increase the O_3 concentration. O_3 react with NO, which makes a negative correlation with NO. However, the low loading on GEM of factor 2 indicated that Factor 2 had no relationship with GEM concentrations at Chongming from the aspect of whole year data."

See the revised manuscript at line 345 - 350

Sorry for the typo. We have corrected this in Table 1. The variance explain showed the

contribution ratio of factor 1 and factor 2 in the total variance. We have revised the data to two valid numbers after decimal point.

Table 1. PCA component loading of GEM and other air pollutants

Air pollutants	2014		Air pollutants	2016	
	Factor 1	Factor 2		Factor 1	Factor 2
SO_2	**0.76**	0.14	SO_2	**0.82**	-0.09
NO_X	**0.76**	-0.20	NO_X	**0.70**	**-0.52**
O_3	-0.11	**0.98**	O_3	-0.41	**0.97**
$PM_{2.5}$	**0.85**	0.05	$PM_{2.5}$	**0.88**	0.05
GEM	**0.66**	0.02	GEM	**0.78**	-0.19
CO	**0.79**	0.12			
Component	Combustion	Transport of air mass from stratosphere	Component	Combustion	Transport of air mass from stratosphere
Variance explain (%)	49.36	17.53	Variance explain (%)	50.63	25.10

Note: Text in bold phase were regarded as high loading (factor loading>0.40 or <-0.40)

Comment 7:

Table 3: The numbers are probably annual emissions but the capture does not say it. The year of the emissions is not given. I wonder about the "other SO2 sources" which are substantially larger than all coal, oil, and biomass burning taken together. If it is not an error, what are the "other SO2 sources"?

Response：

Thank for the comments. It is the emission in 2014. The emission for "other SO_2 sources" is a typo and we have revised this. The other sectors contain municipal solid incineration, copper smelting, aluminum production, gold production, other coal combustion, stationary oil combustion, and cremation. Table 2 has been revised as follows.

Table 2. Emissions of the main air pollutants in YRD region in 2014

Sectors	Annual emissions			
	SO_2 (kt)	NO_x (kt)	$PM_{2.5}$ (kt)	GEM (t)
Coal-fired power plants	918.31	991.62	118.42	14.00
Coal-fired industrial boilers	311.03	271.94	79.91	9.80
Residential coal combustion	68.48	42.11	163.93	0.40
Cement clinker production	207.48	371.13	208.02	4.70
Iron and steel production	480.97	142.80	169.84	2.30
Mobile oil combustion	38.43	1786.74	98.00	1.90

Other sectors	348.83	316.28	382.48	2.50

"The studied emission sectors included coal-fired power plants, coal-fired industrial boilers, residential coal-combustion, cement clinker production, iron and steel production, and other small emission sectors (eg., zinc smelting, lead smelting, municipal solid incineration, copper smelting, aluminum production, gold production, other coal combustion, stationary oil combustion, and cremation)."

See the revised manuscript at line 209 - 212

Comment 8:

Chapter 3.4 misses a major point: Table 3 of SO_2, NO_x, $PM_{2.5}$, and GEM emissions is only for one undefined year and only for the YRD region. Table 4 and Figure 6 show a dominant influence of transport from NW of China which is mostly outside of the YRD region. To illustrate convincingly the major conclusion of the paper one would need a table with the inventories for NW and SW (perhaps separately) and for 2014 and 2016.

Response :

According to our response to comment 4, we have adjusted the cluster. Based on the adjusted cluster, we added the emissions of SO_2, NO_x and $PM_{2.5}$ in both 2014 and 2016 to illustrate the change of emission inventory in NCP and SW-YRD region (Table S5). According to the table, we observed obvious emission decline of the above pollutants.

Table S5. Emission inventories of the main pollutants from the studied regions in 2014 and 2016

Air pollutants	2014		2016		Decline proportion	
	NCP	SW-YRD	NCP	SW-YRD	NCP	SW-YRD
$PM_{2.5}$ (kt)	2019	1209	1849	1109	-8%	-8%
NO_x (kt)	5697	4022	5424	3855	-5%	-4%
SO_2 (kt)	3780	1993	3450	1780	-9%	-11%
GEM (t)	118	72	103	67	-13%	-7%

Note: According to the contribution of trajectory, the dominant provinces in the NCP region included Beijing, Tianjin, Hebei, Shandong and Liaoning province. The SW-YRD mainly contained Shanghai, Zhejiang, Jiangsu, Jiangxi and Anhui province.

See the revised manuscript at line 377 - 379

Comment 9:

Table 1: The paper is about regional trend and I wonder why it is necessary to discuss global background trends in such detail. Also because the reasons of downward trend of mercury at many background stations of the world at the time of increasing global emissions is still not well understood (compare Horowitz et al., EST 48, 10242-10250, 2014, with Soerensen et al., GRL, 39, L21810, doi:10.1029/2012GL053736, 2012).

Response :

We agree with your valuable comment. Our paper is about regional trend, so we have curtailed the discussion and move the original Table 1 to supporting information (Table S4) so as to focus on our topic.

Comment 10:

Figure S1: Here the least square fit of 4 points provides R2 of 0.487 for January and 0.613 for February for which the authors claim $p < 0.01$ in both cases. This is surely incorrect because 4 points result statistically in only 2 degrees of freedom. Please explain.

Response :

The purpose of original Figure S1 is to obtain the fitting curve so as to calculate the Hg concentrations in the January and February of 2016. But it did not work actually because the points are not enough. Therefore, we deleted the original Figure S1. Instead, the average value of the GEM concentrations in January of 2015 and 2017 were used to represent the GEM observation in January of 2016. The same method is used to simulate the GEM observation in February of 2016.

The Figure S1 was revised as below.

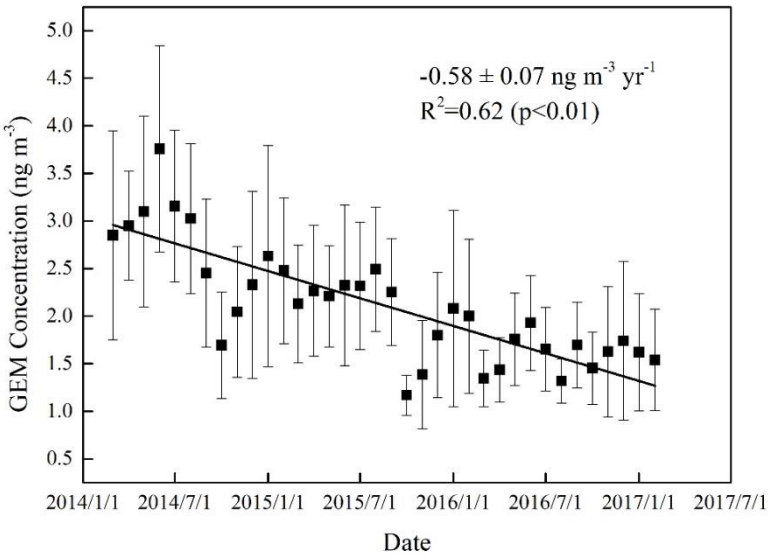


Figure S1. The trend of monthly average GEM concentration from March 2014 to February 2017. The monthly average of GEM in January of 2016 is simulated as the average value that in the January of 2015 and 2017. The same method is used for the data in February of 2016.

Comment 11:

Figure S2: The downward annual rate should be given with its standard error.

Response:

The downward annual rate was given with its standard error as follows. See the Figure S1 in the response of comment 10.

Comment 12:

Line 51: "Both GOM and PBM are more soluble." than what? PBM is not necessarily more soluble than GEM but it is scavenged by wet deposition. Low solubility of GEM need to be mentioned before this statement.

Response :

The statement has been revised as follows.

"GOM is more soluble than GEM, and PBM can be quickly scavenged by both dry and wet deposition. Therefore, the residence time of both GOM and PBM is shorter than that of GEM, generally several days to a few weeks for GOM and 0.5 – 2 year for GEM (Schroeder and

Munthe, 1998). ”

See the revised manuscript at line 52 - 55.

Comment 13:

Lines 71/72: …have been estimated to decrease…

Response :

We have revised the manuscript as follow.

“However, recently atmospheric Hg emissions in China have been estimated to decrease since 2012 (Wu et al., 2016).”

See the revised manuscript at line 82

Comment 14:

Line 86: …is located…

Response:

We have revised the manuscript as suggested.

“As China’s third largest island, Chongming Island is located in the east of Yangtze River Delta region with a typical subtropical monsoon climate.”

See the revised manuscript at line 97

Comment 15:

Lines 102/103: … the error between gold trap A and gold trap B was limited to…? Probably the difference instead of error was limited. What happens if the difference is more than the limit?

Response:

We have replaced the error with differences. If the difference is more than the limit, it means that the gold traps are passivizing and we need to replace the old gold trap A and gold trap B.

Comment 16:

Lines 171/172: “uncertainties” would be better than “errors”

Response:

We have revised the manuscript as suggested.

Comment 17:

Line 199: Please state the decrease rate with its standard error.

Response

We have revised the manuscript as suggested.

Comment 18:

Lines 207-209: A reference to Martin et al (2017) is not correct because the paper does not contain annual averages and the authors of this paper do not mention a gap in the measurements between 2004 and 2007. The correct reference would be: annual average GEM concentration decreased from 1.29 ng m-3 in 1996 to 1.19 ng m-3 in 2004 (Slemr et al., GRL 35, L11807, doi:10.1029/2008GL033741, 2008) and were increasing from 0.93 ng m-3 in 2007 (Slemr et al., ACP 15, 3125-3133, 2015) until 2016 (Martin et al, 2017).

Response:

We have revised the manuscript as suggested.

"In South Africa, annual average GEM concentration at Cape Point decreased from 1.29 ng m^{-3} in 1996 to 1.19 ng m^{-3} in 2004 (Slemr et al., 2008) and were increasing from 0.93 ng m^{-3} in 2007 (Slemr et al., 2015) until 2016 (Martin et al, 2017)."

See the manuscript at line 257– 259

References:

Fishman J, Seiler W. Correlative Nature of Ozone and Carbon Monoxide in the Troposphere: Implications for the Tropospheric Ozone Budget. Journal of Geophysical Research, 88(C6), 1983.

Jaffe, D.: Relationship betwen surface and free tropospheric ozone in the western U.S., Environmental Science & Technology, 45, 432-438, 10.1021/es1028102, 2010.

Li S, Gao W, Wang S X, Zhang L, Li Z J, Wang L, Hao J M. Characteristics of Speciated Atmospheric Mercury in Chongming Island, Shanghai. Environmental Science 2016, 37(9): 3290 -

3299.

Pirrone N, Cinnirella S, Feng X, Finkelman R B, Friedli H R, Leaner J, Mason R, Mukherjee A B, Stracher G B, Streets D G, Telmer K. Global Mercury Emissions to the Atmosphere from Anthropogenic and Natural Sources. Atmospheric Chemistry and Physics, 2010, 10(13): 5951-5964.

Slemr F, Angot H, Dommergue A, Magand O, Barret M, Weigelt A, Ebinghaus R, Brunke E G, Pfaffhuber K A, Edwards G, Howard D, Powell J, Keywood M, Wang F. Comparison of Mercury Concentrations Measured at Several Sites in the Southern Hemisphere. Atmospheric Chemistry and Physics, 2015, 15(6): 3125-3133.

Sprovieri F, Pirrone N, Bencardino M, amp, apos, Amore F, Carbone F, Cinnirella S, Mannarino V, Landis M, Ebinghaus R, Weigelt A, Brunke E-G, Labuschagne C, Martin L, Munthe J, Wängberg I, Artaxo P, Morais F, Barbosa H d M J, Brito J, Cairns W, Barbante C, Diéguez M d C, Garcia P E, Dommergue A, Angot H, Magand O, Skov H, Horvat M, Kotnik J, Read K A, Neves L M, Gawlik B M, Sena F, Mashyanov N, Obolkin V, Wip D, Feng X B, Zhang H, Fu X, Ramachandran R, Cossa D, Knoery J, Marusczak N, Nerentorp M, Norstrom C. Atmospheric Mercury Concentrations Observed at Ground-Based Monitoring Sites Globally Distributed in the Framework of the Gmos Network. Atmospheric Chemistry and Physics, 2016, 16(18): 11915-11935.

Wang X, Lin C J, Yuan W, Sommar J, Zhu W, Feng X. Emission-Dominated Gas Exchange of Elemental Mercury Vapor over Natural Surfaces in China. Atmospheric Chemistry and Physics, 2016, 16(17): 11125-11143.

Wang X, Lin C J, Feng X. Sensitivity Analysis of an Updated Bidirectional Air–Surface Exchange Model for Elemental Mercury Vapor. Atmospheric Chemistry and Physics, 2014, 14(12): 6273-6287.

Zhang L, Wang L, Wang S, Dou H, Li J, Li S, Hao J. Characteristics and Sources of Speciated Atmospheric Mercury at a Coastal Site in the East China Sea Region. Aerosol and Air Quality Research, 2017, 17(12): 2913-2923.

Zhang, L. M., Wright, L. P., and Blanchard, P.: A review of current knowledge concerning dry deposition of atmospheric mercury, Atmos. Environ., 43, 5853-5864, 2009.

Zhang Y, Jacob D J, Horowitz H M, Chen L, Amos H M, Krabbenhoft D P, Slemr F, St Louis V L, Sunderland E M. Observed Decrease in Atmospheric Mercury Explained by Global Decline in Anthropogenic Emissions. Proceedings of the National Academy of Sciences of the United States of America, 2016, 113(3): 526.

Reply to Comments from Reviewer #2

We thank the editor and reviewers' comments which help us improve the manuscript. We have carefully revised our manuscript following the reviewers' comments. A point-to-point response is given below. The reviewers' comments are in black and our replies are in blue.

To Reviewer

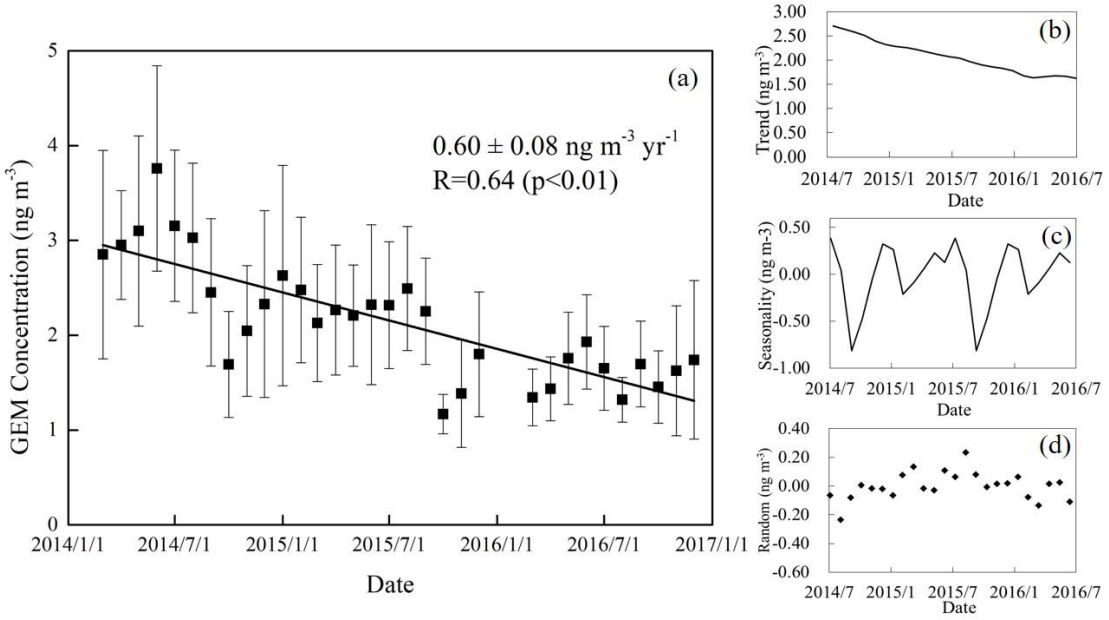
Major Comment:

I really think you should perform a trend decomposition of the signal (signal = seasonal + trend + random, example here: https://anomaly.io/seasonal-trend-decomposition-in-r/). There is a very strong seasonal cycle and you conclude that "the seasonal GEM cycle was dominated by the natural emissions". However, how can you explain that the seasonality is way more pronounced in 2014? To me, presenting emissions inventories is not convincing enough; how you can you be sure that the decreasing trend is not driven by a change in seasonality?

Response:

The trend decomposition of the signal is performed in Figure 2 (signal = trend + average variation + random). By using this method, the decreasing trend was shown in Figure 2b and the seasonality is approximately 12 months. In each cycle, we observed very strong seasonal cycle where GEM peak was observed in July and the GEM trough was in September.

The seasonal variation was more pronounced in 2014 can attribute to the lower wet deposition and GEM oxidation. On one aspect, as a costal site, the Chongming island is abundant with •OH radicals. The increase of O_3 concentration from the summer of 2014 to 2016 may contribute to a higher oxidation of GEM in 2016. On another aspect, higher wet Hg deposition in summer is approximately 6.6 times of that in the winter at Chongming (Zhang et al., 2010). Meanwhile, the rainfall in 2016 summer (546 mm) was higher than the rainfall in 2014 (426 mm). Therefore, the higher oxidation and wet deposition rate of mercury in the summer of 2016 will reduce the peak of GEM concentration and then shrink the concentration difference between summer and winter, which lead to a less pronounced seasonal variation in 2016.



425

426 **Figure 2.** Monthly average GEM concentrations during the studied period (a) observed monthly
427 GEM concentrations (b) GEM trend after decomposition (c) GEM seasonality after
428 decomposition (d) GEM random after decomposition

429 Note: The observed concentrations during July 2015-April 2016 were TGM concentrations
430 indeed due to the problems of Tekran 1130/1135. However, the GOM concentrations at
431 Chongming island accounted for less than 1% of TGM. Thus, the GEM concentrations were
432 approximated to TGM concentrations during July 2015-April 2016.

433

434 The revised manuscript of decomposition trend is revised as below.

435 "From another aspect, the trend decomposition of the GEM concentration signal (signal =
436 trend + seasonal + random) from March 2014 to December 2016 were performed in Figure 2
437 (https://anomaly.io/seasonal-trend-decomposition-in-r/). By using this method, we also
438 observed a pronounced trend (Figure 2b) and the random was limited in the range of -0.24 –
439 0.24 ng m^{-3} (Figure 2d)."

440 **See the revised manuscript at line 234 - 239**

441

442 "From Figure 2, we also observed more pronounced seasonal variation in 2014, which can
443 be attributed to the lower wet deposition and GEM oxidation. On one aspect, as a costal site,
444 the Chongming Island is abundant with •OH. The increase of O_3 concentration from the summer

of 2014 to 2016 may contribute to a higher oxidation of GEM in 2016. On another aspect, higher wet Hg deposition in summer is approximately 6.6 times of that in the winter at Chongming (Zhang et al., 2010). Meanwhile, the rainfall in 2016 summer (546 mm) was higher than the rainfall in 2014 (426 mm). Therefore, the higher oxidation and wet deposition rate of Hg in the summer of 2016 will reduce the concentration difference between summer and winter, which lead to a less pronounced seasonal variation in 2016. Meanwhile, the higher oxidation and wet deposition in 2016 also contributed to the downward trend of GEM by reducing the seasonality in spring and summer (Figure S3)."

See the revised manuscript at line 314 - 324

While SO_2, NO_2, and PM concentrations were monitored, data are not presented nor discussed. Do you also observe a decreasing trend? That would be the best way to support that "air pollution control policies targeting SO_2, NO_2, and PM reductions had significant co-benefits on atmospheric Hg".

Response:

To further verify the cause of downward trend of atmospheric Hg, we give the emission inventory (Table S6) and concentrations (Table S5) of other air pollutants in the studied regions in both 2014 and 2016. Both the emissions and concentrations of SO_2, NO_2, and PM showed a decreasing trend, which is used to support that "air pollution control policies targeting SO_2, NO_2, and PM reductions had significant co-benefits on atmospheric Hg".

"Table 3 showed the detailed data of the three classifications. From 2014 to 2016, the whole China region (NCP, SW-YRD) contributed to 70% of GEM decline at Chongming Island. Considering downward trend of emission inventory and atmospheric pollutant from 2014 to 2016 in NCP and SW-YRD region (Table S5, Table S6), the reason of downward trend can be attributed to the effectiveness of existing air pollution control measures in China (SC, 2013; MEP, 2014)."

See the revised manuscript at line 375 – 379

Table S5. The annual concentration of SO_2, NO_x, O_3 and $PM_{2.5}$ at Chongming site, NCP, and

SW-YRD regions

Year		2014			2016			Change		
Pollutants	Region	NCP	SW-YRD	Chongming	NCP	SW-YRD	Chongming	NCP	SW-YRD	Chongming
$PM_{2.5}$ ($\mu g\ m^{-3}$)		71.93	53.05	25.09	60.75	44.75	23.89	-16%	-16%	-5%
SO_2 ($\mu g\ m^{-3}$)		34.52	21.01	1.60	24.37	16.40	1.47	-29%	-22%	-8%
NO_2 ($\mu g\ m^{-3}$)		45.07	34.34	12.62	41.55	34.40	10.84	-8%	0%	-14%
O_3 ($\mu g\ m^{-3}$)		60.29	56.27	41.70	61.84	60.92	44.38	3%	8%	6%
GEM ($ng\ m^{-3}$)		No data		2.68	No data		1.60	No data		-40%

Note: According to the contribution of trajectory, the dominant provinces in the NCP region included Beijing, Tianjin, Hebei, Shandong and Liaoning province. The SW-YRD mainly contained Shanghai, Zhejiang, Jiangsu, Jiangxi and Anhui province.

Table S6. Emission inventories of the main pollutants from the studied regions in 2014 and 2016

Air pollutants	2014		2016		Decline proportion	
	NCP	SW-YRD	NCP	SW-YRD	NCP	SW-YRD
$PM_{2.5}$ (kt)	2019	1209	1849	1109	-8%	-8%
NO_x (kt)	5697	4022	5424	3855	-5%	-4%
SO_2 (kt)	3780	1993	3450	1780	-9%	-11%
GEM (t)	118	72	103	67	-13%	-7%

Note: According to the contribution of trajectory, the dominant provinces in the NCP region included Beijing, Tianjin, Hebei, Shandong and Liaoning province. The SW-YRD mainly contained Shanghai, Zhejiang, Jiangsu, Jiangxi and Anhui province.

Finally, I wonder why GOM and PBM data are not reported and discussed. Do you also observe a decreasing trend? You may have encountered issues with the speciation unit. If so, was the experimental setup identical in 2014 and 2016, or did you analyze GEM when the speciation unit was working vs. TGM when it wasn't? A discussion on analytical uncertainties would be much welcomed.

The GOM and PBM data are not reported finally due to the following reasons. First, the main purpose of this manuscript is to validate the anthropogenic Hg emissions reduction through observation data. The Chongming sit is a background site. Considering the stability of GEM in

the air, we choose GEM as an index to reflect the emission control effect. Second, the method we developed to explain the GEM trend is not applicable for GOM and PBM. Except emissions, we think the potential reactions in the air are significant factors to impact both GOM and PBM concentrations. But we need more evidence to prove our assumptions. Therefore, we deleted the discussion of GOM and PBM in our final manuscript.

We also observed decreasing trend of PBM. But the GOM kept increasing. Currently, we need more study to explain this phenomenon.

Yes, we also encountered issues with the speciation unit. From July 5, 2015 to April 30 2016, the Tekran 1130/1135 speciation unit was damaged by the rainstorm, the Tekran 2537X were operated without speciation units but with PTFE filter to protect the instrument from particles and sea salt. The average concentration of GOM and PBM during sampling period was 14.81 $\pm$ 13.21 and 20.10 $\pm$ 34.02 pg m^{-3}, respectively. The GOM fraction in TGM was less than 1% at Chongming island (Li et al., 2016; Zhang et al., 2017). Therefore, the GEM concentrations were approximated to TGM concentrations July 5, 2015 to April 30 2016 when the speciation unit does not work, as most of other studies have done (Slemr et al., 2015; Sprovieri et al., 2016). In addition, uncertainty analysis was performed to point out potential impact. The revision is as follows.

"From July 5, 2015 to April 30 2016, the Tekran 1130/1135 speciation unit was damaged by the rainstorm, the Tekran 2537X were operated without speciation units but with PTFE filter to protect the instrument from particles and sea salt. Therefore, the observed concentrations during July 2015-April 2016 were TGM concentrations indeed. However, the GOM concentrations at Chongming Island accounted for less than 1% of TGM (TGM=GOM+GEM). Thus, the GEM concentrations were approximated to TGM concentrations from July 2015 to April 2016."

See the revised manuscript at line 109 – 115

"In our research, random uncertainties of individual measurement had been averaged out and the systematic uncertainties need to be considered. The overall practically achievable systematic uncertainty would be 10% considering that the instrument was not in ideal performance (Slemr et al., 2015; Steffen et al., 2012). For example, slow deactivation of the traps, contamination of the switching valves and leaks would increase the uncertainties but were difficult to quantify (Slemr et al., 2015;Steffen et al., 2012). Because of the consistency of

instrument and the quality assurance/quality control have been paid special attention to during the sampling campaign, the systematic differences of instrument did not affect the huge variation between 2014 and 2016."

See the revised manuscript at line 122 – 129

Comment 1:

Line 26: "GEM concentrations showed a significant decrease with a rate of -0.60 ng/m3/yr". According to Table 1, the rate is -0.52 ng/m3/yr.

Response:

Thanks for the comments. This is a typo in the table. It is -0.60±0.08 ng m^{-3} yr^{-1} actually. We have corrected this in the revised manuscript.

.

Comment 2:

Line 33: "It was find" should be "It was found".

Response:

We have corrected in the manuscript as below.

"It was found that the reduction of domestic emissions was the main driver of GEM decline in Chongming Island, accounting for 70% of the total decline."

See the revised manuscript at line 33 – 34

Comment 3:

Lines 47-48: "In the atmosphere, Hg mainly presents as GEM, accounting for over 95% or the total". Can you please add a reference? Is that also true at your site?

Response:

We have added references in the manuscript. During the sampling period (March 2014 to June 2015 and May 2016 to December 2016), the GOM concentration is 14.81 ± 13.21 pg m^{-3} and GEM concentration is 2.15 ± 0.94 ng m^{-3}. Thus, the GOM concentration accounted for 0.68% and the conclusion in the reference is also true at our site. We also added reference in

our manuscript as below.

"In the atmosphere, Hg mainly presents as GEM, accounting for over 95% of the total in most observation sites (Fu et al., 2015; Li et al., 2016; Zhang et al., 2017)."

See the revised manuscript at line 49

Comment 4:

Line 61-62: "(: : :) there is no official national monitoring network of atmospheric Hg". Out of curiosity, what is the current status of the Asian-Pacific Mercury Monitoring Network (http://nadp.sws.uiuc.edu/newIssues/asia/)? Do you think that Chinese sites will be included?

Response:

Thanks for the comment. Asian-Pacific Mercury Monitoring Network (APMMN) was established in 2013 with founding countries and regions including the U.S, China Taiwan, Thailand, Vietnam, Indonesia, Japan, Korea and Canada (http://apmmn.org/AboutAPMMN.html). APMMN has been monitoring atmospheric mercury deposition in the Asia-Pacific region and holds annual meetings since 2013. Currently, there is no monitoring site of mainland China in the APMMN (see the Figure R2).

Figure R2. The participating country of APMMN (http://apmmn.org/AboutAPMMN.html)

Considering the large Hg emissions in mainland China, including the Chinese sites into the monitoring network will helpful for the research of Hg behavior in the regional or global scale. However, nearly all mercury monitoring sites belong to individual researchers in China currently. Therefore, whether the Chinese sites will be included mainly depend on multiple factors such as individual interests and potential benefit. We also revised our expression in the manuscript as below.

"For the developing countries such as China, limited atmospheric Hg observations have been carried out (Fu et al., 2008b; Zhang H et al., 2016; Hong et al., 2016) and there is no official national monitoring network of atmospheric Hg in mainland China."

See the revised manuscript at line 63 - 66

Comment 5:

Lines 64-67: "Atmospheric Hg emissions in China accounted for 27% of the global total in 2010 (UNEP, 2013), which led to high air Hg concentrations in China. Therefore, atmospheric Hg observations in China are critical to understand the Hg cycling at both regional and global scale". Please define "high" air Hg concentrations. Additionally, in order to emphasize the fact that observations in China are critical to understand the Hg cycling on a global scale, you could perhaps add a sentence about 1) future projections (e.g., Chen et al., 2018; Pacyna et al., 2016), and 2) long-range transport of Chinese emissions to other regions (e.g., Chen et al., 2018; Corbitt et al., 2011; Sung et al., 2018).

Response:

Hg concentration in remote site in China and Northern Hemisphere are compared to illustrate the level of Hg pollution in China. Long-range transport and future projections are added to emphasize observation in China are critical to understand Hg cycling on a global scale.

The related paragraph is revised as below.

"China contributes to the largest Hg emissions in the world and will continue to be one significant Hg emitter for global Hg emissions in the coming future (UNEP, 2013, Wu et al., 2016, Chen et al., 2018; Pacyna et al., 2016). Large Hg emissions in China have led to the average air Hg concentrations of 2.86 ± 0.95 ng m^{-3} (in the range of 1.60-5.07 ng m^{-3}) at the

598 remote sites in China (Fu et al., 2015). Such Hg concentration level is approximately 1.3 ng m-
599 ^{3} higher than the background concentration of GEM in Northern Hemisphere (Zhang et al.,
600 2016;Sprovieri et al., 2017;Fu et al., 2015). In addition, the large Hg emissions in China will
601 also impact the air Hg concentrations in East Asia and even North America through long-range
602 transport (Sung et al., 2018;Zhang et al., 2017)."
603 **See the revised manuscript at line 67 – 75.**
604

605 ***Comment 6:***

606 Lines 93-94: "we used Tekran 2537X/1130/1135 instruments to monitor speciated Hg in the
607 atmosphere". I wonder why GOM and PBM concentrations are not reported in the manuscript.
608

609 ***Response:***

610 We didn't report the GOM and PBM concentration because the main purpose of this
611 manuscript is to validate the anthropogenic Hg emissions reduction through observation data.
612 The Chongming site is a background site. Considering the stability of GEM in the air, we
613 choose GEM as an index to reflect the emission control effect. In addition, the method we
614 developed to explain the GEM trend is not applicable for GOM and PBM. Except emissions,
615 we think the potential reactions in the air are more significant factors for GOM and PBM. But
616 we need more evidence to prove our assumptions. Therefore, we deleted the discussion of GOM
617 and PBM in our final manuscript.
618

619 If concentrations were recorded, it would be interesting to discuss the results. Do you also see
620 a decreasing trend from 2014 to 2016? From 1978 to 2014, the fractions of GEM and PBM
621 decreased, while the GOM emission share gradually increased (Wu et al., 2016). What about
622 the speciation of emissions since 2014? Can you observe a trend in GOM/PBM concentrations?
623

624 ***Response:***

625 Yes. The downward trend of PBM concentration was observed to decrease from 2014 (24.51
626 $\pm$43.31 pg m^{-3}) to 2016 (22.07 $\pm$30.55 pg m^{-3}), which was also consistent with the downwawrd

trend of GEM. However, the GOM concentration increased from (15.41 ± 16.02 pg m^{-3}) in 2014 to (18.97 ± 9.28 pg m^{-3}) in 2016. Speciated Hg emissions were showed in Table R1. All speciated Hg emissions have decreased since 2014 in the YRD regions. However, we observed significant GEM decreasing. But the decrease of GOM and PBM was quite slight.

Table R1. Speciated Hg emissions in YRD region and concentration at Chongming island in 2014 and 2016

Year	Emission			Concentration		
	GEM (t)	GOM (t)	PBM (t)	GEM (ng m^{-3})	GOM (pg m^{-3})	PBM (pg m^{-3})
2014	34.26	30.41	1.50	2.68	15.41	24.51
2016	27.65	29.16	1.39	1.60	18.97	22.07

Alternately, did you have issues with the speciation unit? It is quite common and I would appreciate an open discussion about that and associated analytical uncertainties. What kind of issues did you encounter? Are you confident that you collected and analyzed GEM (vs. TGM) during the entire experiment? Was the instrumental setup exactly the same during the entire experiment? If not, how can you compare GEM concentrations without discussing analytical uncertainties? See major comment.

Yes, we encountered issues with the speciation unit. The Tekran 2537X was consistent and in good condition during the sampling period. There was no data in January and February in 2016 due to equipment failure. The Tekran 1130/1135 was accidentally rained by rain, so there was no data of speciated mercury between July 2015 and April 2016. From July 2015 to April 2016, we used Tekran 2537X only with PTFE filter to monitor atmospheric mercury. The average concentration of GOM during sampling period (March, 2014 to June 2015, May 2016 to December 2016) was 14.81 ± 13.21 pg m^{-3}, which is approximately 1% of GEM concentration. Thus, the GEM concentrations were approximated to TGM concentrations July 5, 2015 to April 30 2016 when the speciation unit does not work, as most of other studies have done (Slemr et al., 2015; Sprovieri et al., 2016). In addition, we have added discussion about the analytical uncertainties to point out potential impact.

The manuscript was revised as below.

"From July 5, 2015 to April 30 2016, the Tekran 1130/1135 speciation unit was damaged by the rainstorm, the Tekran 2537X were operated without speciation units but with PTFE filter to protect the instrument from particles and sea salt. Therefore, the observed concentrations during July 2015-April 2016 were TGM concentrations indeed. However, the GOM concentrations at Chongming Island accounted for less than 1% of TGM (TGM=GOM+GEM). Thus, the GEM concentrations were approximated to TGM concentrations from July 2015 to April 2016."

See the revised manuscript at line 108 – 114

"In our research, random uncertainties of individual measurement had been averaged out and the systematic uncertainties need to be considered. The overall practically achievable systematic uncertainty would be 10% considering that the instrument was not in ideal performance (Slemr et al., 2015; Steffen et al., 2012). For example, slow deactivation of the traps, contamination of the switching valves and leaks would increase the uncertainties but were difficult to quantify (Slemr et al., 2015;Steffen et al., 2012). Because of the consistency of instrument and the quality assurance/quality control have been paid special attention to during the sampling campaign, the systematic differences of instrument did not affect the huge variation between 2014 and 2016."

See the revised manuscript at line 122 – 129

Comment 7:

Lines 103-104: "The impactor plates and quartz filter were changed in every two weeks. The quartz filter was changed once a month". Did you change the quartz filter every two weeks or once a month?

Response:

Yes, the impactor plates, Teflon filter and quartz filter were changed in every two weeks. The soda lime was changed once a month. We have corrected this sentence in the revised manuscript as below.

"The impactor plates and quartz filter were changed in every two weeks. The soda lime was changed once a month."

See the revised manuscript at line 119 – 120

Comment 8:

Line 106: "During the sampling campaigns, $PM_{2.5}$, O_3, NO_x, CO and SO_2 were monitored". Why aren't you discussing the data, especially SO_2, NO_x, $PM_{2.5}$ while your main conclusion is that Hg decreasing trend in due to air pollution control policies targeting SO_2, NO_x, and $PM_{2.5}$. I agree that you present emissions inventories, but I would really appreciate to see a real interpretation and discussion of these data. Do you also observe a decreasing trend? See major comment.

Response:

To further verify the cause of downward trend of atmospheric Hg, we give the emission inventory (Table S6) and concentrations (Table S5) of other air pollutants in the studied regions in both 2014 and 2016. Both the emissions and concentrations of SO_2, NO_2, and PM showed a decreasing trend, which is used to support that "air pollution control policies targeting SO_2, NO_2, and PM reductions had significant co-benefits on atmospheric Hg".

"Table 3 showed the detailed data of the three classifications. From 2014 to 2016, the whole China region (NCP, SW-YRD) contributed to 70% of GEM decline at Chongming Island. Considering downward trend of emission inventory and atmospheric pollutant from 2014 to 2016 in NCP and SW-YRD region (Table S5, Table S6), the reason of downward trend can be attributed to the effectiveness of existing air pollution control measures in China (SC, 2013; MEP, 2014)."

See the revised manuscript at line 375 – 379

Table S5. The annual concentration of SO_2, NO_x, O_3 and $PM_{2.5}$ at Chongming site, NCP, and SW-YRD regions

Year		2014			2016			Change		
Pollutants	Region	NCP	SW-YRD	Chongming	NCP	SW-YRD	Chongming	NCP	SW-YRD	Chongming
$PM_{2.5}$ ($\mu g\ m^{-3}$)		71.93	53.05	25.09	60.75	44.75	23.89	-16%	-16%	-5%
SO_2		34.52	21.01	1.60	24.37	16.40	1.47	-29%	-22%	-8%

($\mu g\ m^{-3}$)									
NO_2 ($\mu g\ m^{-3}$)	45.07	34.34	12.62	41.55	34.40	10.84	-8%	0%	-14%
O_3 ($\mu g\ m^{-3}$)	60.29	56.27	41.70	61.84	60.92	44.38	3%	8%	6%
GEM ($ng\ m^{-3}$)	No data		2.68	No data		1.60	No data		-40%

Note: According to the contribution of trajectory, the dominant provinces in the NCP region included Beijing, Tianjin, Hebei, Shandong and Liaoning province. The SW-YRD mainly contained Shanghai, Zhejiang, Jiangsu, Jiangxi and Anhui province.

Table S6. Emission inventories of the main pollutants from the studied regions in 2014 and 2016

Air pollutants	2014		2016		Decline proportion	
	NCP	SW-YRD	NCP	SW-YRD	NCP	SW-YRD
$PM_{2.5}$ (kt)	2019	1209	1849	1109	-8%	-8%
NO_x (kt)	5697	4022	5424	3855	-5%	-4%
SO_2 (kt)	3780	1993	3450	1780	-9%	-11%
GEM (t)	118	72	103	67	-13%	-7%

Note: According to the contribution of trajectory, the dominant provinces in the NCP region included Beijing, Tianjin, Hebei, Shandong and Liaoning province. The SW-YRD mainly contained Shanghai, Zhejiang, Jiangsu, Jiangxi and Anhui province.

Comment 9:

Lines 173-175: "Besides, this method required similar meteorological conditions of the periods participated in comparison so as to reduce the interference from meteorology". I am not sure I understand this sentence. Do you mean that you used similar meteorological data in 2014 and 2016 to compute the back-trajectories? Or are you referring to the fact that meteorological conditions were pretty much similar in 2014 and 2016 (lines 266-270)?

Response:

Thanks for the comments. Yes, this sentence is referring to the fact that meteorological conditions were pretty similar in 2014 and 2016. We have revised the sentence as suggested to make it easier to understand.

"Besides, meteorological conditions were pretty similar in 2014 and 2016 so as to reduce the interference from meteorology (Table S2).

See the revised manuscript at line 200 – 201

Comment 10:

Lines 188: "For small emission sectors (: : :)". Which ones?

Response:

We have added the explanation of small emission sectors in the revised manuscript as below.

"The emission sectors included coal-fired power plants, coal-fired industrial boilers, residential coal-combustion, cement clinker production, iron and steel production, mobile oil combustion, and other small emission sectors (eg., zinc smelting, lead smelting, municipal solid incineration, copper smelting, aluminum production, gold production, other coal combustion, stationary oil combustion, and cremation)."

See the revised manuscript at line 208-212.

Comment 11:

Lines 193-194: "The average concentrations of GEM in 2014 and 2016 were (: : :)". What about the mean concentration in 2015? Additionally, are the average annual concentrations actually referring to March-December? If so, please add something like "The average concentrations of GEM in 2014 (Mar-Dec) and 2016 (Mar-Dec) were (: : :)".

Response:

The average concentration of GEM in 2015 was 2.14 ± 0.82 ng m^{-3}. And we have added the concentration of 2015 and remark in the revised manuscript.

"The average concentrations of GEM in 2014 (March to December), 2015 and 2016 (March to December) were 2.68 ± 1.07 ng m^{-3}, 2.14 ± 0.82 ng m^{-3}, and 1.60 ± 0.56 ng m^{-3}, respectively."

See the revised manuscript at line 226 – 227

Comment 12:

Lines 194-195: How does it compare to concentrations reported in Sprovieri et al. (2016)?

Response:

t test was used to compare the GEM concentration at Chongming and background concentration of Northern Hemisphere. The *p* value ($p<0.01$) of the *t* test were added in the revised manuscript as below.

"The GEM concentrations in 2014 (2.68±1.07 ng m^{-3}) were higher (*t* test, $p<0.01$) than the Northern Hemisphere back-ground concentration (about 1.5 ng m^{-3}) (Sprovieri et al., 2010) and those measured in other remote and rural locations in China (Zhang H et al., 2015; Fu et al., 2008a; Fu et al., 2009)."

See the revised manuscript, line 229 – 231

Comment 13:

Lines 199-200: "During this period, monthly GEM concentrations showed a significant decrease with a rate of -0.60 ng/m3/yr". Table 1 refers to TGM concentrations, not GEM. Additionally, as mentioned earlier, the rate is -0.52 ng/m3/yr in Table 1. Please, try to be consistent throughout the manuscript.

Response:

Thanks for the comments. It is a typo. We have gone through the whole paper so as to make the manuscript consistent.

Comment 14:

Lines 201-216: To me, "GEM" and "TGM" are not interchangeable (see previous comment). While the difference between TGM and GEM is usually smaller than 1% (Soerensen et al., 2010), it might not be the case everywhere. What is the fraction of GOM at your site? I would appreciate a discussion on analytical uncertainties and instrumental setups.

Response:

We agree that the GEM and TGM are not always interchangeable. The average concentration of GOM during sampling period was 14.81 ± 13.21 pg m^{-3}, which was less than 1% of TGM. Thus, the GEM concentrations were approximated to TGM concentrations July 5, 2015 to April 30 2016 when the speciation unit does not work, as most of other studies have done (Slemr et al., 2015; Sprovieri et al., 2016). We have pointed out this in the revised manuscript.

A discussion on analytical uncertainties and instrumental setups has been added in the following text as below.

"From July 5, 2015 to April 30 2016, the Tekran 1130/1135 speciation unit was damaged by the rainstorm, the Tekran 2537X were operated without speciation units but with PTFE filter to

protect the instrument from particles and sea salt. Therefore, the observed concentrations during July 2015-April 2016 were TGM concentrations indeed. However, the GOM concentrations at Chongming Island accounted for less than 1% of TGM (TGM=GOM+GEM). Thus, the GEM concentrations were approximated to TGM concentrations from July 2015 to April 2016."

See the revised manuscript at line 108 – 114

"In our research, random uncertainties of individual measurement had been averaged out and the systematic uncertainties need to be considered. The overall practically achievable systematic uncertainty would be 10% considering that the instrument was not in ideal performance (Slemr et al., 2015; Steffen et al., 2012). For example, slow deactivation of the traps, contamination of the switching valves and leaks would increase the uncertainties but were difficult to quantify (Slemr et al., 2015;Steffen et al., 2012). Because of the consistency of instrument and the quality assurance/quality control have been paid special attention to during the sampling campaign, the systematic differences of instrument did not affect the huge variation between 2014 and 2016."

See the revised manuscript at line 124 – 129

The sentence "at the Cape Point of South Africa, GEM concentrations decreased from 1.35 ng/m3 in 1996 to 0.9 ng/m3 in 2008" is not entirely true. A downward trend has been observed from 1996 to 2005, while an upward one is observed since 2007 (Martin et al., 2017; Slemr et al., 2015).

Response:

We have revised our expression about observation trend in Cape Point as follows

"In South Africa, annual average GEM concentration at Cape Point decreased from 1.29 ng m^{-3} in 1996 to 1.19 ng m^{-3} in 2004 (Slemr et al., 2008) and were increasing from 0.93 ng m^{-3} in 2007 (Slemr et al., 2015) until 2016 (Martin et al, 2017)."

Additionally, the instrumental setup changed: a manual amalgamation technique was used from 1995 to 2004 while a Tekran instrument has been used since 2007 (Martin et al., 2017). It might also be the case at other stations in Table 1. How does it influence the various trends reported in Table 1?

Response:

Table 1 was moved to supporting information (Table S4) so as to focus on our topic. In the Table S4, all the stations used Tekran instruments except for the monitoring in South Korea. The instrument of Canadian sites were maintended by the Environment Canada- developed Research Data Management and Quality Assurance System (RDMQ). At Zeppelin, the instruement maintenance were carried out by the protocols of Norwegian mercury monitoring program. Instruments at these plances have been maintended under the guidance of similar quality control criteria. In South Korea, the concentration of TGM were measured using an automatic on-line analytical system called a AM-series analyzer. Although the intruments used in the stations listed in the Table 1 were not totally the same, the instruments at each site remained unchanged during the monitoring period. Therefore, the downward trend at different sites can be compared in the Table S4. We have revised the Table S4 and give expression about the different instrument in the revised manusript.

"All the stations in Table S4 used Tekran instruments except for the observation in South Korea. Different instruments could cause potential differences in the observation, but they were comparable and did not affect the conclusion of comparison in downward trend (Slemr et al., 2015; Sprovieri et al., 2016)."

See the revised manuscript at line 249 - 252

Table S4. Historical variation trends of atmospheric Hg in previous studies

Monitoring site	Duration	TGM trend (pg m^{-3} yr^{-1})	Variation trend	Site description	Monitoring instrument	References
Alert, Canada	2000-2009	-13(-21,0)	-0.9% y^{-1}	Remote	2537A	Cole et al. 2013
Kuujjuarapik, Canada	2000-2009	-33(-50,-18)	-2.1% y^{-1}	Remote	2537A	Cole et al. 2013
Egbert, Canada	2000-2009	-35(-44,-27)	-2.2% y^{-1}	Remote	2537A	Cole et al. 2013
Zeppelin Stn, Norway	2000-2009	+2(-7,+12)	-	Remote	2537A	Cole et al. 2013
St.Anicet, Canada	2000-2009	-29(-31,-27)	-1.9% y^{-1}	Remote	2537A	Cole et al. 2013
Kejimkujik, Canada	2000-2009	-23(-33,-13)	-1.6% y^{-1}	Remote	2537A	Cole et al. 2013

Head, Ireland	1996-2009	-	-1.3±0.2% y^{-1}	Rural	2537A	Weigelt et al. 2015
Yong San, South Korea	2004-2011	No trend (3.54±1.46 ng m^3)		Urban	AM-3	Kim et al. 2016
Yong San South Korea	2013-2014	Decrease to 2.34±0.73 ng m^3			AM-3	Kim et al. 2016
Mt. Changbai	2013-2015	Decrease from 1.74 ng m^{-3} to 1.58 ng m^{-3}		Remote	2537B	Fu et al. 2015
Chongming Island, China	2014-2016	-600	-29.4%/y	Remote	2537X	This study

Comment 15:

Lines 212-214: "The decreasing trend observed in our study was accordant with the unpublished data in Mt. Changbai during 2014-2015 cited in the review of Fu et al. (2015). But much sharper decrease of Hg concentrations was observed in our study". Aren't the data at Mt. Changbai you are referring to in Sprovieri et al. (2016)? What is the trend at that site? Why isn't included in Table 1?

Response:

The data of Mt. Changbai reported by Sprovieri et al. (2016) is the observation in 2013. The observation period is not in the range of our study period. Therefore, we cited the data reported by Fu et al.(2015), where the observation is in 2014-2015.

"The decreasing trend observed in our study was accordant with the data in Mt. Changbai during 2014-2015 cited in the review of Fu et al. (2015). The atmospheric mercury at Chongming was influenced by and in turn reflected regional mercury emission and cycle. Although the decline in atmospheric mercury was observed in many sites of the Northern Hemisphere, much sharper decrease of Hg concentrations was observed in our study."

See the revised manuscript at line 262 - 267

Comment 16:

Line 224: Are you referring to Figure 2?

Response:

Refer to figure 3 in our revised manuscript. We have corrected this in the revised manuscript.

Comment 17:

Lines 225-227: Is that based on the _3 years of data?

Response:

Figure 3 is calculated based on the 3 years data. We have revised it in the manuscript as below.

"According to the decomposition result (Figure 2c), we observed strong seasonal cycle at Chongming. The GEM concentrations were highest in July and lowest in September, so GEM concentrations in the same month from different years were averaged to understand the detrended seasonal circle (Figure 3)."

See the revised manuscript at line 270 - 276

Comment 18:

Line 234: "The higher Hg concentrations in cold seasons in Mt. Ailao and Mt. Waliguan (: : :)". You say above that concentrations are lower in the cold season at these sites. This is confusing.

Response:

Sorry for the mistakes. We have revised the manuscript as below.

"The higher Hg concentrations in cold seasons in Mt. Leigong were mainly explained by coal-combustion for urban and residential heating during cold seasons. Whereas, increasing solar radiation and soil/air temperature dominate the higher Hg concentrations in Mt. Ailao."

See the revised manuscript at line 283- 286

Comment 19:

Line 250-251: "Therefore, we supposed that the seasonal cycle of GEM concentrations was dominated by natural emissions". How can you explain that the seasonal cycle is more pronounced in 2014 than in 2016? See major comment.

Response:

The seasonal variation was more pronounced in 2014 can attribute to the lower wet deposition and GEM oxidation. On one aspect, as a costal site, the Chongming island is

abundant with •OH. The increase of O_3 concentration from the summer of 2014 to 2016 may contribute to a higher oxidation of GEM in 2016. On another aspect, higher wet Hg deposition in summer is approximately 6.6 times of that in the winter at Chongming (Zhang et al., 2010). Meanwhile, the rainfall in 2016 summer (546 mm) was higher than the rainfall in 2014 (426 mm). Therefore, the higher oxidation and wet deposition rate of mercury in the summer of 2016 will reduce the concentration difference between summer and winter, which lead to a less pronounced seasonal variation in 2016.

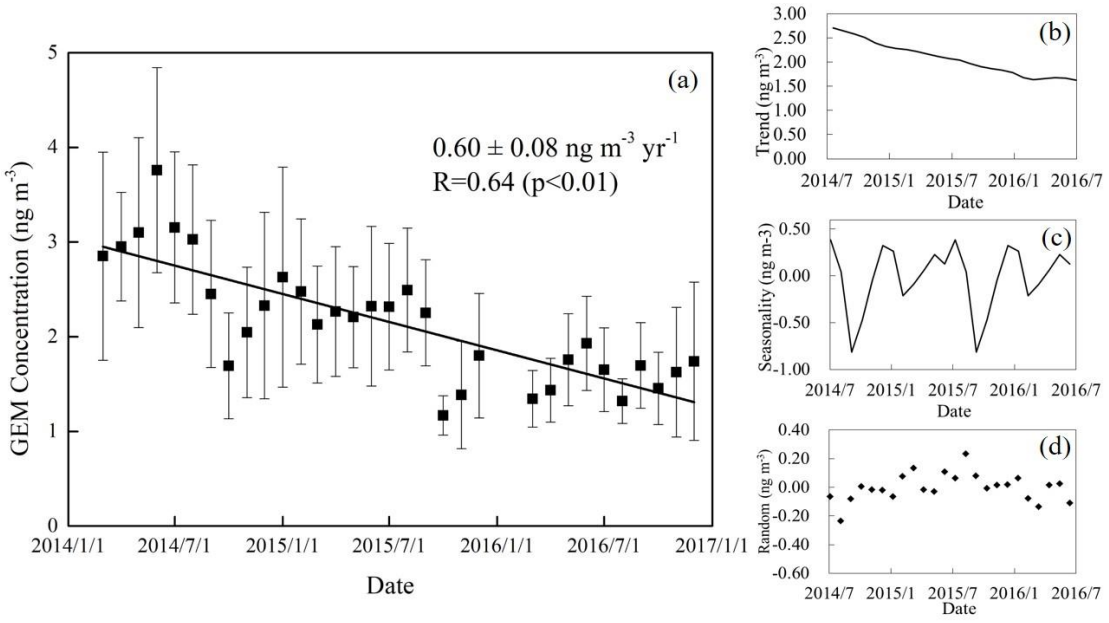


Figure 2. Monthly average GEM concentrations during the studied period (a) observed monthly GEM concentrations (b) GEM trend after decomposition (c) GEM seasonality after decomposition (d) GEM random after decomposition

Note: The observed concentrations during July 2015-April 2016 were TGM concentrations indeed due to the problems of Tekran 1130/1135. However, the GOM concentrations at Chongming island accounted for less than 1% of TGM. Thus, the GEM concentrations were approximated to TGM concentrations during July 2015-April 2016.

"From Figure 2, we also observed more pronounced seasonal variation in 2014, which can be attributed to the lower wet deposition and GEM oxidation. On one aspect, as a costal site, the Chongming Island is abundant with •OH. The increase of O_3 concentration from the summer of 2014 to 2016 may contribute to a higher oxidation of GEM in 2016. On another aspect,

higher wet Hg deposition in summer is approximately 6.6 times of that in the winter at Chongming (Zhang et al., 2010). Meanwhile, the rainfall in 2016 summer (546 mm) was higher than the rainfall in 2014 (426 mm). Therefore, the higher oxidation and wet deposition rate of Hg in the summer of 2016 will reduce the concentration difference between summer and winter, which lead to a less pronounced seasonal variation in 2016. Meanwhile, the higher oxidation and wet deposition in 2016 also contributed to the downward trend of GEM by reducing the seasonality in spring and summer (Figure S3)."

See the revised manuscript at line 313 - 323

Comment 20:

Lines 275-276: "This decline may be contributed by the downward trend of GEM concentrations in north hemisphere". Please, elaborate on this idea. I don't really understand what you mean here.

Response:

Sorry for the obscure expression. The decline of PSCF value means that East China Sea has less contribution to Chongming in 2016. The potential reason of the decline on PSCF value in the East China Sea may be attributed to the reduction of emissions in Japan and Korea. The downward trend in Japan and Korea will lead to clean air mass transport from Japan and Koran to East China Sea and then to Chongming. We have revised the manuscript as below:

"The decline from the East China Sea may be contributed by the downward trend of GEM concentrations in South Korea and Japan (Kim et al., 2016; Kim et al., 2013), where the anthropogenic Hg emissions of Japan and South Korea have been reduced by 13% and 4% during 2010-2015, respectively (UNEP 2013; UNEP 2018). The air mass from Japan and South Korea would pass through the East China Sea to Chongming."

Table R2. Total Hg emission from Japan and South Korea in 2010 and 2015

Country	Mercury emissions (t)		Decline	Reference
	2010	2015		
Japan	17.07	14.86	-13%	UNEP Technical Report (2013)
South Korea	7.32	7.01	-4%	UNEP Technical Report (2018)

See the revised the manuscript at line 331-335

Comment 21:

Lines 315-325: Do you get the same results if you perform this analysis on SO2, NOx, and PM concentrations?

Response:

The SO_2, NO_x and $PM_{2.5}$ concentrations at Chongming island also show downward trend. However, such kind of analysis is not so suitable for SO2, NOx, and $PM_{2.5}$ due to the following reasons. First, the residential time of SO_2, NOx, and $PM_{2.5}$ is 2-4 d, 8-10d, and several days to few weeks, respectively (Pirrone, et al., 1996, Seinfeld, Spyros, 2016). Such residential time is much shorter than that for Hg^0. Second, SO2, NO_x and $PM_{2.5}$ are more reactive in the atmosphere compared with Hg^0 (Pirrone, et al., 1996, Seinfeld, Spyros, 2016).

Comment 22:

Line 318: 34% should be 35% according to Table 4. Additionally, how can you explain this result? Is there a decline in anthropogenic emissions and a GEM decreasing trend in this region (China Sea, Japan, South Korea) as well? Cluster EAST explains 35% of the decline, i.e., 0.35 x 0.52 = 0.182 ng/m3/yr. Is that consistent with trends reported in this region (e.g., Kim et al., 2016)?

Response:

Yes. This is a mistake that 34% should be 35% in the original manuscript.

However, we have changed the definition of cluster according to the suggestion. The NCP region, SW-YRD region, and ABROAD region causes 26%, 44%, and 30% for GEM decline, respectively. The whole China region (NCP, SW-YRD) contributed to 70% of GEM decline at Chongming Island while ABROAD region contributed to 30%. The decline in NCP and SW-YRD indicated effective air pollution control policy in China since 2013. The decline in ABROAD region was originated from GEM decline in South Korea and Japan.

The decline in Chongming was consistent with the decline in anthropogenic emission and a GEM decreasing trend in the ABROAD region. In South Korea, the decline of GEM at Seoul can be calculated as 0.47 ng m^{-3} yr^{-1} from 2011 to 2013 (Kim, et al., 2016, Kim, et al., 2013). In Japan, there is no published data about long term trend since 2010. But the emission

inventory of Japan decreased from 2010 to 2015 (Table R2). Therefore, the decline in ABROAD can be attributed to the decline in South Korea and Japan.

Table R2. Total Hg emission from Japan and South Korea in 2010 and 2015

Country	Mercury emissions (t)		Decline	Reference
	2010	2015		
Japan	17.07	14.86	-13%	UNEP Technical Report (2013)
South Korea	7.32	7.01	-4%	UNEP Technical Report (2018)

We also revised the manuscript as below.

"The decline from the East China Sea may be contributed by the downward trend of GEM concentrations in South Korea and Japan (Kim et al., 2016; Kim et al., 2013), where the anthropogenic Hg emissions of Japan and South Korea have been reduced by 13% and 4% during 2010-2015, respectively (UNEP 2013; UNEP 2018). The air mass from Japan and South Korea would pass through the East China Sea to Chongming."

See the revised manuscript at line 331 -335

Comment 23:

Lines 321-323: "We also noted that the largest decline of Hg concentrations was observed in the cluster SW, which indicated more effective air pollution control in the regions where the air mass of the cluster SW passed". What about the seasonality of GEM concentrations in the various clusters (NW, SW and EAST)? Could a difference in seasonality explain the observed Hg decline?

Response:

The seasonality of GEM concentration in the various clusters was showed in Figure S3. In our revised manuscript, cluster NW, SW and EAST were modified to cluster NCP, SW-YRD and ABROAD. The seasonality of cluster NCP, SW-YRD and ABROAD were similar to the seasonality at Chongming. The GEM concentration of different clusters reached the highest in the summer of 2014. And the seasonality in 2014 for the three clusters was more pronounced than their seasonality in 2016.

The seasonality also explained the observed decline. From 2014 to 2016, all the clusters

declined in all season. In 2014, the seasonality was more pronounced than the seasonality in 2016. It can be attributed to the higher oxidation of GEM and higher wet deposition in 2016. The smaller seasonal variation also had an effect on the decline. We revised our expression in the revised manuscript.

"Therefore, the higher oxidation and wet deposition rate of mercury in the summer of 2016 will reduce the concentration difference between summer and winter, which lead to a less pronounced seasonal variation in 2016. Meanwhile, the higher oxidation and wet deposition in 2016 also contributed to the downward trend by reducing the seasonality of spring and summer (Figure S3)."

See the revised manuscript in line 319 – 323.

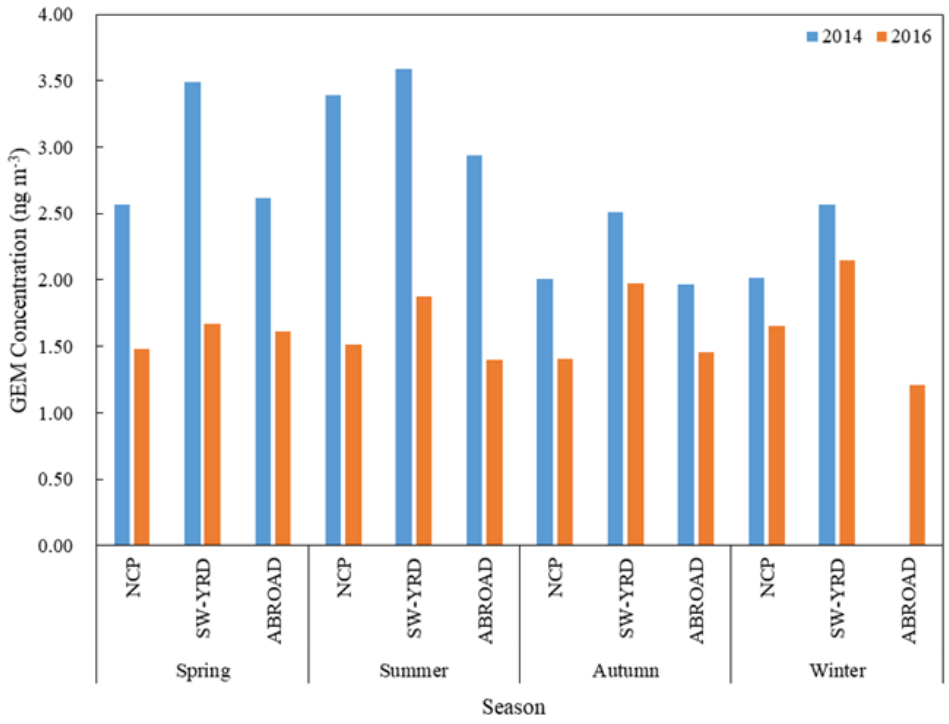


Figure S3. The seasonality of GEM concentration in the NCP, SW-YRD and ABROAD (No trajectory transport though ABROAD in winter of 2014)

See the revised manuscript at Figure S3.

Comment 24:

Figure 3: Could you please add the standard deviations? Is that the average over several years?

Response:

We have revised as suggested. It is the average in the three years. We also give expression in

the manuscript as below.

"According to the decomposition result (Figure 2c), we observed strong seasonal cycle at Chongming. The GEM concentrations were highest in July and lowest in September, so GEM concentrations in the same month from different years were averaged to understand the detrended seasonal circle (Figure 3). The error bars in the Figure 3 meant the standard deviation of the monthly average."

See the revised manuscript in line 270– 274.

References:

Chen, L., Zhang, W., Zhang, Y., Tong, Y., Liu, M., Wang, H., Xie, H., and Wang, X.: Historical and future trends in global source-receptor relationships of mercury, Sci Total Environ, 610-611, 24-31, 10.1016/j.scitotenv.2017.07.182, 2018.

Cheng, Y. F., Zheng, G. J., Wei, C., Mu, Q., Zheng, B., Wang, Z. B., Gao, M., Zhang, Q., He, K. B., Carmichael, G., Poschl, U., and Su, H.: Reactive nitrogen chemistry in aerosol water as a source of sulfate during haze events in China, Science Advances, 2, no. 12, e1601530, 10.1126/sciadv.1601530, 2016.

Fu, X. W., Zhang, H., Yu, B., Wang, X., Lin, C. J., and Feng, X. B.: Observations of atmospheric mercury in China: a critical review, Atmospheric Chemistry and Physics, 15, 9455-9476, 10.5194/acp-15-9455-2015, 2015.

Li, S., Gao, W., Wang, S. X., Zhang, L., Li, Z. J., Wang, L., and Hao, J. M.: Characteristics of Speciated Atmospheric Mercury in Chongming Island, Shanghai, Environmental Science 37, 3290 - 3299, 2016.

Kim, K.-H., Yoon, H.-O., Brown, R. J. C., Jeon, E.-C., Sohn, J.-R., Jung, K., Park, C.-G., and Kim, I.-S.: Simultaneous monitoring of total gaseous mercury at four urban monitoring stations in Seoul, Korea, Atmospheric Research, 132-133, 199-208, 10.1016/j.atmosres.2013.05.023, 2013.

Kim, K.-H., Brown, R. J. C., Kwon, E., Kim, I.-S., and Sohn, J.-R.: Atmospheric mercury at an urban station in Korea across three decades, Atmospheric Environment, 131, 124-132, 10.1016/j.atmosenv.2016.01.051, 2016

Pirrone N, Keeler G J, Nriagu J O. Regional Differences in Worldwide Emissions of Mercury to the Atmosphere. Atmospheric Environment, 1996, 30(30): 2981-2987.

Seinfeld J H, Spyros N P. Atmospheric Chemistry and Physics: From Air Pollution to Clumate Change. John Wiley & Sons, 2016.

Slemr, F., Angot, H., Dommergue, A., Magand, O., Barret, M., Weigelt, A., Ebinghaus, R., Brunke, E. G., Pfaffhuber, K. A., Edwards, G., Howard, D., Powell, J., Keywood, M., and Wang, F.: Comparison of mercury concentrations measured at several sites in the Southern Hemisphere, Atmospheric Chemistry and Physics, 15, 3125-3133, 10.5194/acp-15-3125-2015, 2015.

Sprovieri, F., Pirrone, N., Bencardino, M., amp, apos, Amore, F., Carbone, F., Cinnirella, S., Mannarino, V., Landis, M., Ebinghaus, R., Weigelt, A., Brunke, E.-G., Labuschagne, C., Martin, L., Munthe, J., Wängberg, I., Artaxo, P., Morais, F., Barbosa, H. d. M. J., Brito, J., Cairns, W., Barbante, C., Diéguez, M. d. C., Garcia, P. E., Dommergue, A., Angot, H., Magand, O., Skov, H., Horvat, M., Kotnik, J., Read, K. A., Neves, L. M., Gawlik, B. M., Sena, F., Mashyanov, N., Obolkin, V., Wip, D., Feng, X. B., Zhang, H., Fu, X., Ramachandran, R., Cossa, D., Knoery, J., Marusczak, N., Nerentorp, M., and Norstrom, C.: Atmospheric mercury concentrations observed at ground-based monitoring sites globally distributed in the framework of the GMOS network, Atmospheric Chemistry and Physics, 16, 11915-11935, 10.5194/acp-16-11915-2016, 2016.

Sprovieri, F., Pirrone, N., Bencardino, M., amp, apos, Amore, F., Angot, H., Barbante, C., Brunke, E.-G., Arcega-Cabrera, F., Cairns, W., Comero, S., Diéguez, M. d. C., Dommergue, A., Ebinghaus, R., Feng, X. B., Fu, X., Garcia, P. E., Gawlik, B. M., Hageström, U., Hansson, K., Horvat, M., Kotnik, J., Labuschagne, C., Magand, O., Martin, L., Mashyanov, N., Mkololo, T., Munthe, J., Obolkin, V., Ramirez Islas, M., Sena, F., Somerset, V., Spandow, P., Vardè, M., Walters, C., Wängberg, I., Weigelt, A., Yang, X., and Zhang, H.: Five-year records of mercury wet deposition flux at GMOS sites in the Northern and Southern hemispheres, Atmospheric Chemistry and Physics, 17, 2689-2708, 10.5194/acp-17-2689-2017, 2017.

Sung, J.-H., Roy, D., Oh, J.-S., Back, S.-K., Jang, H.-N., Kim, S.-H., Seo, Y.-C., Kim, J.-H., Lee, C. B., and Han, Y.-J.: Trans-boundary movement of mercury in the Northeast Asian region predicted by CAMQ-Hg from anthropogenic emissions distribution, Atmospheric Research, 203, 197-206, 10.1016/j.atmosres.2017.12.015, 2018.

Wu, Q., Wang, S., Li, G., Liang, S., Lin, C. J., Wang, Y., Cai, S., Liu, K., and Hao, J.: Temporal Trend and Spatial Distribution of Speciated Atmospheric Mercury Emissions in China During 1978-2014, Environ Sci Technol, 50, 13428-13435, 10.1021/acs.est.6b04308, 2016.

Zhang, H., Fu, X. w., Lin, C. J., Shang, L. h., Zhang, Y. p., Feng, X. b., and Lin, C.: Monsoon-facilitated characteristics and transport of atmospheric mercury at a high-altitude background site in southwestern China, Atmospheric Chemistry and Physics, 16, 13131-13148, 10.5194/acp-16-13131-2016, 2016.

Zhang, L., Wang, S. X., Wang, L., Wu, Y., Duan, L., Wu, Q. R., Wang, F. Y., Yang, M., Yang, H., Hao, J. M., and Liu, X.: Updated emission inventories for speciated atmospheric mercury from anthropogenic sources in China, Environ Sci Technol, 49, 3185-3194, 10.1021/es504840m, 2015.

Zhang, L., Wang, L., Wang, S., Dou, H., Li, J., Li, S., and Hao, J.: Characteristics and Sources of Speciated Atmospheric Mercury at a Coastal Site in the East China Sea Region, Aerosol and Air Quality Research, 17, 2913-2923, 10.4209/aaqr.2016.09.0402, 2017.

Recent decrease trend of atmospheric mercury concentrations in East China: the influence of anthropogenic emissions

Yi Tang[1, 2], Shuxiao Wang[1, 2*], Qingru Wu[1, 2*], Kaiyun Liu[1, 2], Long Wang[3], Shu Li[1], Wei Gao[4], Lei Zhang[5], Haotian Zheng[1, 2], Zhijian Li[1], Jiming Hao[1, 2]

[1] State Key Joint Laboratory of Environmental Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing 100084, China

2 State Environmental Protection Key Laboratory of Sources and Control of Air Pollution Complex, Beijing 100084, China

3 School of Environment and Energy, South China University of Technology, Guangzhou, 510006, China

4 Yangtze River Delta Center for Environmental Meteorology Prediction and Warning, Shanghai, 20030, China

5 State Key Laboratory of Pollution Control & Resource Reuse, School of the Environment, Nanjing University, Nanjing, 210023, China

* *Correspondence to:* Shuxiao Wang (shxwang@tsinghua.edu.cn)

Qingru Wu (qrwu@tsinghua.edu.cn)

Abstract

Measurements of gaseous elemental Hg (GEM), other air pollutants including SO_2, NO_X, O_3, $PM_{2.5}$, CO, and meteorological conditions were carried out at Chongming Island in East China from March 1 in 2014 to December 31 in 2016. During the sampling period, GEM concentrations significantly decreased from 2.68±1.07 ng m^{-3} in 2014 (March to December) to 1.60 ±0.56 ng m^{-3} in 2016 (March to December). Monthly mean GEM concentrations showed a significant decrease with a rate of -0.60 ±0.08ng m^{-3} yr^{-1} (R^2=0.64, p<0.01 significance level). Combining the analysis of potential source contribution function (PSCF), principle component analysis (PCA), and emission inventory, we found that Yangtze River Delta (YRD) region was the dominant source region of GEM in Chongming Island and the main source industries included coal-fired power plants, coal-fired industrial boilers, and cement clinker production. We further quantified the effect of emission change on the air Hg concentration variations at Chongming Island through a coupled method of trajectory clusters and air Hg concentrations. It was found that the reduction of domestic emissions was the main driver of GEM decline in Chongming Island, accounting for 70% of the total decline. The results indicated that air pollution control policies targeting SO_2, NO_x and particulate matter reductions had significant co-benefits on GEM.

1 Introduction

Mercury (Hg) is of crucial concern to public health and the global environment for its neurotoxicity, long-distance transport, and bioaccumulation. The atmosphere is an important channel for global Hg transport. Once atmospheric Hg deposits to the aquatic system, it can be transformed into methylmercury (MeHg) which bio-accumulates through the food web and affects the central nervous system of human beings (Mason et al., 1995). Hg is therefore on the priority list of several international agreements and conventions dealing with environmental protection, including the *Minamata Convention on Mercury*.

Atmospheric Hg exists in three operationally defined forms: gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM), and particulate-bound mercury (PBM). And the sum of GEM and GOM is known as total gaseous mercury (TGM). In the atmosphere, Hg mainly presents as GEM, accounting for over 95% of the total in the most observation sites (Fu et al., 2015; Li et al., 2016; Zhang et al., 2017). GEM is stable and with low solubility in the troposphere with a long residence time and can be transported at regional and global scale (Lindberg et al., 2007). GEM can be oxidized through photochemical reaction to GOM, which can be converted to PBM upon adsorption/absorption on aerosol surfaces. GOM is much soluble than GEM, and PBM can be quickly scavenged by both dry and wet deposition. Therefore, the residence time of both GOM and PBM is shorter than that of GEM, generally several days to a few weeks for GOM and 0.5 – 2 year for GEM (Schroeder and Munthe, 1998).

The atmospheric Hg observation results are important evidences to assess the effect of Hg emission control. During the past decades, significant decreases of GEM concentrations in Europe and North America have been observed (Cole et al., 2013; Weigelt et al., 2015). Air Hg concentrations in the northern hemisphere are reported to decline by 30-40% between 1990 and 2010 (Zhang Y et al., 2016). Such a decrease is consistent with the decrease in anthropogenic Hg emissions inventory in Europe and North America (Streets et al., 2011). So far, most of the long-term observations on the ground sites have been carried out in the developed countries. For the developing countries such as China, limited atmospheric Hg observations have been carried out (Fu et al., 2008b; Zhang H et al., 2016; Hong et al., 2016) and there is no official national observing network of atmospheric Hg in mainland China. Therefore, there are few continuous multi-year

observation records of China's air Hg concentrations published (Fu et al., 2015).

China contributes to the largest Hg emissions in the world and will continue to be one significant Hg emitter for global Hg emissions in the coming future (UNEP, 2013, Wu et al., 2016, Chen et al., 2018; Pacyna et al., 2016). Large Hg emissions in China have led to the average air Hg concentrations of 2.86 ± 0.95 ng m^{-3} (in the range of 1.60-5.07 ng m^{-3}) at the remote sites in China (Fu et al., 2015). Such Hg concentration level is approximately 1.3 ng m^{-3} higher than the background concentration of GEM in Northern Hemisphere (Zhang et al., 2016;Sprovieri et al., 2017;Fu et al., 2015). In addition, the large Hg emissions in China will also impact the air Hg concentrations in East Asia and even North America through long-range transport (Sung et al., 2018;Zhang et al., 2017). Meanwhile, China has a great potential for Hg emission reduction during implementation of the *Minamata Convention on Mercury* (Chen et al., 2018). Therefore, long-term atmospheric Hg observations in China are critical to understand the Hg cycling at both regional and global scale. China's Hg emissions had increased from 147 t yr^{-1} in 1978 to around 538 t yr^{-1} in 2010 due to the dramatic economic development (Zhang L et al., 2015; Wu et al., 2016; Hui et al., 2017). Atmospheric Hg monitoring that spanned the longest periods (from 2002 to 2010) in Guiyang, southwestern China witnessed the increase of Hg emissions in China (Fu et al., 2011). However, recently atmospheric Hg emissions in China have been estimated to decrease since 2012 (Wu et al., 2016). This decreasing trend needs to be confirmed by atmospheric Hg observations.

In this study, we measured GEM, other air pollutants (eg., $PM_{2.5}$ and NO_x), and meteorological parameters (eg., temperature and wind speed) at a remote marine site of Chongming Island in East China during 2014-2016. We analyzed annual and seasonal variation of GEM and the potential impact factors. Combining the analysis of potential source contribution function (PSCF), principle component analysis (PCA), and emission inventory, the potential source regions and source industries of atmospheric Hg pollution at the monitoring site are identified. In addition, a coupled trajectories and air Hg concentration method is developed to assess the effect of Hg emission change from different regions on air GEM concentration variation at the monitoring site.

2 Materials and methods

2.1 Site descriptions

The monitoring remote site (31°32′13″N, 121°58′04″E, about 10 m above sea level) locates at the top of weather station in Dongtan Birds National Natural Reserve, Chongming Island, China (Figure 1). As China's third largest island, Chongming Island is located in the east of Yangtze River Delta region with a typical subtropical monsoon climate. It is rainy, hot, with southern and southeastern winds in summer and is dry, cold, and with northwestern wind in winter. The dominant surface types are farmland and wetland. There are no large anthropogenic emission sources in the island and no habitants within 5 km distance from the site. The downtown Shanghai area is 50 km to the southwest of the site.

2.2 Sampling methods and analysis

During the monitoring period, we used Tekran 2537X/1130/1135 instruments to monitor speciated Hg in the atmosphere, which was widely used for air Hg observation in the world. The sampling inlet was 1.5 m above the instrument platform. Continuous 5-minute of GEM was measured by Tekran 2537X Hg vapor analyzer with the detection limit of 0.1 ng m^{-3} at a sampling flow rate of 1.0 L min^{-1} during two campaigns: March 1, 2014 to December 31, 2015 and March 26 to December 31, 2016. From July 5, 2015 to April 30 2016, the Tekran 1130/1135 speciation unit was damaged by the rainstorm, the Tekran 2537X were operated without speciation units but with PTFE filter to protect the instrument from particles and sea salt. Therefore, the observed concentrations during July 2015-April 2016 were TGM concentrations indeed. However, the GOM concentrations at Chongming Island accounted for less than 1% of TGM (TGM=GOM+GEM). Thus, the GEM concentrations were approximated to TGM concentrations from July 2015 to April 2016.

The 2537X analyzer was calibrated automatically every 25 h using the internal Hg permeation source inside the instrument, and the internal permeation source was calibrated every 12 months with manual injection of Hg by a syringe from an external Hg source (module 2505). Two zero and two span calibrations were performed for each calibration of gold trap A and B, respectively. The difference between gold trap A and gold trap B was limited to ±10 %. The impactor plates and quartz filter were changed in every two weeks. The soda lime was changed once a month. The denuders

were recoated once every two weeks following the procedure developed by Landis et al. (2002).

In our research, random uncertainties of individual measurement had been averaged out and the systematic uncertainties need to be considered. The overall practically achievable systematic uncertainty would be 10% considering that the instrument was not in ideal performance (Slemr et al., 2015; Steffen et al., 2012). For example, slow deactivation of the traps, contamination of the switching valves and leaks would increase the uncertainties but were difficult to quantify (Slemr et al., 2015;Steffen et al., 2012). Because of the consistency of instrument and the quality assurance/quality control have been paid special attention to during the sampling campaign, the systematic differences of instrument did not affect the huge variation between 2014 and 2016.

During the sampling campaigns, $PM_{2.5}$, O_3, NO_x, CO and SO_2 were also monitored by Thermo Scientific TEOM 1405D, Model 49i O_3 Analyzer, Model 48i CO Analyzer, Model 42i-TL NOx Analyzer and Model 43i SO_2 Analyzer, respectively. The detection limits of O_3, SO_2, NO_x, CO and $PM_{2.5}$ are 1.0, 0.5, 0.4, 0.04 and 0.1 μg m^{-3}, respectively. The meteorological parameters including air temperature, wind speed, and wind direction are measured by Vantage Pro2 weather station (Davis Instruments). The instruments are tested and calibrated periodically. All data are hourly averaged in this study.

2.3 Sources apportionment of atmospheric Hg pollution

2.3.1 PSCF model

To identify the source areas for pollutants with a relatively long lifetime such as GEM (Xu and Akhtar, 2010), the PSCF values for mean GEM concentrations in grid cells in a study domain are calculated by counting the trajectory segment endpoints that terminate within each cell. The number of endpoints that fall in the *ij*-th cell are designated n_{ij}. The number of endpoints for the same cell having arrival times at the monitoring site corresponding to GEM concentrations higher than a specific criterion is defined to be m_{ij}. The criterion in this study is set as the average Hg concentration during our study period. The PSCF value for the *ij*-th cell is then defined as:

$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}} W_{ij} \quad (1)$$

where W_{ij} is an empirical weight to reduce the effects of grid cells with small n_{ij} values. In this study, W_{ij} is defined as in the following formula, in which *Avg* is the mean n_{ij} of all grid cells with n_{ij} greater than zero:

$$W_{ij} = \begin{cases} 1.0 & n_{ij} > 2 * Avg \\ 0.7 & Avg < n_{ij} \leq 2 * Avg \\ 0.42 & 0.5 * Avg < n_{ij} \leq Avg \\ 0.17 & n_{ij} \leq 0.5 * Avg \end{cases} \quad (2)$$

The PSCF value indicates the probability of a grid cell through which polluted events occurs. More method details can be found in the study of Polissar et al. (Polissar et al., 1999). In this study, the domain that covered the potential contribution source region (105 °–135 °E, 15 °–45 °N) was divided into 22500 grid cells with 0.2 ° ×0.2 °resolution. 72-hour back trajectories were generated hourly from 1 March, 2014 to 31 December, 2015 and from March 26 to December 31 in 2016 by TrajStat, a software including HYSPLIT for trajectory calculation with trajectory statistics modules (Wang et al., 2009). PSCF map was plotted using ArcGIS version 10.1.

2.3.2 Principal component analysis (PCA)

Correlation between Hg and other pollutant concentrations are used to identify source industries. Strong positive loadings (loading>0.40) with SO_2 and $PM_{2.5}$ typically indicate the impact of coal combustion, and strong positive loadings with GEM and CO have often been used as an indicator for regional transport because both pollutants have similar source and stable chemical properties (Lin et al., 2006; Pirrone et al., 1996). In this study, PCA was applied to infer the possible influencing factors of GEM in 2014 and 2016. Prior to analysis, each variable was normalized by dividing its mean, and pollutant concentrations (SO_2, CO, NO_X, $PM_{2.5}$) were averaged to 1-h sampling intervals to match the hourly Hg monitoring during sampling period. The results in 2016 had no CO data due to instrument broken. Statistics analyses were carried out by using SPSS 19.0 software.

2.4 Quantification method of source contribution

To further quantitatively assess the effect of change in emissions from different regions on air concentrations variation at a certain monitoring site, a quantitative estimation method which coupled trajectories with air Hg concentrations was developed. We firstly identified the trajectories by using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model. The gridded meteorological data at a horizontal resolution of 1 °×1 °were obtained from the Global Data Assimilation System (GDAS) (Draxler and Hess, 1998). The starting heights were set to be 500 m above ground level to represent the center height of boundary layer where pollutants are usually well mixed in boundary layer. Secondly, each trajectory was assigned with GEM concentration by matching the arriving time in Chongming site.

Third, the backward trajectories which coupled with Hg concentrations were clustered into groups according to transport patterns by using NOAA HYSPLIT 4.7. Thus, the grouped clusters were applied to identify the Hg source regions. The Hg average concentration of the cluster *j* was then calculated as equation (3). And, the trajectory weighted concentration in the cluster *j* as equation (4). At last, the contribution of reduction at a certain region on Hg concentration at monitoring sites in a certain period can be calculated as equation (5).

$$C_{j,t} = \frac{\sum_{i=1}^{n} C_{i,j,t}}{\sum_{i=1}^{n} N_{i,j,t}} \quad (3)$$

$$TWC_{j,t} = AR \times C_{j,t} \quad (4)$$

where *N* refers to a certain trajectory. *j* refers to a certain cluster. *t* is the studied period, and *n* is the number of trajectory. *m* is the number of cluster. *C* is the GEM concentration, ng m^{-3}. *TWC* refers to the trajectory weighted concentration, ng m^{-3}. In order to reduce the influence of trajectory changes in different region between calculated years, the average ratio (AR) was used here for calculating TWC.

$$CR_j = \frac{TWC_{j,t_2} - TWC_{j,t_1}}{\sum_{j=1}^{m} TWC_{j,t_2} - \sum_{j=1}^{m} TWC_{j,t_1}} \quad (5)$$

where *CR* refers to the contribution of GEM reduction. t_1 and t_2 refers to the two period participating to comparison, namely year 2014 and 2016 in this study, respectively.

This approach is a simple method to quantify the influence of anthropogenic emissions on GEM concentration variation. It should be noted that uncertainties always exist in calculating trajectories, causing uncertainties in all trajectory-based approaches. Trajectory errors vary considerably in different situation. Draxler (1996) suggested uncertainties might be 10% of the travel distance. Besides, meteorological conditions were pretty similar in 2014 and 2016 so as to reduce the interference from meteorology (Table S2).

2.5 Regional atmospheric Hg emissions

Regional anthropogenic GEM emissions by month are calculated by using both the technology-

based emission factor methods and transformed normal distribution function method. Detailed introduction of these two methods and the speciation profile of the emitted Hg for each sector are described in our previous study (Wu et al., 2016). Conventional air pollutant (SO_2, $PM_{2.5}$, and NO_x) emissions were calculated following the study of Zhao et al. (2013). The source regions included in the emission inventory consisted of Shanghai, Jiangsu, Zhejiang, and Anhui provinces according to the PSCF results (See section 3.3). The studied emission sectors included coal-fired power plants, coal-fired industrial boilers, residential coal-combustion, cement clinker production, iron and steel production, mobile oil combustion and other small emission sectors (eg., zinc smelting, lead smelting, municipal solid incineration, copper smelting, aluminum production, gold production, other coal combustion, stationary oil combustion, and cremation). The monthly Hg emissions were mainly distributed according to fuel combustions or products productions by month (Table S1). For small emission sectors, the annual emissions were equally distributed into monthly emissions. The GEM emissions from natural sources E_N are calculated as followings.

$$E_N = \sum_i F_i \times A_i \times t \quad (6)$$

where F_i is a bi-directional Hg flux of canopy i, ng km^{-2} yr^{-1}; A is the studied area, km^{-2}; t is the studied year, yr. The bi-directional Hg flux was obtained from the study of Wang et al. (2016) directly. It should be pointed out that the natural emission is a concept of net emission in this manuscript, which reflected a net effect of two competing processes (Zhang, 2009): total Hg natural emissions and total Hg deposition. The total natural emissions included primary natural release and re-emission of legacy Hg stored in the terrestrial and water surface (Wang et al., 2016). When the value is positive, it means the net effect is Hg emissions to air. Otherwise, Hg deposited.

3 Results and discussions

3.1 Decreasing trends of atmospheric Hg during 2014-2016

The average concentrations of GEM in 2014 (March to December), 2015 and 2016 (March to December) were 2.68 ±1.07 ng m^{-3}, 2.14 ± 0.82 ng m^{-3}, and 1.60 ±0.56 ng m^{-3}, respectively. The GEM concentrations in 2014 were higher (t test, p<0.01) than the Northern Hemisphere background concentration (about 1.5 ng m^{-3}) (Sprovieri et al., 2010) and those measured in other remote and rural locations in China (Zhang H et al., 2015; Fu et al., 2008a; Fu et al., 2009). However, in

2016, the GEM concentrations were similar to the background concentrations in the Northern Hemisphere. During this period, monthly GEM concentrations showed a significant decrease with a rate of -0.60 ± 0.08 ng m^{-3} yr^{-1} (R^2=0.64, p<0.01 significance level, n = 32) (Figure 2a). The amount of valid data for each mouth was shown in Table S3. From another aspect, the trend decomposition of the GEM concentration signal (signal = trend + seasonal + random) from March 2014 to December 2016 were performed in Figure 2 (https://anomaly.io/seasonal-trend-decomposition-in-r/). By using this method, we also observed a pronounced trend (Figure 2b) and the random was limited in the range of -0.24 – 0.24 ng m^{-3} (Figure 2d).

One potential worry is that the calculated trend will be sensitive to seasonal variation and the missing data in January and February of 2016 may impact the downward trend. To evaluate the impact of the missing data, we estimate the Hg concentrations in the missing months based on the data of the same months in 2015 and 2017 (Figure S1). Combining the estimated data, we re-fit the Hg concentrations and downward trend still maintained robust and similar to the downward trend in manuscript (Figure S1). Thus, we assume that the missing data is not very important and will not impact our main conclusion.

Table S4 showed the Hg variation trends in different regions. Significant decreases of GEM concentrations in North hemisphere over the past two decades have been well documented (Weigelt et al., 2015; Cole et al., 2013; Kim et al., 2016). All the stations in Table S4 used Tekran instruments except for the observation in South Korea. Different instruments could cause potential differences in the observation, but they were comparable and did not affect the conclusion of comparison in downward trend (Slemr et al., 2015; Sprovieri et al., 2016). Weigelt et al. (2015) showed that GEM concentrations decreased from 1.75 ng m^{-3} in 1996 to 1.4 ng m^{-3} in 2009 at Mace Head, Europe. Ten-year trends of GEM concentrations at six ground-based sites in the Arctic and Canada also showed a decreasing trend at a rate of 13-35 pg m^{-3} y^{-1} (Cole et al., 2013). In South Korea, the observed GEM concentration also had significant decrease in recent years (Kim et al., 2016). In South Africa, annual average GEM concentration at Cape Point decreased from 1.29 ng m^{-3} in 1996 to 1.19 ng m^{-3} in 2004 (Slemr et al., 2008) and were increasing from 0.93 ng m^{-3} in 2007 (Slemr et al., 2015) until 2016 (Martin et al, 2017). However, limited GEM monitoring sites and relative short-time spans in China restricted the views of long-term trends in atmospheric Hg concentration in this region. A preliminary assessment indicated that atmospheric Hg concentrations

in China kept increasing before 2012 (Fu et al., 2015). The decreasing trend observed in our study was accordant with reported data in Mt. Changbai during 2014-2015 cited in the review of Fu et al. (2015). The atmospheric Hg at Chongming was influenced by and in turn reflected regional Hg emission and cycle. Although the decline in atmospheric Hg was observed in many sites of the Northern Hemisphere, much sharper decrease of Hg concentrations was observed at Chongming in our study. The specific reasons for the Hg concentration decrease in our study will be discussed in section 3.4.

3.2 Seasonal variation of GEM concentrations

According to the decomposition result (Figure 2c), we observed strong seasonal cycle at Chongming. The GEM concentrations were highest in July and lowest in September, so GEM concentrations in the same month from different years were averaged to understand the detrended seasonal circle (Figure 3). The error bars in the Figure 3 meant the standard deviation of the monthly average. Observed GEM concentrations showed an obvious seasonal cycle. The mean GEM concentration in warm season (from April to September) is 0.29 ng m^{-3} higher than that in cold season. Such seasonal variation trend is also observed at Nanjing, Miyun, Mt. Ailao, Mt. Waliguan, and Shangri-La (Zhang et al., 2013; Zhang et al., 2016; Fu et al., 2015; Zhu et al., 2012). On the other hand, the means of GEM at Mt. Gongga, Mt. Daimei, Mt.Leigong, and Mt. Changbai in China are relatively higher in cold seasons. The average of atmospheric Hg concentrations in the north hemisphere also have a trough value in summer (Sprovieri et al., 2016).

Seasonal variations of GEM concentration are generally attributed to the following factors, including natural and anthropogenic emissions, atmospheric chemical reaction, and air mass transportation. The higher Hg concentrations in cold seasons in Mt. Leigong were mainly explained by coal-combustion for urban and residential heating during cold seasons. Whereas, increasing solar radiation and soil/air temperature dominate the higher Hg concentrations in Mt. Ailao. In addition, sites in southern, eastern, and northeastern China also impacted from anthropogenic emissions of GEM from the north and west by the northerly winter monsoon while the sites located in western, southwestern, and northern China were impacted in the warm season (Fu et al., 2015). As to most sites in the northern hemisphere, high wet Hg precipitation induced probably by faster GEM oxidation led to lower Hg concentrations in summer.

Source emission is one significant factor on GEM concentrations in the air. The GEM

concentrations at a remote site are generally regarded under the impact of regional emissions. Therefore, the emissions in the YRD regions (Anhui, Zhejiang, Jiangsu, and Shanghai) were calculated. However, the anthropogenic emissions were in the range of 2.5-2.7 t, which is almost unchanged. Compared to the anthropogenic emissions, we observed almost synchronized trends between natural emissions and air Hg concentrations in Figure 4. The natural emissions showed a huge seasonal variation, from -5.4 t to 8.4 t. The largest natural emissions were observed in summer when the highest GEM concentrations were monitored. In the autumn, the natural emissions performed as the largest deposition direction amount and the GEM concentrations were the lowest in the whole year. Therefore, natural emissions instead of anthropogenic were supposed to be one significant factor of the seasonal cycle of GEM concentrations (Figure 4). The seasonal trend of natural emissions is closely related with the canopy types in YRD areas, where widely subtropical forests, paddy field, and dry farming were observed (Figure S2). The high temperature will speed up decomposition of organic compound in soil, which leads to Hg emissions from farmland and forest in YRD region (Luo et al., 2016; Yu et al., 2017). In autumn and winter, with the decrease of temperature (Table S2), the role of soil changed from Hg source to sink, which reduces the Hg concentrations in the air (Wang et al., 2016). At the same time, the growing vegetation in autumn also absorbs air Hg, resulting lower Hg concentrations compared to that in winter. Transport also overall enhanced the observed seasonal variation of GEM concentrations at Chongming Island. According to the statistics of backward trajectories in section 3.4, the GEM concentrations in the air mass which did not pass via the YRD regions also showed high GEM concentration in warm season in 2014 (Figure S3).

From Figure 2, we also observed more pronounced seasonal variation in 2014, which can be attributed to the lower wet deposition and GEM oxidation. On one aspect, as a costal site, the Chongming Island is abundant with •OH. The increase of O_3 concentration from the summer of 2014 to 2016 may contribute to a higher oxidation of GEM in 2016. On another aspect, higher wet Hg deposition in summer is approximately 6.6 times of that in the winter at Chongming (Zhang et al., 2010). Meanwhile, the rainfall in 2016 summer (546 mm) was higher than the rainfall in 2014 (426 mm). Therefore, the higher oxidation and wet deposition rate of Hg in the summer of 2016 will reduce the concentration difference between summer and winter, which lead to a less pronounced seasonal variation in 2016. Meanwhile, the higher oxidation and wet deposition in

2016 also contributed to the downward trend of GEM by reducing the seasonality in spring and summer (Figure S3).

3.3 Source apportionment of atmospheric Hg pollutions

According to the PSCF result, YRD region, including Shanghai, Jiangsu, Anhui, and Zhejiang provinces, was the dominant source region in both 2014 and 2016 (Figure 5). Therefore, Hg emissions from these areas would contribute to high proportion of Hg pollution at Chongming Island. The offshore area mainly around Jiangsu province also has a high PSCF value because some trajectories from North China, especially Shandong province, transport to Chongming Island through this area. Compared to the result in 2014, the PSCF value had an obvious decline in East China Sea in 2016. The decline from the East China Sea may be contributed by the downward trend of GEM concentrations in South Korea and Japan (Kim et al., 2016; Kim et al., 2013), where the anthropogenic Hg emissions of Japan and South Korea have been reduced by 13% and 4% during 2010-2015, respectively (UNEP 2013; UNEP 2018). The air mass from Japan and South Korea would pass through the East China Sea to Chongming.

PCA method was applied to preliminarily identify the source industries. In the studied period, totally 2 factors were identified in 2014 and 2016, respectively. The factor 1 had strong factor loadings of GEM, SO_2, NO_x, CO, and $PM_{2.5}$ in both 2014 and 2016 (No CO data in 2016 due to equipment problems). The factor 1 accounted for 49% variance in 2014 and 50% variance in 2016 (Table 1). The results indicated common significant source sectors of the above five air pollutants, which can also be proven from emission inventories (Table 2). The dominant source industries included coal-fired power plants, coal-fired industrial boilers, and cement clinker production. The PCA results showed that anthropogenic emissions were the main sources of GEM during the sampling period.

The factor 2 in both 2014 and 2016 had a strong positive loading on O_3 and negative loading on NO_x. Considering the low loading of CO and high loading of O_3, the factor 2 can be viewed as a sign of the transport of air mass from stratosphere (Fishman and Seiler, 1983; Jaffe, 2010). The air mass from stratosphere will increase the O_3 concentration. O_3 react with NO, which makes a negative correlation with NO. However, the low loading on GEM of factor 2 indicated that Factor 2 had no relationship with GEM concentrations at Chongming from the aspect of whole year data.

3.4 The influence of anthropogenic emissions

To further understand the reason of the downward trend, we firstly compared the meteorological conditions in both 2014 and 2016. We noted that the difference of annual temperature, solar radiation, and relative humidity were constrained in the range of 17.13±7.48 ℃, 165.55±45.87 W m^{-2} and 75.38±5.82%, respectively (Table S2). The coefficient of variation for annual mean of these meteorological conditions in 2014 and 2016 was 2.6%, 6.7% and 0.2%, respectively. In addition, the wind rose was similar, and the dominating wind was from SE in both 2014 and 2016 (Figure S4). The HYSPLIT results also provided similar trajectories in 2014 and 2016 (Figure 6). Therefore, we assumed that the meteorological condition was not the dominant reason of GEM decline at Chongming site.

To further quantify the driver of GEM decline, a trajectory-based analysis method was used in this study. The 72-h air mass back trajectories were calculated using HYSPLIT for every 8 hours starting at the observation site. Approximately 918 and 832 trajectories were calculated in sampling period in 2014 (Mar 1 to Dec 31, 2014) and 2016 (Mar 26 to Dec 31, 2016), respectively. The trajectories were grouped into 3 clusters in each year according to geographical regions (Figure 6). The first cluster of trajectories mainly passed through the regions (eg., North China) north and northwest to Chongming Island before arriving to our monitoring site, which was denoted as cluster NCP. The second cluster mainly passed YRD region to Chongming, which was signed as cluster SW-YRD. The third type mainly originated from the East China Seas, South Korea, Japan and Northeast Asia continent, and then arrived to our monitoring sites directly without passing the mainland China. This type of trajectories was named as cluster ABROAD. Some trajectories originated from the East China Sea and crossed the mainland China before arriving Chongming were grouped into cluster NCP or SW-YRD depending on the regions it crossed. The trajectories for each of the three clusters in 2014 and 2016 were shown in Table 3.

Table 3 showed the detail statistics data of the three classifications. From 2014 to 2016, the whole China region (NCP, SW-YRD) contributed to 70% of GEM decline at Chongming Island. Considering downward trend of emission inventory and atmospheric pollutant from 2014 to 2016 in NCP and SW-YRD region (Table S5, Table S6), the reason of downward trend can be attributed to the effectiveness of existing air pollution control measures in China (SC, 2013; MEP, 2014). Meanwhile, the cluster NCP, cluster SW-YRD, and cluster ABROAD caused 26%, 44%, and 30%

for GEM decline, respectively (Table 3). The cluster SW-YRD contributed to 44% of reduction, suggesting that air pollution controls on anthropogenic emissions in YRD region dominated the recent decrease of GEM concentrations at Chongming site. The largest decline of Hg concentration (1.32 ng m^{-3}) was also observed in the cluster SW-YRD demonstrated the efficiency of emission reduction in YRD region (Table S5, Table S6). Moreover, ABROAD region caused 30% of GEM decline from 2014 to 2016, which implies global effort on atmospheric Hg emission control under the guidance of *Minamata Convention on Mercury*.

4 Conclusion

Atmospheric Hg was continuously measured for three years at a regional background site in the YRD region. During the sampling period, a downward trend for GEM concentrations (-0.60 ± 0.08 ng m^{-3} y^{-1}) at Chongming Island was observed. The seasonal GEM cycle was dominated by the natural emissions while the annual GEM concentration trend was mainly impacted by anthropogenic emissions. By using a new approach that considers both cluster frequency and the Hg concentration associated with each cluster, we quantified that atmospheric Hg from NCP region, SW-YRD region, and ABROAD region have caused 26%, 44%, and 30% decline of GEM concentrations at Chongming monitoring site, respectively. The result suggested that reduction of anthropogenic emissions in mainland China was the main cause of the recent decreasing trend of GEM concentration at Chongming site. The air pollution control policies in China, especially the pollution control in the coal-fired power plants, coal-fired industrial boilers, and cement clinker production in YRD region and Shandong province, have received significant co-benefit of atmospheric Hg emission reductions. On the other hand, emission reduction from the ABROAD region, where clusters arrived to Chongming monitoring site directly without passing the mainland China, implies global effort on atmospheric Hg emission control under the guidance of *Minamata Convention on Mercury*. Considering that the *Minamata Convention on Mercury* had come into force in 2017, continuous long-term observation of atmospheric Hg in China will be required for the assessment of policy effectiveness.

Data availability. All data are available from the authors upon request.

Competing interests. The authors declare that they have no conflict of interest.

Acknowledge. This work is sponsored by the Natural Science Foundation of China (No. 21607090), Major State Basic Research Development Program of China (973 Program) (No. 2013CB430000), National Key R&D Program of China (No. 2016YFC0201900)

References

Cole, A. S., Steffen, A., Pfaffhuber, K. A., Berg, T., Pilote, M., Poissant, L., Tordon, R., and Hung, H.: Ten-year trends of atmospheric mercury in the high Arctic compared to Canadian sub-Arctic and mid-latitude sites, Atmospheric Chemistry and Physics, 13, 1535-1545, 2013.

Chen, L., Zhang, W., Zhang, Y., Tong, Y., Liu, M., Wang, H., Xie, H., and Wang, X.: Historical and future trends in global source-receptor relationships of mercury, Science of the Total Environment, 610-611, 24-31, 2018.

Draxler, R. R.: Trajectory Optimization for Balloon Flight Planning, International Journal for Numerical Methods in Fluids, 5, 13-23, 1996.

Draxler, R. R., and Hess, G. D.: An overview of the hysplit-4 modeling system for trajectories, Australian Meteorological Magazine, 47, 295-308, 1998.

Fishman J, Seiler W. Correlative Nature of Ozone and Carbon Monoxide in the Troposphere: Implications for the Tropospheric Ozone Budget. Journal of Geophysical Research, 88(C6), 1983.

Fu, X. W., Feng, X. B, Zhu, W. Z., Wang, S. F., and Lu, J. L.: Total gaseous mercury concentrations in ambient air in the eastern slope of Mt. Gongga, South-Eastern fringe of the Tibetan plateau, China, Atmospheric Environment, 42, 970-979, 2008a.

Fu, X. W., Feng, X. B., Zhu, W. Z., Zheng, W., Wang, S. F., and Lu, J. Y.: Total particulate and reactive gaseous mercury in ambient air on the eastern slope of the Mt. Gongga area, China, Applied Geochemistry, 23, 408-418, 2008b.

Fu, X. W., Feng, X. B., Wang, S., Rothenberg, S., Shang, L., Li, Z., and Qiu, G.: Temporal and spatial distributions of total gaseous mercury concentrations in ambient air in a mountainous area in southwestern China: implications for industrial and domestic mercury emissions in remote areas in China, Science of the Total Environment, 407, 2306-2314, 2009.

Fu, X. W., Feng, X. B., Qiu, G. L., Shang, L. H., and Zhang, H.: Speciated atmospheric mercury and its potential source in Guiyang, China, Atmospheric Environment, 45, 4205-4212, 2011.

Fu, X. W., Zhang, H., Yu, B., Wang, X., Lin, C. J., and Feng, X. B.: Observations of atmospheric mercury in China: a critical review, Atmospheric Chemistry and Physics, 15, 9455-9476, 2015.

Hong, Q. Q., Xie, Z. Q., Liu, C., Wang, F. Y., Xie, P. H., Kang, H., Xu, J., Wang, J. C., Wu, F. C., He, P. Z., Mou, F. S., Fan, S. D., Dong, Y. S., Zhan, H. C., Yu, X. W., Chi, X. Y., and Liu, J. G.:

Speciated atmospheric mercury on haze and non-haze days in an inland city in China, Atmospheric Chemistry and Physics, 16, 13807-13821, 2016.

Hui, M. L., Wu, Q. R., Wang, S. X., Liang, S., Zhang, L., Wang, F. Y., Lenzen, M., Wang, Y. F., Xu, L. X., Lin, Z. T., Yang, H., Lin, Y., Larssen, T., Xu, M., and Hao, J. M.: Mercury flows in China and global drivers, Environmental Science & Technology, 51, 222-231, 2017.

Jaffe, D.: Relationship betwen surface and free tropospheric ozone in the western U.S., Environmental Science & Technology, 45, 432-438, 2010.

Kim, K.-H., Yoon, H.-O., Brown, R. J. C., Jeon, E.-C., Sohn, J.-R., Jung, K., Park, C.-G., and Kim, I.-S.: Simultaneous monitoring of total gaseous mercury at four urban monitoring stations in Seoul, Korea, Atmospheric Research, 132-133, 199-208, 2013.

Kim, K. H., Brown, R. J. C., Kwon, E., Kim, I. S., and Sohn, J. R.: Atmospheric mercury at an urban station in Korea across three decades, Atmospheric Environment, 131, 124-132, 2016.

Landis, M. S., Stevens, R. K., Schaedlich, F., and Prestbo, E. M.: Development and characterization of an annular denuder methodology for the measurement of divalent inorganic reactive gaseous mercury in ambient air, Environmental Science & Technology, 36, 3000-3009, 2002.

Lindberg, S., Bullock, R., Ebinghaus, R., Engstrom, D., Feng, X. B., Fitzgerald, W., Pirrone, N., Prestbo, E., and Seigneur, C.: A synthesis of progress and uncertainties in attributing the sources of mercury in deposition, Ambio, 36, 19, 2007.

Li, S., Gao, W., Wang, S. X., Zhang, L., Li, Z. J., Wang, L., and Hao, J. M.: Characteristics of Speciated Atmospheric Mercury in Chongming Island, Shanghai, Environmental Science 37, 3290 - 3299, 2016.

Luo, Y., Duan, L., Driscoll, C. T., Xu, G., Shao, M., Taylor, M., Wang, S. X, and Hao, J. M.: Foliage/atmosphere exchange of mercury in a subtropical coniferous forest in south China, Journal of Geophysical Research Biogeosciences, 121, 2016.

Martin, L. G., Labuschagne, C., Brunke, E. G., Weigelt, A., Ebinghaus, R., and Slemr, F.: Trend of atmospheric mercury concentrations at Cape Point for 1995–2004 and since 2007, Atmospheric Chemistry and Physics, 17, 2393-2399, 2017.

Mason, R. P., Reinfelder, J. R., and Morel, F. M. M.: Bioaccumulation of mercury and methylmercury, Springer Netherlands, 915-921 pp., 1995.

Ministry of Environmental Protection (MEP) and State Administration for Quality Supervision and

Inspection and Quarantine (AQSIQ): Emission standard of air pollutants for boilers, MEP, Beijing, China, 2014.

Pirrone, N., Keeler, G. J., and Nriagu, J. O.: Regional differences in worldwide emissions of mercury to the atmosphere, Atmospheric Environment, 30, 2981-2987, 1996.

Polissar, A. V., Hopke, P. K., Paatero, P., Kaufmann, Y. J., Hall, D. K., Bodhaine, B. A., Dutton, E. G., and Harris, J. M.: The aerosol at Barrow, Alaska: long-term trends and source locations, Atmospheric Environment, 33, 2441-2458, 1999.

State Council of the People's Republic of China (SC): Action plan of national air pollution prevention and control, SC, Beijing, China, 2013.

Schroeder, W. H., and Munthe, J.: Atmospheric mercury—An overview, Atmospheric Environment, 32, 809-822, 1998.

Slemr, F., Angot, H., Dommergue, A., Magand, O., Barret, M., Weigelt, A., Ebinghaus, R., Brunke, E. G., Pfaffhuber, K. A., Edwards, G., Howard, D., Powell, J., Keywood, M., and Wang, F.: Comparison of mercury concentrations measured at several sites in the Southern Hemisphere, Atmospheric Chemistry and Physics, 15, 3125-3133, 2015.

Sprovieri, F., Pirrone, N., Ebinghaus, R., Kock, H., and Dommergue, A.: A review of worldwide atmospheric mercury measurements, Atmospheric Chemistry and Physics, 10, 8245-8265, 2010.

Streets, D. G., Devane, M. K., Lu, Z., Bond, T. C., Sunderland, E. M., and Jacob, D. J.: All-Time releases of mercury to the atmosphere from human activities, Environmental Science & Technology, 45, 10485-10491, 2011.

Sprovieri, F., Pirrone, N., Bencardino, M., amp, apos, Amore, F., Carbone, F., Cinnirella, S., Mannarino, V., Landis, M., Ebinghaus, R., Weigelt, A., Brunke, E.-G., Labuschagne, C., Martin, L., Munthe, J., Wängberg, I., Artaxo, P., Morais, F., Barbosa, H. d. M. J., Brito, J., Cairns, W., Barbante, C., Diéguez, M. d. C., Garcia, P. E., Dommergue, A., Angot, H., Magand, O., Skov, H., Horvat, M., Kotnik, J., Read, K. A., Neves, L. M., Gawlik, B. M., Sena, F., Mashyanov, N., Obolkin, V., Wip, D., Feng, X. B., Zhang, H., Fu, X., Ramachandran, R., Cossa, D., Knoery, J., Marusczak, N., Nerentorp, M., and Norstrom, C.: Atmospheric mercury concentrations observed at ground-based monitoring sites globally distributed in the framework of the GMOS network, Atmospheric Chemistry and Physics, 16, 11915-11935, 2016.

Sprovieri, F., Pirrone, N., Bencardino, M., amp, apos, Amore, F., Angot, H., Barbante, C., Brunke,

E.-G., Arcega-Cabrera, F., Cairns, W., Comero, S., Diéguez, M. d. C., Dommergue, A., Ebinghaus, R., Feng, X. B., Fu, X., Garcia, P. E., Gawlik, B. M., Hageström, U., Hansson, K., Horvat, M., Kotnik, J., Labuschagne, C., Magand, O., Martin, L., Mashyanov, N., Mkololo, T., Munthe, J., Obolkin, V., Ramirez Islas, M., Sena, F., Somerset, V., Spandow, P., Vardè, M., Walters, C., Wängberg, I., Weigelt, A., Yang, X., and Zhang, H.: Five-year records of mercury wet deposition flux at GMOS sites in the Northern and Southern hemispheres, Atmospheric Chemistry and Physics, 17, 2689-2708, 2017

Steffen, A., Scherz, T., Olson, M., Gay, D., and Blanchard, P.: A comparison of data quality control protocols for atmospheric mercury speciation measurements, J Environ Monit, 14, 752-765, 2012.

Sung, J.-H., Roy, D., Oh, J.-S., Back, S.-K., Jang, H.-N., Kim, S.-H., Seo, Y.-C., Kim, J.-H., Lee, C. B., and Han, Y.-J.: Trans-boundary movement of mercury in the Northeast Asian region predicted by CAMQ-Hg from anthropogenic emissions distribution, Atmospheric Research, 203, 197-206, 2018.

Arctic Monitoring and Assessment Programme and United Nations Environment Programme (AMAP/UNEP): Global Hg assessment 2013: sources, emissions, releases and environmental transport, AMAP/UNEP, Geneva, Switzerland, 2013

Arctic Monitoring and Assessment Programme and United Nations Environment Programme (AMAP/UNEP): Global mercury assessment 2018 - draft technical background document, AMAP/UNEP, Geneva, Switzerland, 2018.

Wang, X., Lin, C.-J., Yuan, W., Sommar, J., Zhu, W., and Feng, X.: Emission-dominated gas exchange of elemental mercury vapor over natural surfaces in China, Atmospheric Chemistry and Physics, 16, 11125-11143, 2016.

Wang, Y. Q., Zhang, X. Y., and Draxler, R. R.: TrajStat: GIS-based software that uses various trajectory statistical analysis methods to identify potential sources from long-term air pollution measurement data, Elsevier Science Publishers B. V., 938-939 pp., 2009.

Weigelt, A., Ebinghaus, R., Manning, A. J., Derwent, R. G., Simmonds, P. G., Spain, T. G., Jennings, S. G., and Slemr, F.: Analysis and interpretation of 18 years of mercury observations since 1996 at Mace Head, Ireland, Atmospheric Environment, 100, 85-93, 2015.

Wu, Q., Wang, S., Li, G., Liang, S., Lin, C. J., Wang, Y., Cai, S., Liu, K., and Hao, J.: Temporal trend and spatial distribution of speciated atmospheric mercury emissions in China during 1978-

2014, Environmental Science & Technology, 50, 13428-13435, 2016.

Xu, X., and Akhtar, U. S.: Identification of potential regional sources of atmospheric total gaseous mercury in Windsor, Ontario, Canada using hybrid receptor modeling, Atmospheric Chemistry and Physics, 10, 7073-7083, 2010.

Yu Q, Luo Y, Wang S, Wang Z, Hao J, Duan L. Gaseous Elemental Mercury (GEM) Fluxes over Canopy of Two Typical Subtropical Forests in South China. Atmospheric Chemistry and Physics, 18(1), 495-509, 2018.

Zhang, G. Y., Zhou, L. M., Zheng, X. M., and Huang, W. D.: Temporal distribution and potential hazards of wet depostion mercury in Yangtze River Estuary, Urban Environmental & Urban Ecology, 1-4, 2010.

Zhang, H., Fu, X. W., Lin, C. J., Wang, X., and Feng, X. B.: Observation and analysis of speciated atmospheric mercury in Shangri-La, Tibetan Plateau, China, Atmospheric Chemistry and Physics, 15, 653-665, 2015.

Zhang, H., Fu, X. W., Lin, C.-J., Shang, L. H., Zhang, Y. P., Feng, X. B., and Lin, C.: Monsoon-facilitated characteristics and transport of atmospheric mercury at a high-altitude background site in southwestern China, Atmospheric Chemistry and Physics, 16, 13131-13148, 2016.

Zhang, L., Wang, S. X., Wang, L., Wu, Y., Duan, L., Wu, Q. R., Wang, F. Y., Yang, M., Yang, H., Hao, J. M., and Liu, X.: Updated emission inventories for speciated atmospheric mercury from anthropogenic sources in China, Environmental Science & Technology, 49, 3185-3194, 2015.

Zhang, L. M., Wright, L. P., and Blanchard, P.: A review of current knowledge concerning dry deposition of atmospheric mercury, Atmospheric Environment, 43, 5853-5864, 2009.

Zhang, Y. X., Jacob, D. J., Horowitz, H. M., Chen, L., Amos, H. M., Krabbenhoft, D. P., Slemr, F., St Louis, V. L., and Sunderland, E. M.: Observed decrease in atmospheric mercury explained by global decline in anthropogenic emissions, Proceedings of the National Academy of Sciences of the United States of America, 113, 526, 2016.

Zhao, B., Wang, S. X., Liu, H., Xu, J. Y., Fu, K., Klimont, Z., Hao, J. M., He, K. B., Cofala, J., and Amann, M.: NOx emissions in China: historical trends and future perspectives, Atmospheric Chemistry and Physics, 13, 9869-9897, 2013.

Zhu, J., Wang, T., Talbot, R., Mao, H., Hall, C. B., Yang, X., Fu, C., Zhuang, B., Li, S., Han, Y., and Huang, X.: Characteristics of atmospheric total gaseous mercury (TGM) observed in urban Nanjing,

China, Atmospheric Chemistry and Physics, 12, 12103-12118, 2012.

Figure citation

Figure 1. The location of the Chongming monitoring site in Shanghai, China

Figure 2. Monthly average GEM concentrations during the studied period (a) observed monthly GEM concentrations (b) GEM trend after decomposition (c) GEM seasonality after decomposition (d) GEM random after decomposition

Note: The observed concentrations during July 2015-April 2016 were TGM concentrations indeed due to the problems of Tekran 1130/1135. However, the GOM concentrations at Chongming Island accounted for less than 1% of TGM. Thus, the GEM concentrations were approximated to TGM concentrations during July 2015-April 2016.

Figure 3. Monthly variations of GEM concentration at remote sites in China

Figure 4. Seasonal cycle of GEM concentrations and emissions during 2014-2016. The error bars represent the standard deviation of seasonal average. Negative values of natural emissions represent mercury deposition and positive values of natural emissions represent natural emissions.

Figure 5. Source regions of GEM at monitoring site from PSCF model in 2014(a) and 2016(b)

Figure 6. The back trajectories map for cluster NCP, SW-YRD and ABROAD in 2014(a) and 2016(b)

(NCP – North China Plain; SW-YRD –Southwest region and Yangtze River Delta; ABROAD – Abroad)

Table citation

Table 1. PCA component loading of GEM and the co-pollutant**s**

Table 2. Main air pollutant emitted by the different sector in YRD region in 2014

Table 3. The statistics of cluster and estimated contribution of GEM reduction in 2014 and 2016

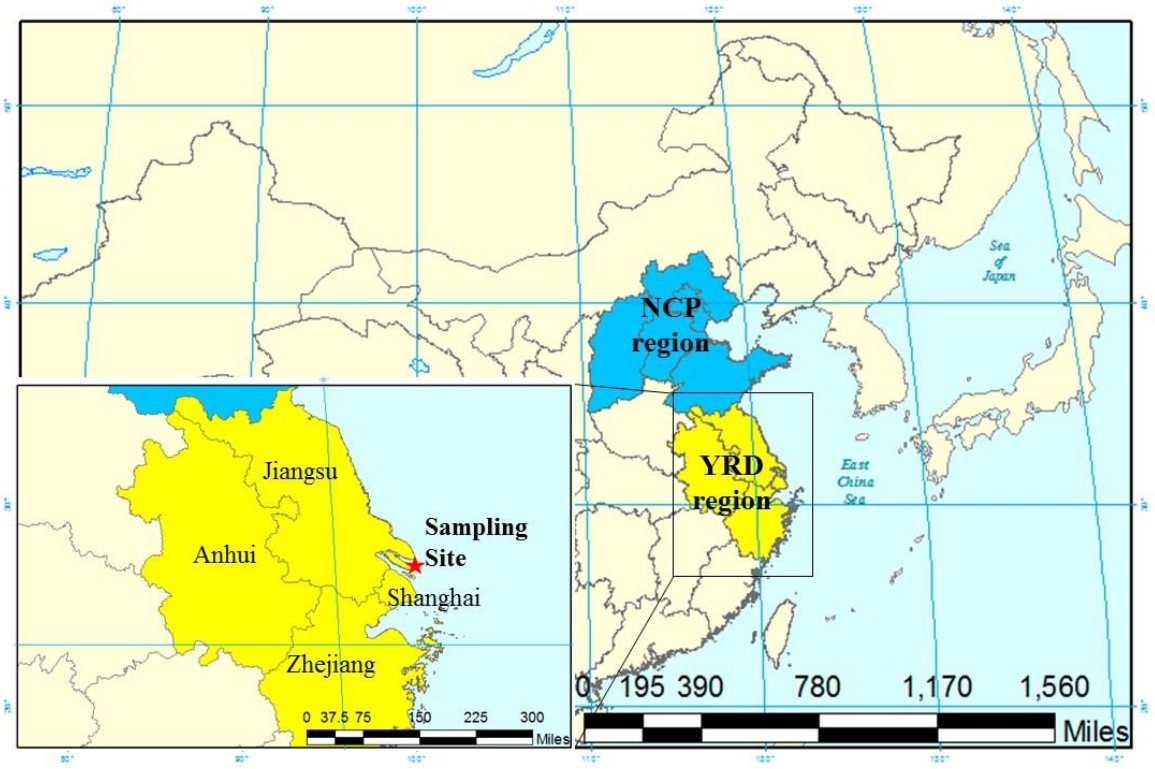


Figure 1. The location of the Chongming monitoring site in Shanghai, China

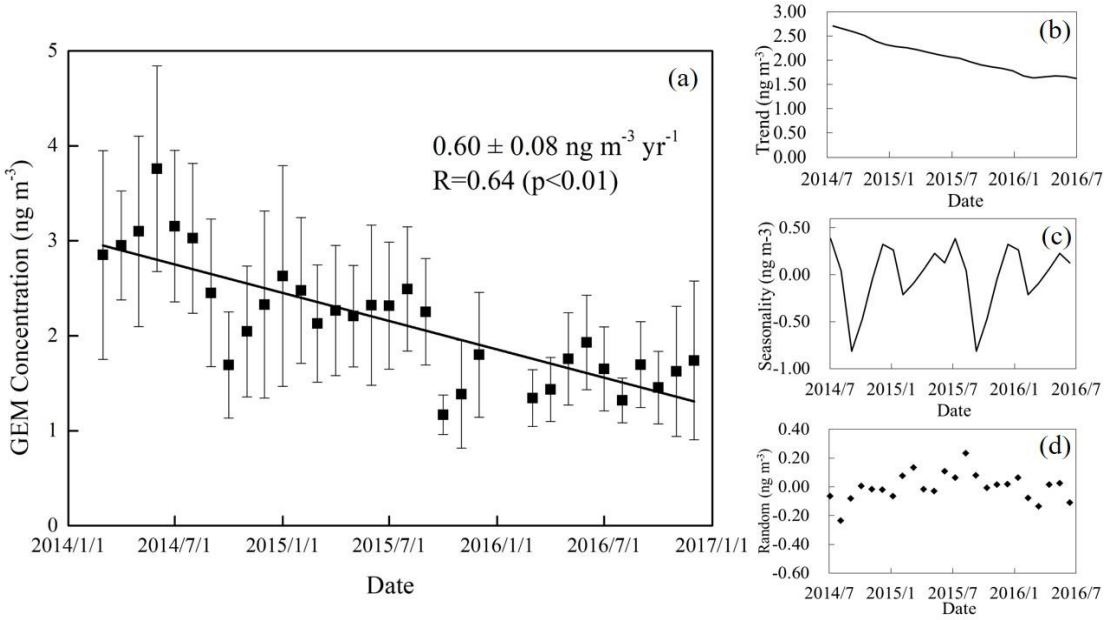


Figure 2. Monthly average GEM concentrations during the studied period (a) observed monthly GEM concentrations (b) GEM trend after decomposition (c) GEM seasonality after decomposition (d) GEM random after decomposition

Note: The observed concentrations during July 2015-April 2016 were TGM concentrations indeed due to the problems of Tekran 1130/1135. However, the GOM concentrations at Chongming Island accounted for less than 1% of TGM. Thus, the GEM concentrations were approximated to TGM concentrations during July 2015-April 2016.

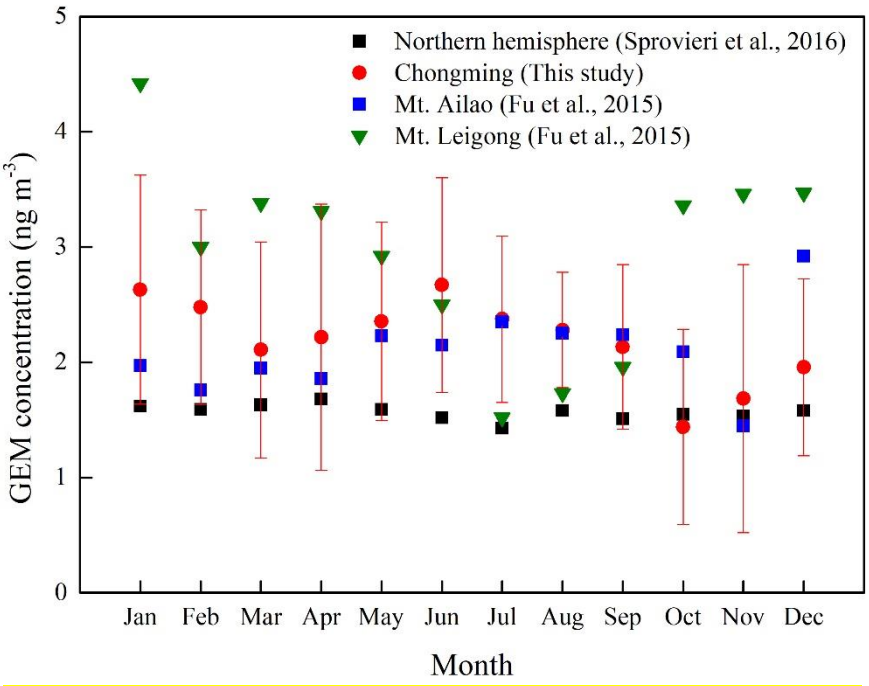


Figure 3. Monthly variations of GEM concentration at remote sites in China

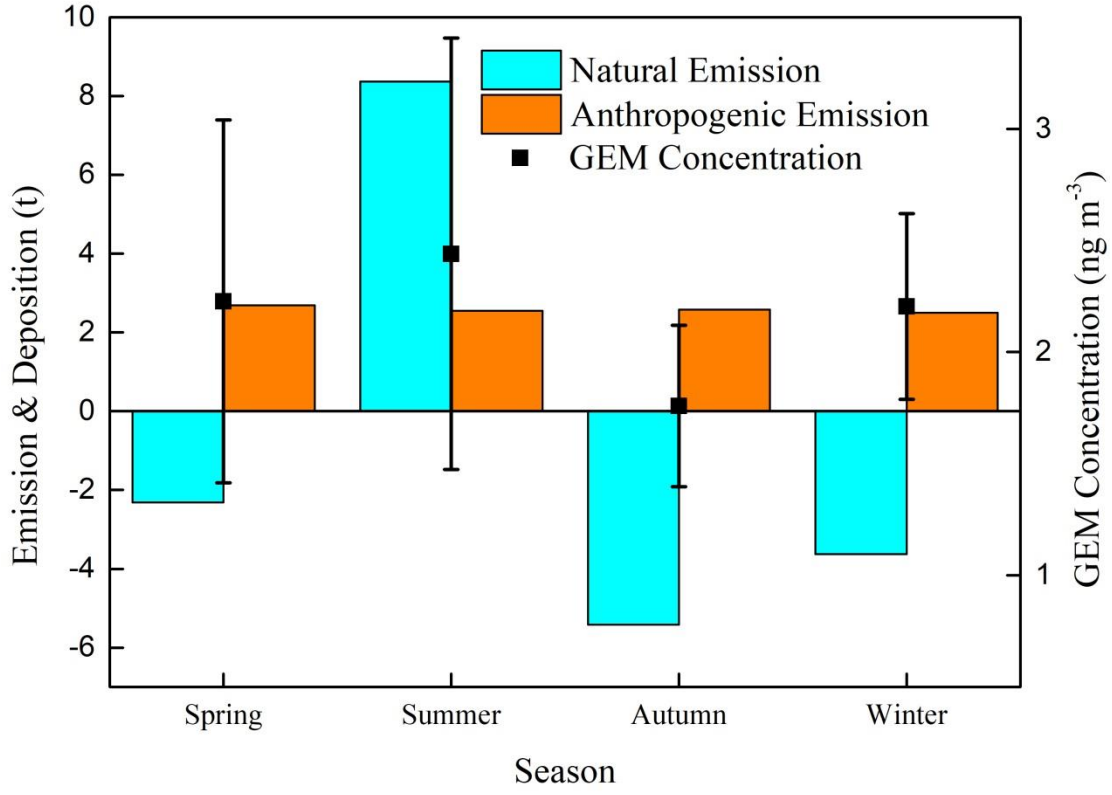
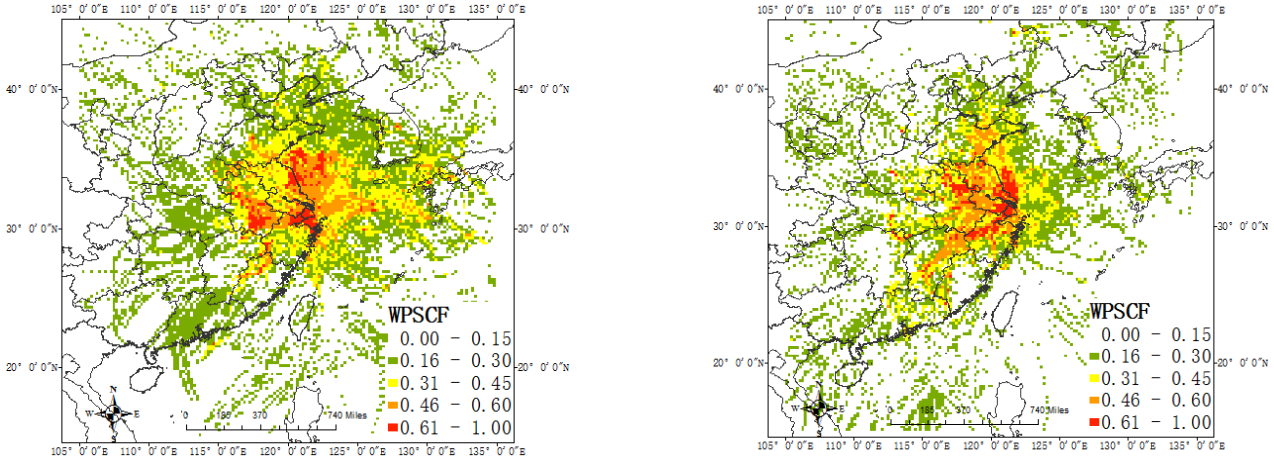
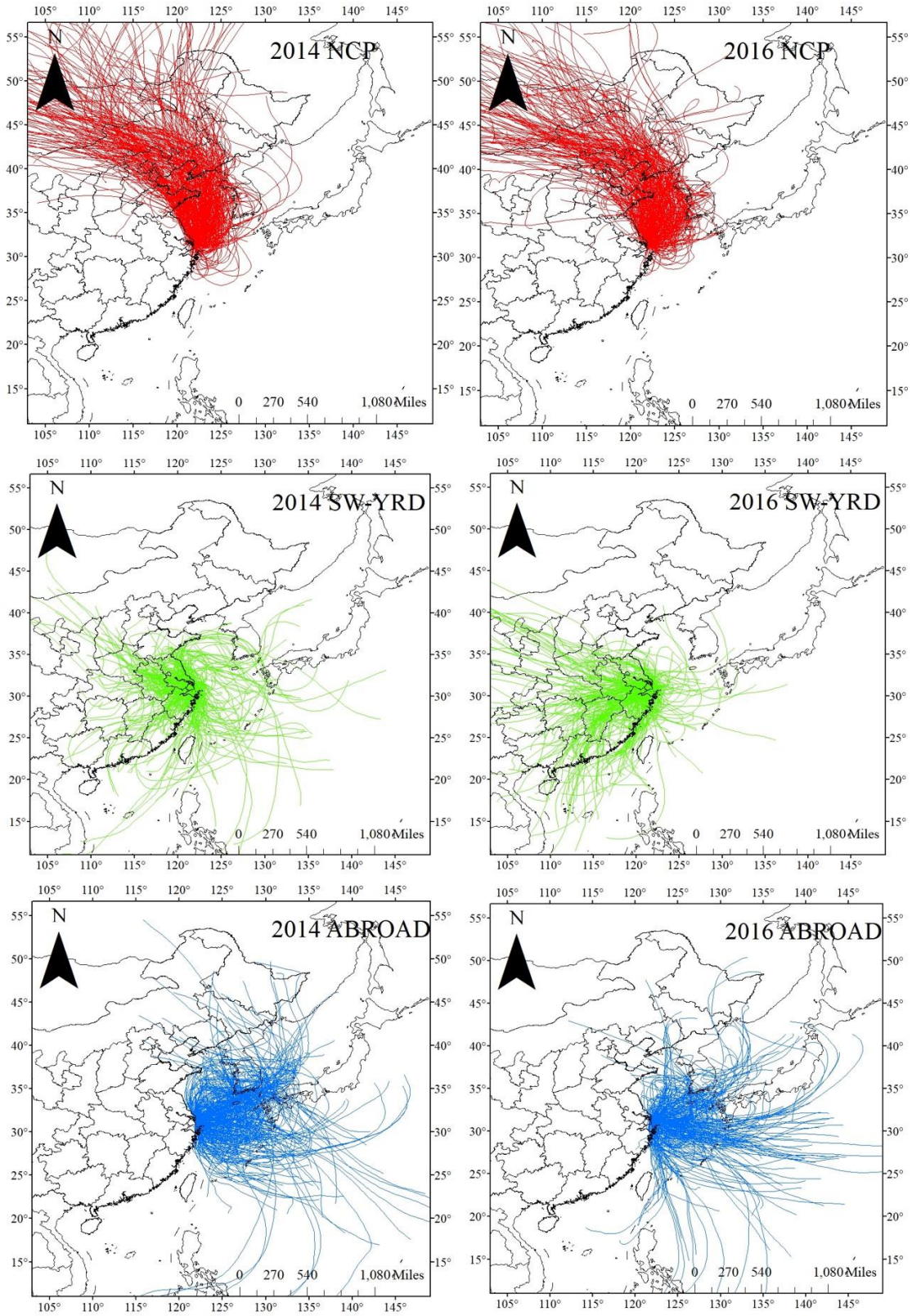


Figure 4. Seasonal cycle of GEM concentrations and natural emissions during 2014-2016. The error bars represent the standard deviation of seasonal average. Positive values of natural emissions represent Hg emitted to air. Otherwise, negative values represent Hg deposition.



(a) 2014 (b) 2016

Figure 5. Source regions of GEM at monitoring site from PSCF model in 2014(a) and 2016(b)



(a) 2014 (b) 2016

Figure 6. The back trajectories map for cluster NCP, SW-YRD and ABROAD in 2014(a) and 2016(b)

(NCP – North China Plain; SW-YRD –Southwest region and Yangtze River Delta; ABROAD – Abroad)

Table 1. PCA component loading of GEM and other air pollutants

Air pollutants	2014		Air pollutants	2016	
	Factor 1	Factor 2		Factor 1	Factor 2
SO_2	**0.76**	0.14	SO_2	**0.82**	-0.09
NO_X	**0.76**	-0.20	NO_X	**0.70**	**-0.52**
O_3	-0.11	**0.98**	O_3	-0.41	**0.97**
$PM_{2.5}$	**0.85**	0.05	$PM_{2.5}$	**0.88**	0.05
GEM	**0.66**	0.02	GEM	**0.78**	-0.19
CO	**0.79**	0.12			
Component	Combustion	Transport of air mass from stratosphere	Component	Combustion	Transport of air mass from stratosphere
Variance explain (%)	49.36	17.53	Variance explain (%)	50.63	25.10

Note: Text in bold phase were regarded as high loading (factor loading>0.40 or <-0.40)

Table 2. Emissions of the main air pollutants in YRD region in 2014

Emission sectors	Annual emissions			
	SO_2 (kt)	NO_x (kt)	$PM_{2.5}$ (kt)	GEM (t)
Coal-fired power plants	918.31	991.62	118.42	14.00
Coal-fired industrial boilers	311.03	271.94	79.91	9.80
Residential coal combustion	68.48	42.11	163.93	0.40
Cement clinker production	207.48	371.13	208.02	4.70
Iron and steel production	480.97	142.80	169.84	2.30
Mobile oil combustion	38.43	1786.74	98.00	1.90
Other sectors	348.83	316.28	382.48	2.50

Table 3. The statistics of cluster and estimated contribution of GEM reduction in 2014 and 2016

Time	Cluster	Trajectories			GEM concentration, C_j (ng m^{-3})	Trajectory weighted concentration, TWC_j,(ng m^{-3})	Contribution to GEM reduction , CR_i
		Numbers	Ratio	Average Ratio (AR)			
2014	NCP	285	33%	32%	2.33	0.79	
	SW-YRD	304	35%	37%	3.19	1.18	
	ABROAD	275	32%	31%	2.58	0.77	
2016	NCP	237	31%	32%	1.48	0.50	26%
	SW-YRD	302	39%	37%	1.87	0.69	44%
	ABROAD	230	30%	31%	1.44	0.43	30%

Supporting information for "*Recent decrease trend of atmospheric mercury concentrations in East China: the influence of anthropogenic emissions* "

Yi Tang[1, 2], Shuxiao Wang[1, 2*], Qingru Wu[1, 2*], Kaiyun Liu[1, 2], Long Wang[3], Shu Li[1], Wei Gao[4], Lei Zhang[5], Haotian Zheng[1, 2], Zhijian Li[1], Jiming Hao[1, 2]

[1] State Key Joint Laboratory of Environmental Simulation and Pollution Control, School of Environment, Tsinghua University, Beijing 100084, China

2 State Environmental Protection Key Laboratory of Sources and Control of Air Pollution Complex, Beijing 100084, China

3 School of Environment and Energy, South China University of Technology, Guangzhou, 510006, China

4 Yangtze River Delta Center for Environmental Meteorology Prediction and Warning, Shanghai, 20030, China

5 State Key Laboratory of Pollution Control & Resource Reuse, School of the Environment, Nanjing University, Nanjing, 210023, China

12 pages (including cover page)

4 Figures (S1, S2, S3, S4)

6 Tables (S1, S2, S3, S4, S5, S6)

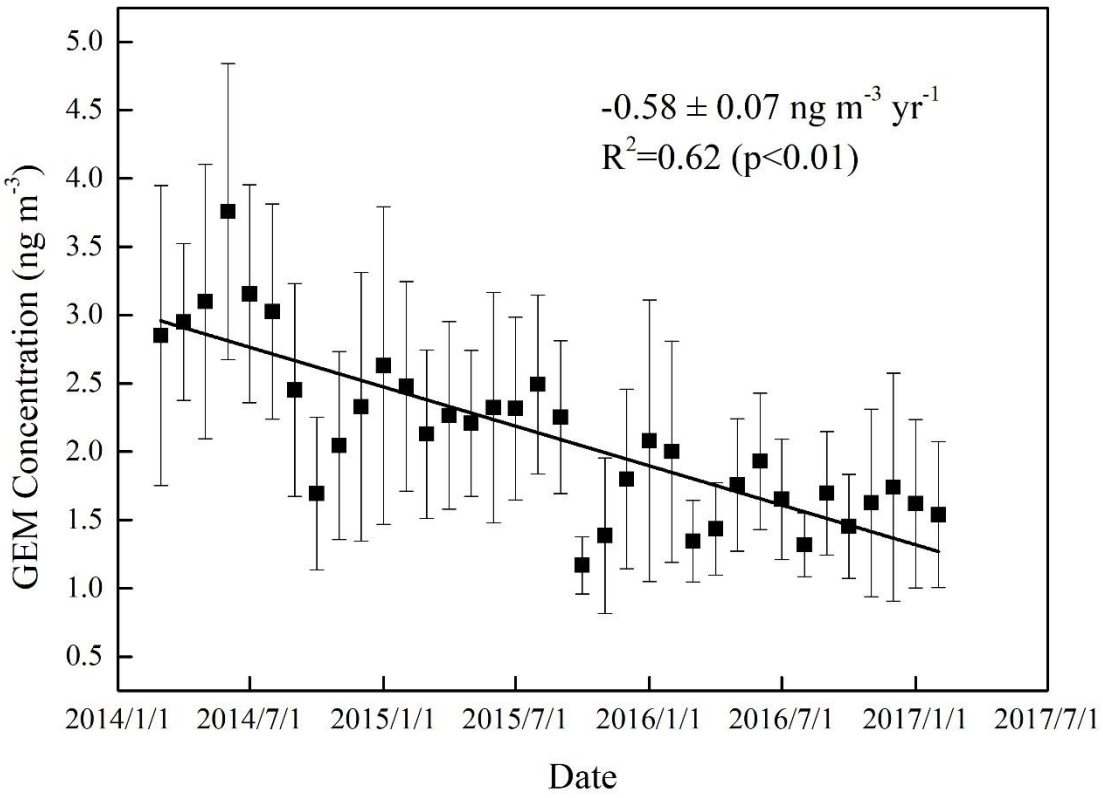


Figure S1. The trend of monthly average GEM concentration from March 2014 to February 2017. The monthly average of GEM in January of 2016 is simulated as the average value that in the January of 2015 and 2017. The same method is used for the data in February of 2016.

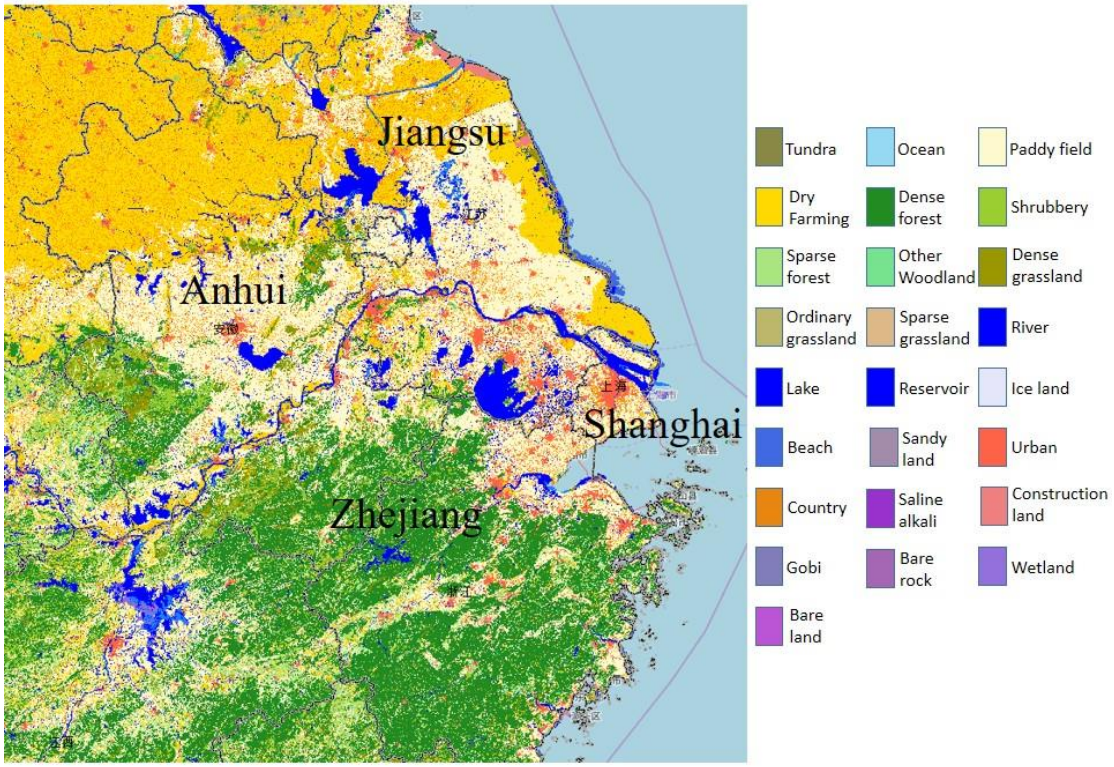


Figure S2. Land use type of YRD region in 2015

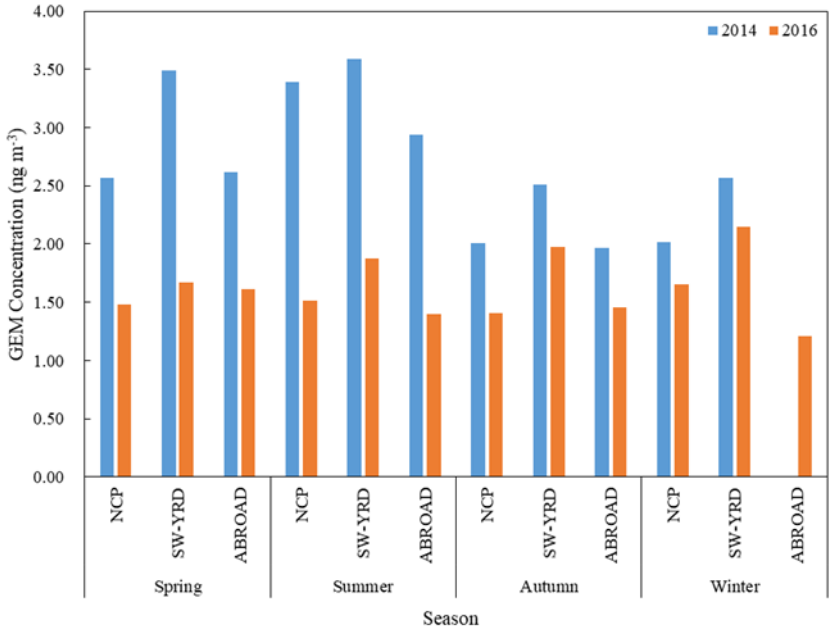


Figure S3. The seasonality of GEM concentration in the NCP, SW-YRD and ABROAD region (No trajectory transport though ABROAD in winter of 2014)

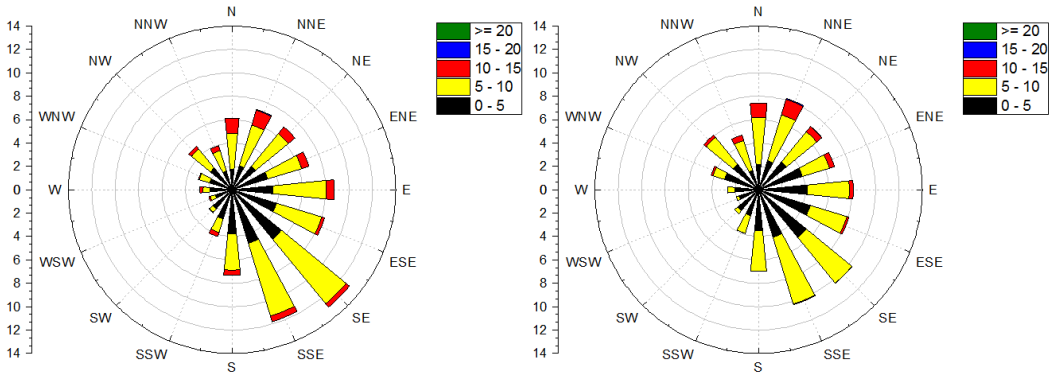


Figure S4. Wind rose at Chongming island in 2014 and 2016

Table S1. Monthly distribution factor for cement clinker production, coal-fired power plants, coal-fired industrial boilers, resident coal combustion, cement clinker production, iron and steel production in potential sources region in 2014 and 2016

Cement clinker production									
2014	Shanghai	Jiangsu	Zhejiang	Anhui	2016	Shanghai	Jiangsu	Zhejiang	Anhui
Jan	0.10	0.06	0.10	0.10	Jan	0.00	0.00	0.00	0.00
Feb	0.00	0.00	0.00	0.00	Feb	0.00	0.00	0.00	0.00
Mar	0.08	0.09	0.08	0.09	Mar	0.04	0.13	0.11	0.11
Apr	0.09	0.09	0.10	0.09	Apr	0.22	0.14	0.10	0.05
May	0.09	0.10	0.09	0.10	May	0.08	0.11	0.10	0.07
Jun	0.09	0.09	0.08	0.09	Jun	0.08	0.13	0.09	0.07
Jul	0.09	0.09	0.08	0.09	Jul	0.09	0.11	0.10	0.07
Aug	0.09	0.09	0.08	0.09	Aug	0.06	0.10	0.08	0.07
Sep	0.08	0.10	0.09	0.08	Sep	0.11	0.09	0.08	0.10
Oct	0.10	0.10	0.10	0.09	Oct	0.08	0.08	0.10	0.09
Nov	0.10	0.10	0.10	0.10	Nov	0.13	0.05	0.11	0.17
Dec	0.10	0.10	0.10	0.10	Dec	0.12	0.06	0.12	0.20
Coal-fired power plants									
2014	Shanghai	Jiangsu	Zhejiang	Anhui	2016	Shanghai	Jiangsu	Zhejiang	Anhui
Jan	0.10	0.08	0.07	0.09	Jan	0.10	0.08	0.07	0.09
Feb	0.10	0.07	0.07	0.08	Feb	0.09	0.07	0.07	0.08
Mar	0.11	0.09	0.09	0.09	Mar	0.09	0.09	0.10	0.08
Apr	0.09	0.10	0.09	0.08	Apr	0.07	0.08	0.08	0.08
May	0.09	0.09	0.09	0.08	May	0.07	0.08	0.08	0.07
Jun	0.07	0.08	0.08	0.08	Jun	0.06	0.08	0.08	0.07
Jul	0.08	0.09	0.09	0.10	Jul	0.10	0.09	0.10	0.09
Aug	0.07	0.08	0.08	0.09	Aug	0.11	0.10	0.10	0.10
Sep	0.05	0.07	0.07	0.07	Sep	0.07	0.08	0.08	0.08
Oct	0.05	0.08	0.08	0.07	Oct	0.07	0.08	0.08	0.07
Nov	0.07	0.08	0.09	0.08	Nov	0.07	0.08	0.08	0.08
Dec	0.11	0.10	0.10	0.09	Dec	0.10	0.09	0.10	0.10
Iron and steel production									
2014	Shanghai	Jiangsu	Zhejiang	Anhui	2016	Shanghai	Jiangsu	Zhejiang	Anhui
Jan	0.09	0.08	0.07	0.07	Jan	0.09	0.08	0.07	0.07
Feb	0.08	0.07	0.07	0.07	Feb	0.09	0.08	0.07	0.07
Mar	0.09	0.08	0.09	0.08	Mar	0.09	0.09	0.09	0.08
Apr	0.09	0.08	0.09	0.08	Apr	0.09	0.09	0.10	0.08
May	0.09	0.08	0.08	0.09	May	0.09	0.08	0.09	0.08
Jun	0.09	0.08	0.09	0.09	Jun	0.08	0.09	0.09	0.08
Jul	0.09	0.08	0.08	0.09	Jul	0.08	0.08	0.09	0.08
Aug	0.09	0.07	0.08	0.09	Aug	0.08	0.08	0.08	0.13
Sep	0.07	0.08	0.08	0.09	Sep	0.08	0.08	0.08	0.08
Oct	0.07	0.08	0.08	0.09	Oct	0.08	0.08	0.08	0.09

Nov	0.07	0.08	0.09	0.08	Nov	0.08	0.08	0.08	0.09
Dec	0.09	0.13	0.10	0.08	Dec	0.08	0.08	0.09	0.09

Coal-fired industrial boilers

2014	Shanghai	Jiangsu	Zhejiang	Anhui	2016	Shanghai	Jiangsu	Zhejiang	Anhui
Jan	0.09	0.08	0.08	0.09	Jan	0.08	0.07	0.08	0.08
Feb	0.08	0.08	0.07	0.09	Feb	0.08	0.07	0.08	0.07
Mar	0.07	0.09	0.09	0.10	Mar	0.09	0.08	0.09	0.09
Apr	0.06	0.08	0.09	0.09	Apr	0.08	0.08	0.09	0.09
May	0.09	0.09	0.08	0.08	May	0.09	0.09	0.08	0.08
Jun	0.09	0.09	0.07	0.08	Jun	0.08	0.08	0.08	0.08
Jul	0.09	0.08	0.08	0.07	Jul	0.08	0.08	0.09	0.09
Aug	0.09	0.07	0.08	0.08	Aug	0.08	0.08	0.08	0.08
Sep	0.08	0.09	0.08	0.09	Sep	0.08	0.08	0.08	0.08
Oct	0.08	0.08	0.09	0.08	Oct	0.09	0.09	0.09	0.09
Nov	0.08	0.08	0.09	0.07	Nov	0.08	0.09	0.09	0.08
Dec	0.09	0.08	0.09	0.08	Dec	0.09	0.09	0.09	0.08

Residential coal combustion

2014	Shanghai	Jiangsu	Zhejiang	Anhui	2016	Shanghai	Jiangsu	Zhejiang	Anhui
Jan	0.33	0.33	0.33	0.33	Jan	0.33	0.33	0.33	0.33
Feb	0.33	0.33	0.33	0.33	Feb	0.33	0.33	0.33	0.33
Mar	0.05	0.05	0.05	0.05	Mar	0.05	0.05	0.05	0.05
Apr	0.00	0.00	0.00	0.00	Apr	0.00	0.00	0.00	0.00
May	0.00	0.00	0.00	0.00	May	0.00	0.00	0.00	0.00
Jun	0.00	0.00	0.00	0.00	Jun	0.00	0.00	0.00	0.00
Jul	0.00	0.00	0.00	0.00	Jul	0.00	0.00	0.00	0.00
Aug	0.00	0.00	0.00	0.00	Aug	0.00	0.00	0.00	0.00
Sep	0.00	0.00	0.00	0.00	Sep	0.00	0.00	0.00	0.00
Oct	0.00	0.00	0.00	0.00	Oct	0.00	0.00	0.00	0.00
Nov	0.05	0.05	0.05	0.05	Nov	0.05	0.05	0.05	0.05
Dec	0.23	0.23	0.23	0.23	Dec	0.23	0.23	0.23	0.23

Table S2. The meteorological condition between 2014 and 2016 in Chongming

Month	Temperature		Solar Radiation		Releative Humidity	
	2014(℃)	2016(℃)	2014(W m^{-2})	2016(W m^{-2})	2014 (%)	2016 (%)
1		4.39		104.47		71.28
2		5.73		143.68		67.69
3	10.14	9.55	156.61	150.31	79.66	71.43
4	14.85	14.77	163.29	161.88	75.11	80.35
5	20.59	19.24	181.49	176.04	78.15	78.50
6	22.87	22.98	171.20	156.63	82.98	83.00
7	26.86	27.82	244.30	224.52	84.62	82.24
8	24.64	28.31	262.20	253.61	78.38	74.03
9	22.08	24.49	197.20	153.34	74.36	75.27
10	17.76	20.82	168.21	122.81	70.47	75.67
11	10.94	13.64	117.01	113.00	68.96	75.25
12	5.34	8.99	114.60	103.87	59.47	71.62
Average	17.61	16.73	177.61	155.35	75.22	75.53

Table S3. The amount of valid data during sampling period

Year	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
2014			5914	6125	6493	5568	4634	6255	6491	7106	7578	5564
2015	5227	4532	5216	3392	4072	4797	7591	6538	3434	2223	4363	8833
2016			1370	8293	7476	5884	5424	5641	3561	4544	6292	4589

Table S4. Historical variation trends of atmospheric Hg in previous studies

Monitoring site	Duration	TGM trend (pg m^{-3} yr^{-1})	Variation trend	Site description	Monitoring instrument	References
Alert, Canada	2000-2009	-13(-21,0)	-0.9% y^{-1}	Remote	2537A	Cole et al. 2013
Kuujjuarapik, Canada	2000-2009	-33(-50,-18)	-2.1% y^{-1}	Remote	2537A	Cole et al. 2013
Egbert, Canada	2000-2009	-35(-44,-27)	-2.2% y^{-1}	Remote	2537A	Cole et al. 2013
Zeppelin Stn, Norway	2000-2009	+2(-7,+12)	no trend	Remote	2537A	Cole et al. 2013
St.Anicet, Canada	2000-2009	-29(-31,-27)	-1.9% y^{-1}	Remote	2537A	Cole et al. 2013
Kejimkujik, Canada	2000-2009	-23(-33,-13)	-1.6% y^{-1}	Remote	2537A	Cole et al. 2013
Head, Ireland	1996-2009		-1.3±0.2% y^{-1}	Rural	2537A	Weigelt et al. 2015
Yong San, South Korea	2004-2011	No trend (3.54±1.46 ng m^3)		Urban	AM-3	Kim et al. 2016
Yong San South Korea	2013-2014	Decrease to 2.34±0.73 ng m^3			AM-3	Kim et al. 2016
Mt. Changbai	2013-2015	Decrease from 1.74 ng m^{-3} to 1.58 ng m^{-3}		Remote	2537B	Fu et al. 2015
Chongming Island, China	2014-2016	-600	-29.4%/y	Remote	2537X	This study

Table S5. The annual concentration of SO_2, NO_x, O_3 and $PM_{2.5}$ at Chongming site, NCP, and SW-YRD regions

Year		2014			2016			Change		
Pollutants	Region	NCP	SW-YRD	Chongming	NCP	SW-YRD	Chongming	NCP	SW-YRD	Chongming
$PM_{2.5}$ ($\mu g\ m^{-3}$)		71.93	53.05	25.09	60.75	44.75	23.89	-16%	-16%	-5%
SO_2 ($\mu g\ m^{-3}$)		34.52	21.01	1.60	24.37	16.40	1.47	-29%	-22%	-8%
NO_2 ($\mu g\ m^{-3}$)		45.07	34.34	12.62	41.55	34.40	10.84	-8%	0%	-14%
O_3 ($\mu g\ m^{-3}$)		60.29	56.27	41.70	61.84	60.92	44.38	3%	8%	6%
GEM ($ng\ m^{-3}$)		No data		2.68	No data		1.60	No data		-40%

Note: According to the contribution of trajectory, the dominant provinces in the NCP region included Beijing, Tianjin, Hebei, Shandong and Liaoning province. The SW-YRD mainly contained Shanghai, Zhejiang, Jiangsu, Jiangxi and Anhui province.

Table S6. Emission inventories of the main pollutants from the studied regions in 2014 and 2016

Air pollutants	2014		2016		Decline proportion	
	NCP	SW-YRD	NCP	SW-YRD	NCP	SW-YRD
$PM_{2.5}$ (kt)	2019	1209	1849	1109	-8%	-8%
NO_x (kt)	5697	4022	5424	3855	-5%	-4%
SO_2 (kt)	3780	1993	3450	1780	-9%	-11%
GEM (t)	118	72	103	67	-13%	-7%

Note: According to the contribution of trajectory, the dominant provinces in the NCP region included Beijing, Tianjin, Hebei, Shandong and Liaoning province. The SW-YRD mainly contained Shanghai, Zhejiang, Jiangsu, Jiangxi and A