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

30　　determine the dominant impact factors. So it is hard to get some additonal evidence for the

31　　reason of GEM seasonal variation from the seasonal variation of GOM and PBM.
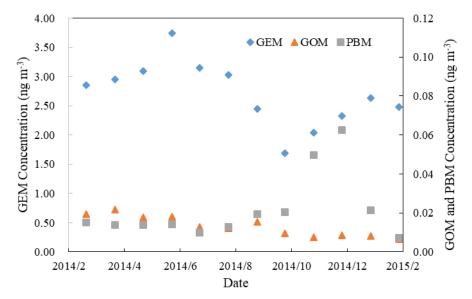

32　　**Figure R1**. Monthly variation of GEM, GOM and PBM at Chongming from March 2014 to

33　　February 2015

34

35　　The calculation process of GEM emissions is revised as below.

36　　　"Regional atmospheric GEM emissions by month are calculated by using both the

37　　technology-based emission factor methods and transformed normal distribution function

38　　method. Detailed introduction of these two methods and the speciation profile of the emitted

39　　Hg for each sector are described in our previous study (Wu et al., 2016)."

40　　**See the revised manuscript, line 206-209**

41

42　　*Comment 2：*

43　　　Section 3.1: Averages and their standard deviations should always be stated with the number

44　　of measurements because only then statistical tests for significance of differences can be made.

45　　In line 194 the authors claim that GEM concentrations in 2014 were significantly higher than…

46　　- at which significance level? Line 199: the annual decrease rate should be given with its

47　　uncertainty and number of months.

48　　*Response:*

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

50   statistical tests of significance of differences.

51   The GEM concentrations in 2014 were significantly higher than the background

52   concentration of Northern Hemisphere at the significance level with p value less than 0.01.

53   "The GEM concentrations in 2014 were higher (*t* test, $p<0.01$) than the Northern Hemisphere

54   back-ground concentration (about 1.5 ng m^{-3}) (Sprovieri et al., 2010) and those measured in

55   other remote and rural locations in China (Zhang H et al., 2015; Fu et al., 2008a; Fu et al.,

56   2009)."

57   **See revised manuscript, line 231 - 234.**

58

59   The annual decrease rate has been given with its uncertainty and the number of months. In

60   addition, the number of valid data to calculate the monthly average is listed in Table S3.

61   "During this period, monthly GEM concentrations showed a significant decrease with a rate of

62   -0.60 ± 0.08 ng m^{-3} yr^{-1} (R^2=0.64, $p<0.01$ significance level, n = 32) (Figure 2a)."

63   **See revised manuscript, line 236 - 237.**

64

65   **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

66   **See revised manuscript, supporting information Table S3.**

67

68   ***Comment 3:***

69   Figure 3: It is not clear how the points in Figure 3 were calculated? Were the data detrended

70   before the averaging? In view of the strong downward trend they should be. What is the

71   standard deviation or standard error of the monthly means – please show them as vertical bars

72   in Fig. 3. Are the differences between the months statistically significant? This is a precondition

73   for the discussion of the seasonal variation.

74   ***Response:***

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

76 average during sampling period. The monthly GEM concentrations were detrended before the

77 average (Figure 2). The standard deviations of the monthly means have been added.

78

79 "According to the decomposition result (Figure 2c), we observed strong seasonal cycle with

80 seasonal GEM peak in July and trough in September, so GEM concentrations in the same month

81 but different years were averaged to discuss the seasonal circle (Figure 3). The average data

82 can eliminate the effect of downward trend and get result of average seasonal variation. The

83 error bars in the Figure 3 mean the standard deviation of the monthly average."
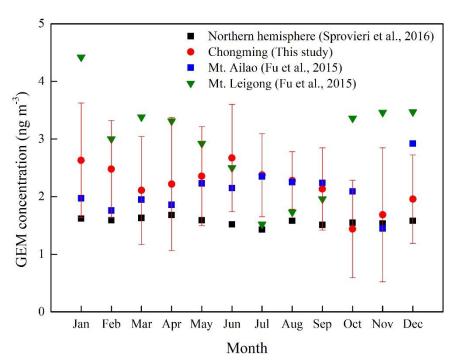
84 **See revised manuscript, line 274 –278.**

85

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

87 **See revised manuscript at Figure 3.**

88

89 The difference between month are statistically significant (F test, p<0.001). In addition, the

90 observed GEM concentration signal was decomposed (signal = trend + seasonal + random,

91 example here: https://anomaly.io/seasonal-trend-decomposition-in-r/). By using this method,

92 we also observed very strong detrended seasonal cycle where GEM peak was observed in July

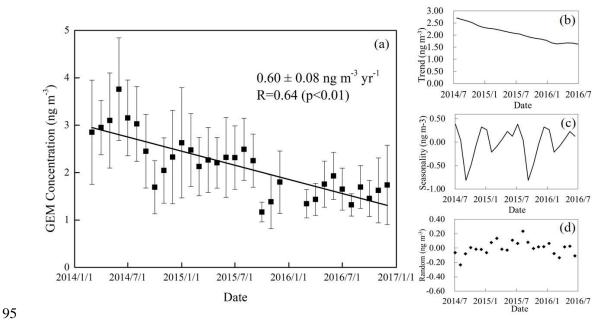93   and the GEM trough was in September.

94

95

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.

103

104   ***Comment 4:***

105   4.1 Figure 4 and its capture: This figure needs substantial revision to illustrate the point the
106   authors make and to make it understandable for the readers. Negative emissions are deposition
107   fluxes and should be named as such. Thus "natural emissions" in spring, autumn and winter are
108   in fact deposition fluxes. Net fluxes are needed to illustrate the point made by the authors but
109   they are not shown. The capture should also state that it is about the emissions and depositions
110   in the YRD region?

111   ***Response:***

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

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

114   Therefore, emissions and flux are different concepts in this study. We use "natural emissions"

115   instead of "natural flux" to correspond to "anthropogenic emissions". It should be pointed out

116   that the natural emission is a concept of net natural emission, which reflected a net effect of

117   two competing processes (Zhang, 2009): total natural Hg emissions and total Hg deposition.

118   When the value is positive, it means the net effect is Hg emissions to air. Otherwise, Hg

119   deposites. We have made this concept clear in both text and Figure 4.

120

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

122
$$E_N = \sum_i F_i \times A_i \times t \tag{6}$$

123   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

124   the studied year, yr. The bi-directional Hg flux was obtained from the study of Wang et al. (2016)

125   directly. It should be pointed out that the natural emission is a concept of net emission in this

126   manuscript, which reflected a net effect of two competing processes (Zhang, 2009): total Hg

127   natural emissions and total Hg deposition. The total natural emissions included primary natural

128   release and re-emission of legacy Hg stored in the terrestrial and water surface (Wang et al.,

129   2016). When the value is positive, it means the net effect is Hg emissions to air. Otherwise, Hg

130   deposited."

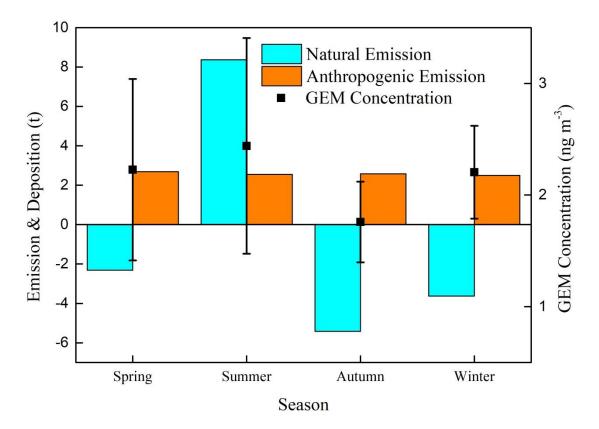131   **See revised manuscript at line 219 – line 227.**

132

133

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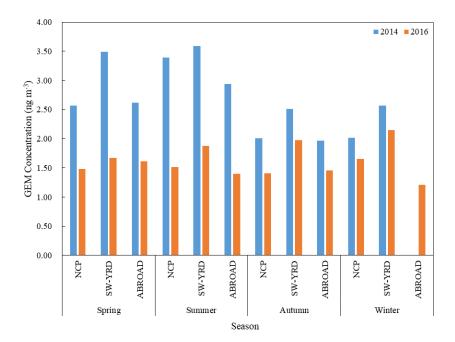
Figure S3. The seasonality of GEM concentration in the NCP, SW-YRD and ABORD region
(No trajectory transport though ABROAD in winter of 2014)

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 314 - 317

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

169 anthropogenically emitted mercury removed from the atmosphere if there are only "natural"

170 deposition fluxes? Please use the correct terminology and separate the natural and

171 anthropogenic emissions from the deposition fluxes of both.

172 ***Response:***

173   Sorry for the misunderstanding. To avoid confusion, we deleted the sentence of "The annual

174   emissions from both natural source and anthropogenic source… was 0.75 and 10.3 t,

175   respectively" in the revised manuscript. The impact of anthropogenic emissions and natural

176   emissions were discussed separately. The anthropogenic emissions were in the range of 2.5 –

177   2.7 t while natural emissions varied from -5.4 – 8.4 t in different season. Thus, one important

178   conclusion of our study is that the seasonal GEM cycle was dominated by the natural

179   emissions.

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

181 concentrations at a remote site are generally regarded under the impact of regional emissions.

182 Therefore, the emissions in the YRD regions (Anhui, Zhejiang, Jiangsu, and Shanghai) were

183 calculated. However, the anthropogenic emissions were in the range of 2.5-2.7 t, which is

184 almost unchanged. Compared to the anthropogenic emissions, we observed almost

185 synchronized trends between natural emissions and air Hg concentrations in Figure 4."

186 **See the revised manuscript at line 296 - 301**

187

188   It is difficult to distinguish whether the deposited Hg is from natural sources or anthropogenic

189 sources. Therefore, the bi-directional Hg flux scheme contained total Hg deposition flux (both

190 so called "natural" and "anthropogenic" deposition fluxes) (Zhang et al., 2009). And the natural

191 Hg emissions in this study have considered the removal of anthropogenic Hg emissions. We

192 have clearly defined the concept of "natural emissions" in this study. The natural emission is a

193 concept of net natural emission, which reflected a net effect of total natural Hg emissions and

194 total Hg deposition amount. Therefore, the data of natural emissions in the four seasons contains

195 both emissions and deposition. The positive value in summer means that net effect is Hg

196 emissions to air. In other three seasons, Hg deposited. Detailed revision is described as follows.

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

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

198

199 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

200 studied year, yr. The bi-directional Hg flux was obtained from the study of Wang et al. (2016)

201 directly. It should be pointed out that the natural emission is a concept of net emission in this

202 manuscript, which reflected a net effect of two competing processes (Zhang, 2009): total Hg

203 natural emissions and total Hg deposition. The total natural emissions included primary natural

204 release and re-emission of legacy Hg stored in the terrestrial and water surface (Wang et al.,

205 2016). When the value of E_N is positive, it means the net effect is Hg emissions to air. Otherwise,

206 Hg deposited."

207 **See the revised manuscript at line 219 - 227**

208

209 ***Comment 6:***

210 The results of PCA analysis and Table 2: The authors attribute the factor 2 to "exchange of

211 PBL with free troposphere" but do not explain why. Last row in the table 2 called "variance

212 explain" lists for 2016 exchange of PBL with the free troposphere 75.735 which together with

213 50.625 for "combustion" makes more than 100. As such the units of "variance explain" cannot

214 be percent. What are the numbers in this row and does it make sense to present them with three

215 valid numbers after decimal point?

216 ***Response:***

217 We have added explanation as follows.

218 "Considering the low loading of CO and high loading of O$_3$, the factor 2 can be viewed as a

219 sign of the invasion of air mass from stratosphere (Fishman and Seiler, 1983; Jaffe, 2010). The

220 air mass from stratosphere will increase the O$_3$ concentration. O$_3$ react with NO, which makes

221 a negative correlation with NO. However, the low loading on GEM of factor 2 indicated that

222 Factor 2 had no relationship with GEM concentrations at Chongming from the aspect of whole

223 year data."

224 **See the revised manuscript at line 350 - 355**

225

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

227 contribution ratio of factor 1 and factor 2 in the total variance. We have revised the data to two

228 valid numbers after decimal point.

229 **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	Invasion of air mass from stratosphere	Component	Combustion	Invasion of air mass from stratosphere
Variance explain (%)	49.36	17.53	Variance explain (%)	50.63	25.10

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

231

232 ***Comment 7:***

233 Table 3: The numbers are probably annual emissions but the capture does not say it. The year

234 of the emissions is not given. I wonder about the "other SO2 sources" which are substantially

235 larger than all coal, oil, and biomass burning taken together. If it is not an error, what are the

236 "other SO2 sources"?

237 ***Response：***

238   Thank for the comments. It is the emission in 2014. The emission for "other SO_2 sources" is

239 a typo and we have revised this. The other sectors contain municipal solid incineration, copper

240 smelting, aluminum production, gold production, other coal combustion, stationary oil

241 combustion, and cremation. Table 2 has been revised as follows.

242 **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

243 "The studied emission sectors included coal-fired power plants, coal-fired industrial boilers,

244 residential coal-combustion, cement clinker production, iron and steel production, and other

245 small emission sectors (eg., zinc smelting, lead smelting, municipal solid incineration, copper

246 smelting, aluminum production, gold production, other coal combustion, stationary oil

247 combustion, and cremation)."

248 **See the revised manuscript at line 213 - 216**

249

250 ***Comment 8:***

251 Chapter 3.4 misses a major point: Table 3 of SO_2, NO_x, $PM_{2.5}$, and GEM emissions is only

252 for one undefined year and only for the YRD region. Table 4 and Figure 6 show a dominant

253 influence of transport from NW of China which is mostly outside of the YRD region. To

254 illustrate convincingly the major conclusion of the paper one would need a table with the

255 inventories for NW and SW (perhaps separately) and for 2014 and 2016.

256 ***Response：***

257 According to our response to comment 4, we have adjusted the cluster. Based on the adjusted

258 cluster, we added the emissions of SO_2, NO_x and $PM_{2.5}$ in both 2014 and 2016 to illustrate the

259 change of emission inventory in NCP and SW-YRD region (Table S5). According to the table,

260 we observed obvious emission decline of the above pollutants.

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

262 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%

263 Note: According to the contribution of trajectory, the dominant provinces in the NCP region

264 included Beijing, Tianjin, Hebei, Shandong and Liaoning province. The SW-YRD mainly

265 contained Shanghai, Zhejiang, Jiangsu, Jiangxi and Anhui province.

266 **See the revised manuscript at line 382 - 384**

267

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.

290   The Figure S1 was revised as below.

The trend plot shows GEM Concentration (ng m⁻³) on the y-axis ranging from 0.5 to 5.0, plotted against Date on the x-axis from 2014/1/1 to 2017/7/1. Annotation: -0.58 ± 0.07 ng m^{-3} yr^{-1}, $R^2=0.62$ ($p<0.01$).
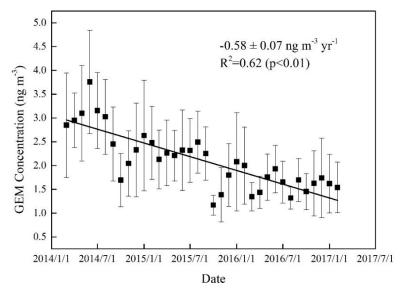
291   **Figure S1.** The trend of monthly average GEM concentration from March 2014 to February

292   2017. The monthly average of GEM in January of 2016 is simulated as the average value that

293   in the January of 2015 and 2017. The same method is used for the data in February of 2016.

294

295   *Comment 11:*

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

297   *Response:*

298   The downward annual rate was given with its standard error as follows. See the Figure S1 in

299   the response of comment 10.

300

301   *Comment 12:*

302   Line 51: "Both GOM and PBM are more soluble." than what? PBM is not necessarily more

303   soluble than GEM but it is scavenged by wet deposition. Low solubility of GEM need to be

304   mentioned before this statement.

305   *Response :*

306   The statement has been revised as follows.

307   "GOM is much soluble than GEM, and PBM can be quickly scavenged by both dry and wet

308   deposition. Therefore, the residence time of both GOM and PBM is shorter than that of GEM,

309   generally several days to a few weeks for GOM and 0.5 – 2 year for GEM (Schroeder and

310   Munthe, 1998). "

311   **See the revised manuscript at line 52 - 55.**

312

313   *Comment 13:*

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

315   *Response ：*

316   We have revised the manuscript as follow.

317   "However, recently atmospheric Hg emissions in China have been estimated to decrease since

318   2012 (Wu et al., 2016)."

319   **See the revised manuscript at line 82**

320

321   *Comment 14:*

322   Line 86: …is located…

323   *Response:*

324   We have revised the manuscript as suggested.

325   "As China's third largest island, Chongming Island is located in the east of Yangtze River Delta

326   region with a typical subtropical monsoon climate."

327   **See the revised manuscript at line 97**

328

329   *Comment 15:*

330   Lines 102/103: … the error between gold trap A and gold trap B was limited to…? Probably

331   the difference instead of error was limited. What happens if the difference is more than the

332   limit?

333   *Response:*

334   We have replaced the error with differences. If the difference is more than the limit, it means

335   that the gold traps are passivizing and we need to replace the old gold trap A and gold trap B.

336

337   *Comment 16:*

338   Lines 171/172: "uncertainties" would be better than "errors"

339 *Response:*

340   We have revised the manuscript as suggested.

341

342 *Comment 17:*

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

344 *Response*

345   We have revised the manuscript as suggested.

346

347 *Comment 18:*

348 Lines 207-209: A reference to Martin et al (2017) is not correct because the paper does not

349 contain annual averages and the authors of this paper do not mention a gap in the measurements

350 between 2004 and 2007. The correct reference would be: annual average GEM concentration

351 decreased from 1.29 ng m-3 in 1996 to 1.19 ng m-3 in 2004 (Slemr et al., GRL 35, L11807,

352 doi:10.1029/2008GL033741, 2008) and were increasing from 0.93 ng m-3 in 2007 (Slemr et

353 al., ACP 15, 3125-3133, 2015) until 2016 (Martin et al, 2017).

354 *Response:*

355   We have revised the manuscript as suggested.

356 "In South Africa, annual average GEM concentration at Cape Point decreased from 1.29 ng

357 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}

358 in 2007 (Slemr et al., 2015) until 2016 (Martin et al, 2017)."

359 **See the manuscript at line 261– 263**

360

361 # References:

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389   Zhang L, Wang L, Wang S, Dou H, Li J, Li S, Hao J. Characteristics and Sources of Speciated

390   Atmospheric Mercury at a Coastal Site in the East China Sea Region. Aerosol and Air Quality

391   Research, 2017, 17(12): 2913-2923.

392   Zhang, L. M., Wright, L. P., and Blanchard, P.: A review of current knowledge concerning dry

393   deposition of atmospheric mercury, Atmos. Environ., 43, 5853-5864, 2009.

394   Zhang Y, Jacob D J, Horowitz H M, Chen L, Amos H M, Krabbenhoft D P, Slemr F, St Louis V L,

395   Sunderland E M. Observed Decrease in Atmospheric Mercury Explained by Global Decline in

396   Anthropogenic Emissions. Proceedings of the National Academy of Sciences of the United States

397   of America, 2016, 113(3): 526.

Reply to Comments from Reviewer #2

399   We thank the editor and reviewers' comments which help us improve the manuscript. We

400 have carefully revised our manuscript following the reviewers' comments. A point-to-point

401 response is given below. The reviewers' comments are in black and our replies are in blue.

402 **To Reviewer**

403 *Major Comment:*

404   I really think you should perform a trend decomposition of the signal (signal = seasonal +

405 trend + random, example here: https://anomaly.io/seasonal-trend-decomposition-in-r/). There

406 is a very strong seasonal cycle and you conclude that "the seasonal GEM cycle was dominated

407 by the natural emissions". However, how can you explain that the seasonality is way more

408 pronounced in 2014? To me, presenting emissions inventories is not convincing enough; how

409 you can you be sure that the decreasing trend is not driven by a change in seasonality?

410 *Response:*

411   The trend decomposition of the signal is performed in Figure 2 (signal = trend + average

412 variation + random). By using this method, the decreasing trend was shown in Figure 2b and

413 the seasonality is approximately 12 months. In each cycle, we observed very strong seasonal

414 cycle where GEM peak was observed in July and the GEM trough was in September.

415   The seasonal variation was more pronounced in 2014 can attribute to the lower wet

416 deposition and GEM oxidation. On one aspect, as a costal site, the Chongming island is

417 abundant with ·OH radicals. The increase of O_3 concentration from the summer of 2014 to 2016

418 may contribute to a higher oxidation of GEM in 2016. On another aspect, and higher wet Hg

419 deposition is approximately 6.6 times of that in the winter at Chongming (Zhang et al., 2010).

420 Meanwhile, the rainfall in 2016 summer (546 mm) was higher than the rainfall in 2014 (426

421 mm). Therefore, the higher oxidation and wet deposition rate of mercury in the summer of 2016

422 will reduce the peak of GEM concentration and then shrink the concentration difference

423 between summer and winter, which lead to a less pronounced seasonal variation in 2016.
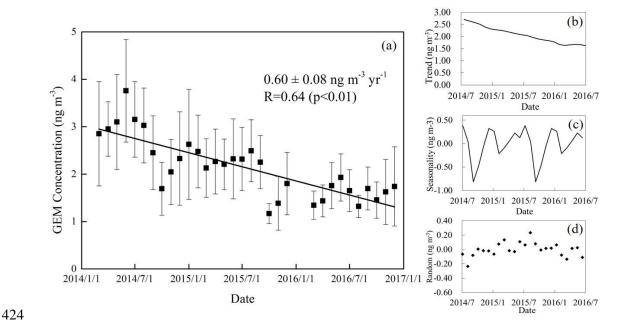
424

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.

The revised manuscript of decomposition trend is revised as below.

   "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)."

See the revised manuscript at line 238 - 243

   "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

444   of 2014 to 2016 may contribute to a higher oxidation of GEM in 2016. On another aspect, and

445   higher wet Hg deposition is approximately 6.6 times of that in the winter at Chongming (Zhang

446   et al., 2010). Meanwhile, the rainfall in 2016 summer (546 mm) was higher than the rainfall in

447   2014 (426 mm). Therefore, the higher oxidation and wet deposition rate of Hg in the summer

448   of 2016 will reduce the concentration difference between summer and winter, which lead to a

449   less pronounced seasonal variation in 2016. Meanwhile, the higher oxidation and wet

450   deposition in 2016 also contributed to the downward trend of GEM by reducing the seasonality

451   in spring and summer (Figure S3)."

452   **See the revised manuscript at line 318 - 328**

453

454   While SO_2, NO_2, and PM concentrations were monitored, data are not presented nor

455   discussed. Do you also observe a decreasing trend? That would be the best way to support that

456   "air pollution control policies targeting SO_2, NO_2, and PM reductions had significant co-

457   benefits on atmospheric Hg".

458   *Response:*

459   To further verify the cause of downward trend of atmospheric Hg, we give the emission

460   inventory (Table S6) and concentrations (Table S5) of other air pollutants in the studied regions

461   in both 2014 and 2016. Both the emissions and concentrations of SO_2, NO_2, and PM showed a

462   decreasing trend, which is used to support that "air pollution control policies targeting SO_2,

463   NO_2, and PM reductions had significant co-benefits on atmospheric Hg".

464

465   "Table 3 showed the detailed data of the three classifications. From 2014 to 2016, the whole

466   China region (NCP, SW-YRD) contributed to 70% of GEM decline at Chongming Island.

467   Considering downward trend of emission inventory and atmospheric pollutant from 2014 to

468   2016 in NCP and SW-YRD region (Table S5, Table S6), the reason of downward trend can be

469   attributed to the effectiveness of existing air pollution control measures in China (SC, 2013;

470   MEP, 2014)."

471   **See the revised manuscript at line 380 – 384**

472

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

Year		2014			2016			Change		
Pollutants	Region	NCP	SW-YRD	Chongming	NCP	SW-YRD	Chongming	NCP	SW-YRD	Chongming
PM$_{2.5}$ (µg m^{-3})		71.93	53.05	25.09	60.75	44.75	23.89	-16%	-16%	-5%
SO$_2$ (µg m^{-3})		34.52	21.01	1.60	24.37	16.40	1.47	-29%	-22%	-8%
NO$_2$ (µg m^{-3})		45.07	34.34	12.62	41.55	34.40	10.84	-8%	0%	-14%
O$_3$ (µg 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%

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

478

479   **Table S6.** Emission inventories of the main pollutants from the studied regions in 2014 and
480   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%

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

484

485   Finally, I wonder why GOM and PBM data are not reported and discussed. Do you also

486   observe a decreasing trend? You may have encountered issues with the speciation unit. If so,

487   was the experimental setup identical in 2014 and 2016, or did you analyze GEM when the

488   speciation unit was working vs. TGM when it wasn't? A discussion on analytical uncertainties

489   would be much welcomed.

490   The GOM and PBM data are not reported finally due to the following reasons. First, the main

491   purpose of this manuscript is to validate the anthropogenic Hg emissions reduction through

492   observation data. The Chongming sit is a background site. Considering the stability of GEM in

493   the air, we choose GEM as an index to reflect the emission control effect. Second, the method

494   we developed to explain the GEM trend is not applicable for GOM and PBM. Except emissions,

495   we think the potential reactions in the air are significant factors to impact both GOM and PBM

496   concentrations. But we need more evidence to prove our assumptions. Therefore, we deleted

497   the discussion of GOM and PBM in our final manuscript.

498   We also observed decreasing trend of PBM. But the GOM kept increasing. Currently, we

499   need more study to explain this phenomenon.

500   Yes, we also encountered issues with the speciation unit. From July 5, 2015 to April 30 2016,

501   the Tekran 1130/1135 speciation unit was damaged by the rainstorm, the Tekran 2537X were

502   operated without speciation units but with PTFE filter to protect the instrument from particles

503   and sea salt. The average concentration of GOM and PBM during sampling period was 14.81

504   ± 13.21 and 20.10 ± 34.02 pg m^{-3}, respectively. The GOM fraction in TGM was less than 1%

505   at Chongming island (Li et al., 2016; Zhang et al., 2017). Therefore, the GEM concentrations

506   were approximated to TGM concentrations July 5, 2015 to April 30 2016 when the speciation

507   unit does not work, as most of other studies have done (Slemr et al., 2015; Sprovieri et al.,

508   2016). In addition, uncertainty analysis was performed to point out potential impact. The

509   revision is as follows.

510   "From July 5, 2015 to April 30 2016, the Tekran 1130/1135 speciation unit was damaged by

511   the rainstorm, the Tekran 2537X were operated without speciation units but with PTFE filter to

512   protect the instrument from particles and sea salt. Therefore, the observed concentrations during

513   July 2015-April 2016 were TGM concentrations indeed. However, the GOM concentrations at

514   Chongming Island accounted for less than 1% of TGM (TGM=GOM+GEM). Thus, the GEM

515   concentrations were approximated to TGM concentrations from July 2015 to April 2016."

516   **See the revised manuscript at line 109 – 115**

517   "In our research, random uncertainties of individual measurement had been averaged out and

518   the systematic uncertainties need to be considered. The overall practically achievable

519   systematic uncertainty would be 10% considering that the instrument was not in ideal

520   performance (Slemr et al., 2015; Steffen et al., 2012). For example, slow deactivation of the

521   traps, contamination of the switching valves and leaks would increase the uncertainties but were

522   difficult to quantify (Slemr et al., 2015;Steffen et al., 2012). Because of the consistency of

523 instrument and the quality assurance/quality control have been paid special attention to during

524 the sampling campaign, the systematic differences of instrument did not affect the huge

525 variation between 2014 and 2016."

526 **See the revised manuscript at line 124 – 131**

527

528 *Comment 1:*

529 Line 26: "GEM concentrations showed a significant decrease with a rate of -0.60 ng/m3/yr".

530 According to Table 1, the rate is -0.52 ng/m3/yr.

531 *Response:*

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

533 have corrected this in the revised manuscript.

534 .

535 *Comment 2:*

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

537 *Response:*

538 We have corrected in the manuscript as below.

539 "It was found that the reduction of domestic emissions was the main driver of GEM decline

540 in Chongming Island, accounting for 70% of the total decline."

541 **See the revised manuscript at line 33 – 34**

542

543 *Comment 3:*

544 Lines 47-48: "In the atmosphere, Hg mainly presents as GEM, accounting for over 95% or the

545 total". Can you please add a reference? Is that also true at your site?

546 *Response:*

547 We have added references in the manuscript. During the sampling period (March 2014 to

548 June 2015 and May 2016 to December 2016), the GOM concentration is 14.81 ± 13.21 pg m^{-3}

549 and GEM concentration is 2.15 ± 0.94 ng m^{-3}. Thus, the GOM concentration accounted for

550 0.68% and the conclusion in the reference is also true at our site. We also added reference in

551   our manuscript as below.

552   "In the atmosphere, Hg mainly presents as GEM, accounting for over 95% of the total in

553   most observation sites (Fu et al., 2015; Li et al., 2016; Zhang et al., 2017)."

554   **See the revised manuscript at line 49**

555

556   *Comment 4:*

557   Line 61-62: "(: : :) there is no official national monitoring network of atmospheric Hg". Out of

558   curiosity, what is the current status of the Asian-Pacific Mercury Monitoring Network

559   (http://nadp.sws.uiuc.edu/newIssues/asia/)? Do you think that Chinese sites will be included?

560   *Response:*

561   Thanks for the comment. Asian-Pacific Mercury Monitoring Network (APMMN) was

562   established in 2013 with founding countries and regions including the U.S, China Taiwan,

563   Thailand, Vietnam, Indonesia, Japan, Korea and Canada

564   (http://apmmn.org/AboutAPMMN.html). APMMN has been monitoring atmospheric mercury

565   deposition in the Asia-Pacific region and holds annual meetings since 2013. Currently, there is

566   no monitoring site of mainland China in the APMMN (see the Figure R2).

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

568　Considering the large Hg emissions in mainland China, including the Chinese sites into the

569　monitoring network will helpful for the research of Hg behavior in the regional or global scale.

570　However, nearly all mercury monitoring sites belong to individual researchers in China

571　currently. Therefore, whether the Chinese sites will be included mainly depend on multiple

572　factors such as individual interests and potential benefit. We also revised our expression in the

573　manuscript as below.

574　　"For the developing countries such as China, limited atmospheric Hg observations have been

575　carried out (Fu et al., 2008b; Zhang H et al., 2016; Hong et al., 2016) and there is no official

576　national monitoring network of atmospheric Hg in mainland China."

577　**See the revised manuscript at line 63 - 66**

578

579　***Comment 5:***

580　Lines 64-67: "Atmospheric Hg emissions in China accounted for 27% of the global total in

581　2010 (UNEP, 2013), which led to high air Hg concentrations in China. Therefore, atmospheric

582　Hg observations in China are critical to understand the Hg cycling at both regional and global

583　scale". Please define "high" air Hg concentrations. Additionally, in order to emphasize the fact

584　that observations in China are critical to understand the Hg cycling on a global scale, you could

585　perhaps add a sentence about 1) future projections (e.g., Chen et al., 2018; Pacyna et al., 2016),

586　and 2) long-range transport of Chinese emissions to other regions (e.g., Chen et al., 2018;

587　Corbitt et al., 2011; Sung et al., 2018).

588　***Response:***

589　　Hg concentration in remote site in China and Northern Hemisphere are compared to illustrate

590　the level of Hg pollution in China. Long-range transport and future projections are added to

591　emphasize observation in China are critical to understand Hg cycling on a global scale.

592　　The related paragraph is revised as below.

593　　"China contributes to the largest Hg emissions in the world and will continue to be one

594　significant Hg emitter for global Hg emissions in the coming future (UNEP, 2013, Wu et al.,

595　2016, Chen et al., 2018; Pacyna et al., 2016). Large Hg emissions in China have led to the

596　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

597   remote sites in China (Fu et al., 2015). Such Hg concentration level is approximately 1.3 ng m$^-$

598   ^{3} higher than the background concentration of GEM in Northern Hemisphere (Zhang et al.,

599   2016;Sprovieri et al., 2017;Fu et al., 2015). In addition, the large Hg emissions in China will

600   also impact the air Hg concentrations in East Asia and even North America through long-range

601   transport (Sung et al., 2018;Zhang et al., 2017)."

602   **See the revised manuscript at line 67 – 75.**

603

604   *Comment 6:*

605   Lines 93-94: "we used Tekran 2537X/1130/1135 instruments to monitor speciated Hg in the

606   atmosphere". I wonder why GOM and PBM concentrations are not reported in the manuscript.

607

608   *Response:*

609   We didn't report the GOM and PBM concentration because the main purpose of this

610   manuscript is to validate the anthropogenic Hg emissions reduction through observation data.

611   The Chongming site is a background site. Considering the stability of GEM in the air, we

612   choose GEM as an index to reflect the emission control effect. In addition, the method we

613   developed to explain the GEM trend is not applicable for GOM and PBM. Except emissions,

614   we think the potential reactions in the air are more significant factors for GOM and PBM. But

615   we need more evidence to prove our assumptions. Therefore, we deleted the discussion of GOM

616   and PBM in our final manuscript.

617

618   If concentrations were recorded, it would be interesting to discuss the results. Do you also see

619   a decreasing trend from 2014 to 2016? From 1978 to 2014, the fractions of GEM and PBM

620   decreased, while the GOM emission share gradually increased (Wu et al., 2016). What about

621   the speciation of emissions since 2014? Can you observe a trend in GOM/PBM concentrations?

622

623   *Response:*

624   Yes. The downward trend of PBM concentration was observed to decrease from 2014 (24.51

625     $\pm$43.31 pg m^{-3}) to 2016 (22.07 $\pm$30.55 pg m^{-3}), which was also consistent with the downwawrd

626   trend of GEM. However, the GOM concentration increased from (15.41 $\pm$ 16.02 pg m^{-3}) in

627   2014 to (18.97 $\pm$ 9.28 pg m^{-3}) in 2016. Speciated Hg emissions were showed in Table R1. All

628   speciated Hg emissions have decreased since 2014 in the YRD regions. However, we observed

629   significant GEM decreasing. But the decrease of GOM and PBM was quite slight.

630

631   **Table R1.** Speciated Hg emissions in YRD region and concentration at Chongming island in

632   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

633

634   Alternately, did you have issues with the speciation unit? It is quite common and I would

635   appreciate an open discussion about that and associated analytical uncertainties. What kind of

636   issues did you encounter? Are you confident that you collected and analyzed GEM (vs. TGM)

637   during the entire experiment? Was the instrumental setup exactly the same during the entire

638   experiment? If not, how can you compare GEM concentrations without discussing analytical

639   uncertainties? See major comment.

640

641   Yes, we encountered issues with the speciation unit. The Tekran 2537X was consistent and

642   in good condition during the sampling period. There was no data in January and February in

643   2016 due to equipment failure. The Tekran 1130/1135 was accidentally rained by rain, so there

644   was no data of speciated mercury between July 2015 and April 2016. From July 2015 to April

645   2016, we used Tekran 2537X only with PTFE filter to monitor atmospheric mercury. The

646   average concentration of GOM during sampling period (March, 2014 to June 2015, May 2016

647   to December 2016) was 14.81 $\pm$ 13.21 pg m^{-3}, which is approximately 1% of GEM

648   concentration. Thus, the GEM concentrations were approximated to TGM concentrations July

649   5, 2015 to April 30 2016 when the speciation unit does not work, as most of other studies have

650   done (Slemr et al., 2015; Sprovieri et al., 2016). In addition, we have added discussion about

651   the analytical uncertainties to point out potential impact.

652 The manuscript was revised as below.

653 "From July 5, 2015 to April 30 2016, the Tekran 1130/1135 speciation unit was damaged by

654 the rainstorm, the Tekran 2537X were operated without speciation units but with PTFE filter to

655 protect the instrument from particles and sea salt. Therefore, the observed concentrations during

656 July 2015-April 2016 were TGM concentrations indeed. However, the GOM concentrations at

657 Chongming Island accounted for less than 1% of TGM (TGM=GOM+GEM). Thus, the GEM

658 concentrations were approximated to TGM concentrations from July 2015 to April 2016."

659 **See the revised manuscript at line 109 – 115**

660 "In our research, random uncertainties of individual measurement had been averaged out and

661 the systematic uncertainties need to be considered. The overall practically achievable

662 systematic uncertainty would be 10% considering that the instrument was not in ideal

663 performance (Slemr et al., 2015; Steffen et al., 2012). For example, slow deactivation of the

664 traps, contamination of the switching valves and leaks would increase the uncertainties but were

665 difficult to quantify (Slemr et al., 2015;Steffen et al., 2012). Because of the consistency of

666 instrument and the quality assurance/quality control have been paid special attention to during

667 the sampling campaign, the systematic differences of instrument did not affect the huge

668 variation between 2014 and 2016."

669 **See the revised manuscript at line 124 – 131**

670

671 *Comment 7:*

672 Lines 103-104: "The impactor plates and quartz filter were changed in every two weeks. The

673 quartz filter was changed once a month". Did you change the quartz filter every two weeks or

674 once a month?

675 *Response:*

676 Yes, the impactor plates, Teflon filter and quartz filter were changed in every two weeks.

677 The soda lime was changed once a month. We have corrected this sentence in the revised

678 manuscript as below.

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

680 changed once a month."

681   **See the revised manuscript at line 120 – 121**

682

683   *Comment 8:*

684   Line 106: "During the sampling campaigns, $PM_{2.5}$, O_3, NO_x, CO and SO_2 were monitored".

685   Why aren't you discussing the data, especially SO_2, NO_x, $PM_{2.5}$ while your main conclusion is

686   that Hg decreasing trend in due to air pollution control policies targeting SO_2, NO_x, and $PM_{2.5}$.

687   I agree that you present emissions inventories, but I would really appreciate to see a real

688   interpretation and discussion of these data. Do you also observe a decreasing trend? See major

689   comment.

690   *Response:*

691   To further verify the cause of downward trend of atmospheric Hg, we give the emission

692   inventory (Table S6) and concentrations (Table S5) of other air pollutants in the studied regions

693   in both 2014 and 2016. Both the emissions and concentrations of SO_2, NO_2, and PM showed a

694   decreasing trend, which is used to support that "air pollution control policies targeting SO_2,

695   NO_2, and PM reductions had significant co-benefits on atmospheric Hg".

696   "Table 3 showed the detailed data of the three classifications. From 2014 to 2016, the whole

697   China region (NCP, SW-YRD) contributed to 70% of GEM decline at Chongming Island.

698   Considering downward trend of emission inventory and atmospheric pollutant from 2014 to

699   2016 in NCP and SW-YRD region (Table S5, Table S6), the reason of downward trend can be

700   attributed to the effectiveness of existing air pollution control measures in China (SC, 2013;

701   MEP, 2014)."

702   **See the revised manuscript at line 380 – 384**

703

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

705   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%

(µg m⁻³)									
NO$_2$ (µg m⁻³)	45.07	34.34	12.62	41.55	34.40	10.84	-8%	0%	-14%
O$_3$ (µg 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%	

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

709

710 **Table S6.** Emission inventories of the main pollutants from the studied regions in 2014 and
711 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%

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

715

716 *Comment 9:*

717 Lines 173-175: "Besides, this method required similar meteorological conditions of the periods

718 participated in comparison so as to reduce the interference from meteorology". I am not sure I

719 understand this sentence. Do you mean that you used similar meteorological data in 2014 and

720 2016 to compute the back-trajectories? Or are you referring to the fact that meteorological

721 conditions were pretty much similar in 2014 and 2016 (lines 266-274)?

722 *Response:*

723 Thanks for the comments. Yes, this sentence is referring to the fact that meteorological

724 conditions were pretty similar in 2014 and 2016. We have revised the sentence as suggested to

725 make it easier to understand.

726 "Besides, meteorological conditions were pretty similar in 2014 and 2016 so as to reduce the

727 interference from meteorology (Table S2).

728 **See the revised manuscript at line 203 – 204**

729

730   *Comment 10:*

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

732   *Response:*

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

734   "The emission sectors included coal-fired power plants, coal-fired industrial boilers,

735   residential coal-combustion, cement clinker production, iron and steel production, mobile oil

736   combustion, and other small emission sectors (eg., zinc smelting, lead smelting, municipal solid

737   incineration, copper smelting, aluminum production, gold production, other coal combustion,

738   stationary oil combustion, and cremation)."

739   **See the revised manuscript at line 212-216.**

740

741   *Comment 11:*

742   Lines 193-194: "The average concentrations of GEM in 2014 and 2016 were (: : :)". What about

743   the mean concentration in 2015? Additionally, are the average annual concentrations actually

744   referring to March-December? If so, please add something like "The average concentrations of

745   GEM in 2014 (Mar-Dec) and 2016 (Mar-Dec) were (: : :)".

746   *Response:*

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

748   concentration of 2015 and remark in the revised manuscript.

749   "The average concentrations of GEM in 2014 (March to December), 2015 and 2016 (March

750   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."

751   **See the revised manuscript at line 230 – 231**

752

753   *Comment 12:*

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

755   *Response:*

756   *t* test was used to compare the GEM concentration at Chongming and background

757   concentration of Northern Hemisphere. The *p* value ($p < 0.01$) of the *t* test were added in the

758   revised manuscript as below.

759 "The GEM concentrations in 2014 (2.68 ± 1.07 ng m^{-3}) were higher (t test, $p<0.01$) than the

760 Northern Hemisphere back-ground concentration (about 1.5 ng m^{-3}) (Sprovieri et al., 2010) and

761 those measured in other remote and rural locations in China (Zhang H et al., 2015; Fu et al.,

762 2008a; Fu et al., 2009)."

763 **See the revised manuscript, line 231 – 234**

764

765 *Comment 13:*

766 Lines 199-200: "During this period, monthly GEM concentrations showed a significant

767 decrease with a rate of -0.60 ng/m3/yr". Table 1 refers to TGM concentrations, not GEM.

768 Additionally, as mentioned earlier, the rate is -0.52 ng/m3/yr in Table 1. Please, try to be

769 consistent throughout the manuscript.

770 *Response:*

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

772 the manuscript consistent.

773

774 *Comment 14:*

775 Lines 201-216: To me, "GEM" and "TGM" are not interchangeable (see previous comment).

776 While the difference between TGM and GEM is usually smaller than 1% (Soerensen et al.,

777 2010), it might not be the case everywhere. What is the fraction of GOM at your site? I would

778 appreciate a discussion on analytical uncertainties and instrumental setups.

779 *Response:*

780 We agree that the GEM and TGM are not always interchangeable. The average concentration

781 of GOM during sampling period was 14.81 ± 13.21 pg m^{-3}, which was less than 1% of TGM.

782 Thus, the GEM concentrations were approximated to TGM concentrations July 5, 2015 to April

783 30 2016 when the speciation unit does not work, as most of other studies have done (Slemr et

784 al., 2015; Sprovieri et al., 2016). We have pointed out this in the revised manuscript.

785 A discussion on analytical uncertainties and instrumental setups has been added in the following

786 text as below.

787 "From July 5, 2015 to April 30 2016, the Tekran 1130/1135 speciation unit was damaged by

788 the rainstorm, the Tekran 2537X were operated without speciation units but with PTFE filter to

789 protect the instrument from particles and sea salt. Therefore, the observed concentrations during

790 July 2015-April 2016 were TGM concentrations indeed. However, the GOM concentrations at

791 Chongming Island accounted for less than 1% of TGM (TGM=GOM+GEM). Thus, the GEM

792 concentrations were approximated to TGM concentrations from July 2015 to April 2016."

793 **See the revised manuscript at line 109 – 115**

794 "In our research, random uncertainties of individual measurement had been averaged out and

795 the systematic uncertainties need to be considered. The overall practically achievable

796 systematic uncertainty would be 10% considering that the instrument was not in ideal

797 performance (Slemr et al., 2015; Steffen et al., 2012). For example, slow deactivation of the

798 traps, contamination of the switching valves and leaks would increase the uncertainties but were

799 difficult to quantify (Slemr et al., 2015;Steffen et al., 2012). Because of the consistency of

800 instrument and the quality assurance/quality control have been paid special attention to during

801 the sampling campaign, the systematic differences of instrument did not affect the huge

802 variation between 2014 and 2016."

803 **See the revised manuscript at line 124 – 131**

804

805 The sentence "at the Cape Point of South Africa, GEM concentrations decreased from 1.35

806 ng/m3 in 1996 to 0.9 ng/m3 in 2008" is not entirely true. A downward trend has been observed

807 from 1996 to 2005, while an upward one is observed since 2007 (Martin et al., 2017; Slemr et

808 al., 2015).

809 *Response:*

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

811 "In South Africa, annual average GEM concentration at Cape Point decreased from 1.29 ng m^{-3}

812 in 1996 to 1.19 ng m^{-3} in 2004 (Slemr et al., 2008) and were increasing from 0.93 ng m^{-3} in

813 2007 (Slemr et al., 2015) until 2016 (Martin et al, 2017)."

814

815 Additionally, the instrumental setup changed: a manual amalgamation technique was used from

816 1995 to 2004 while a Tekran instrument has been used since 2007 (Martin et al., 2017). It might

817 also be the case at other stations in Table 1. How does it influence the various trends reported

818 in Table 1?

Response:

820   Table 1 was moved to supporting information (Table S4) so as to focus on our topic. In the

821   Table S4, all the stations used Tekran instruments except for the monitoring in South Korea.

822   The instrument of Canadian sites were maintended by the Environment Canada- developed

823   Research Data Management and Quality Assurance System (RDMQ). At Zeppelin, the

824   instruement maintenance were carried out by the protocols of Norwegian mercury monitoring

825   program. Instruments at these plances have been maintended under the guidance of similar

826   quality control criteria. In South Korea, the concentration of TGM were measured using an

827   automatic on-line analytical system called a AM-series analyzer. Although the intruments used

828   in the stations listed in the Table 1 were not totally the same, the instruments at each site

829   remained unchanged during the monitoring period. Therefore, the downward trend at different

830   sites can be compared in the Table S4. We have revised the Table S4 and give expression about

831   the different instrument in the revised manusript.

832   "All the stations in Table S4 used Tekran instruments except for the observation in South

833   Korea. Different instruments could cause potential differences in the observation, but they were

834   comparable and did not affect the conclusion of comparison in downward trend (Slemr et al.,

835   2015; Sprovieri et al., 2016)."

836   **See the revised manuscript at line 253 - 256**

837   **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

838

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 266 - 271

Comment 16:

Line 224: Are you referring to Figure 2?

Response:

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

860

861   *Comment 17:*

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

863   *Response:*

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

865   below.

866   "According to the decomposition result (Figure 2c), we observed strong seasonal cycle with

867   seasonal GEM peak in July and trough in September, so GEM concentrations in the same month

868   but different years were averaged to discuss the seasonal circle (Figure 3)."

869   **See the revised manuscript at line 274 - 276**

870

871   *Comment 18:*

872   Line 234: "The higher Hg concentrations in cold seasons in Mt. Ailao and Mt. Waliguan (: : :)".

873   You say above that concentrations are lower in the cold season at these sites. This is confusing.

874   *Response:*

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

876   "The higher Hg concentrations in cold seasons in Mt. Leigong were mainly explained by coal-

877   combustion for urban and residential heating during cold seasons. Whereas, increasing solar

878   radiation and soil/air temperature dominate the higher Hg concentrations in Mt. Ailao."

879   **See the revised manuscript at line 288- 291**

880

881   *Comment 19:*

882   Line 250-251: "Therefore, we supposed that the seasonal cycle of GEM concentrations was

883   dominated by natural emissions". How can you explain that the seasonal cycle is more

884   pronounced in 2014 than in 2016? See major comment.

885   *Response:*

886   The seasonal variation was more pronounced in 2014 can attribute to the lower wet

887   deposition and GEM oxidation. On one aspect, as a costal site, the Chongming island is

888   abundant with •OH. The increase of O_3 concentration from the summer of 2014 to 2016 may

889 contribute to a higher oxidation of GEM in 2016. On another aspect, and higher wet Hg

890 deposition is approximately 6.6 times of that in the winter at Chongming (Zhang et al., 2010).

891 Meanwhile, the rainfall in 2016 summer (546 mm) was higher than the rainfall in 2014 (426

892 mm). Therefore, the higher oxidation and wet deposition rate of mercury in the summer of 2016

893 will reduce the concentration difference between summer and winter, which lead to a less

894 pronounced seasonal variation in 2016.

895

896

897 **Figure 2.** Monthly average GEM concentrations during the studied period (a) observed monthly

898 GEM concentrations (b) GEM trend after decomposition (c) GEM seasonality after

899 decomposition (d) GEM random after decomposition

900 Note: The observed concentrations during July 2015-April 2016 were TGM concentrations

901 indeed due to the problems of Tekran 1130/1135. However, the GOM concentrations at

902 Chongming island accounted for less than 1% of TGM. Thus, the GEM concentrations were

903 approximated to TGM concentrations during July 2015-April 2016.

904 "From Figure 2, we also observed more pronounced seasonal variation in 2014, which can

905 be attributed to the lower wet deposition and GEM oxidation. On one aspect, as a costal site,

906 the Chongming Island is abundant with •OH. The increase of O_3 concentration from the summer

907 of 2014 to 2016 may contribute to a higher oxidation of GEM in 2016. On another aspect, and

908 higher wet Hg deposition is approximately 6.6 times of that in the winter at Chongming (Zhang

909 et al., 2010). Meanwhile, the rainfall in 2016 summer (546 mm) was higher than the rainfall in

910 2014 (426 mm). Therefore, the higher oxidation and wet deposition rate of Hg in the summer

911 of 2016 will reduce the concentration difference between summer and winter, which lead to a

912 less pronounced seasonal variation in 2016. Meanwhile, the higher oxidation and wet

913 deposition in 2016 also contributed to the downward trend of GEM by reducing the seasonality

914 in spring and summer (Figure S3)."

915 **See the revised manuscript at line 318 - 328**

916

917 ***Comment 20:***

918 Lines 275-276: "This decline may be contributed by the downward trend of GEM

919 concentrations in north hemisphere". Please, elaborate on this idea. I don't really understand

920 what you mean here.

921 ***Response:***

922   Sorry for the obscure expression. The decline of PSCF value means that East China Sea has

923 less contribution to Chongming in 2016. The potential reason of the decline on PSCF value in

924 the East China Sea may be attributed to the reduction of emissions in Japan and Korea. The

925 downward trend in Japan and Korea will lead to clean air mass transport from Japan and Koran

926 to East China Sea and then to Chongming. We have revised the manuscript as below:

927   "The decline from the East China Sea may be contributed by the downward trend of GEM

928 concentrations in South Korea and Japan (Kim et al., 2016; Kim et al., 2013), where the

929 anthropogenic Hg emissions of Japan and South Korea have been reduced by 13% and 4%

930 during 2010-2015, respectively (UNEP 2013; UNEP 2018). The air mass from Japan and South

931 Korea would pass through the East China Sea to Chongming."

932   **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)

933 **See the revised the manuscript at line 336-340**

934

935 *Comment 21:*

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

937 concentrations?

938 *Response:*

939   The SO_2, NO_x and $PM_{2.5}$ concentrations at Chongming island also show downward trend.

940 However, such kind of analysis is not so suitable for SO2, NOx, and $PM_{2.5}$ due to the following

941 reasons. First, the residential time of SO_2, NOx, and $PM_{2.5}$ is 2-4 d, 8-10d, and several days to

942 few weeks, respectively (Pirrone, et al., 1996, Seinfeld, Spyros, 2016). Such residential time is

943 much shorter than that for Hg^0. Second, SO2, NO_x and $PM_{2.5}$ are more reactive in the

944 atmosphere compared with Hg^0 (Pirrone, et al., 1996, Seinfeld, Spyros, 2016).

945

946 *Comment 22:*

947 Line 318: 34% should be 35% according to Table 4. Additionally, how can you explain this

948 result? Is there a decline in anthropogenic emissions and a GEM decreasing trend in this region

949 (China Sea, Japan, South Korea) as well? Cluster EAST explains 35% of the decline, i.e., 0.35

950 x 0.52 = 0.182 ng/m3/yr. Is that consistent with trends reported in this region (e.g., Kim et al.,

951 2016)?

952 *Response:*

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

954   However, we have changed the definition of cluster according to the suggestion. The NCP

955 region, SW-YRD region, and ABROAD region causes 26%, 44%, and 30% for GEM decline,

956 respectively. The whole China region (NCP, SW-YRD) contributed to 70% of GEM decline at

957 Chongming Island while ABROAD region contributed to 30%. The decline in NCP and SW-

958 YRD indicated effective air pollution control policy in China since 2013. The decline in

959 ABROAD region was originated from GEM decline in South Korea and Japan.

960   The decline in Chongming was consistent with the decline in anthropogenic emission and a

961 GEM decreasing trend in the ABROAD region. In South Korea, the decline of GEM at Seoul

962 can be calculated as 0.47 ng m^{-3} yr^{-1} from 2011 to 2013 (Kim, et al., 2016, Kim, et al., 2013).

963 In Japan, there is no published data about long term trend since 2010. But the emission

964 inventory of Japan decreased from 2010 to 2015 (Table R2). Therefore, the decline in

965   ABROAD can be attributed to the decline in South Korea and Japan.

966   **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)

967

968   We also revised the manuscript as below.

969 "The decline from the East China Sea may be contributed by the downward trend of GEM

970 concentrations in South Korea and Japan (Kim et al., 2016; Kim et al., 2013), where the

971 anthropogenic Hg emissions of Japan and South Korea have been reduced by 13% and 4%

972 during 2010-2015, respectively (UNEP 2013; UNEP 2018). The air mass from Japan and South

973 Korea would pass through the East China Sea to Chongming."

974 **See the revised manuscript at line 336 -340**

975

976 *Comment 23:*

977 Lines 321-323: "We also noted that the largest decline of Hg concentrations was observed in

978 the cluster SW, which indicated more effective air pollution control in the regions where the air

979 mass of the cluster SW passed". What about the seasonality of GEM concentrations in the

980 various clusters (NW, SW and EAST)? Could a difference in seasonality explain the observed

981 Hg decline?

982 *Response:*

983   The seasonality of GEM concentration in the various clusters was showed in Figure S3. In

984 our revised manuscript, cluster NW, SW and EAST were modified to cluster NCP, SW-YRD

985 and ABROAD. The seasonality of cluster NCP, SW-YRD and ABROAD were similar to the

986 seasonality at Chongming. The GEM concentration of different clusters reached the highest in

987 the summer of 2014. And the seasonality in 2014 for the three clusters was more pronounced

988 than their seasonality in 2016.

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

990 declined in all season. In 2014, the seasonality was more pronounced than the seasonality in

991 2016. It can be attributed to the higher oxidation of GEM and higher wet deposition in 2016.

992 The smaller seasonal variation also had an effect on the decline. We revised our expression in

993 the revised manuscript.

994 "Therefore, the higher oxidation and wet deposition rate of mercury in the summer of 2016

995 will reduce the concentration difference between summer and winter, which lead to a less

996 pronounced seasonal variation in 2016. Meanwhile, the higher oxidation and wet deposition in

997 2016 also contributed to the downward trend by reducing the seasonality of spring and summer

998 (Figure S3)."

999 **See the revised manuscript in line 324 – 328.**

1000

1001 **Figure S3.** The seasonality of GEM concentration in the NCP, SW-YRD and ABROAD (No

1002 trajectory transport though ABROAD in winter of 2014)

1003 **See the revised manuscript at Figure S3.**

1004

1005 *Comment 24:*

1006 Figure 3: Could you please add the standard deviations? Is that the average over several years?

1007 *Response:*

1008 We have revised as suggested. It is the average in the three years. We also give expression in

1009 the manuscript as below.

1010   "According to the decomposition result (Figure 2c), we observed strong seasonal cycle with

1011   seasonal GEM peak in July and trough in September, so GEM concentrations in the same month

1012   but different years were averaged to discuss the seasonal circle (Figure 3). The average data

1013   can eliminate the effect of downward trend and get result of average seasonal variation. The

1014   error bars in the Figure 3 mean the standard deviation of the monthly average."

1015   **See the revised manuscript in line 274– 278.**

1016

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1079

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

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Abstract

1100

1101   Measurements of gaseous elemental Hg (GEM), other air pollutants including SO_2, NO_X, O_3,

1102 $PM_{2.5}$, CO, and meteorological conditions were carried out at Chongming Island in East China from

1103   March 1 in 2014 to December 31 in 2016. During the sampling period, GEM concentrations

1104   significantly decreased from 2.68 ± 1.07 ng m^{-3} in 2014 (March to December) to 1.60 ± 0.56 ng m^{-3}

1105   in 2016 (March to December). Monthly mean GEM concentrations showed a significant decrease

1106   with a rate of -0.60 ± 0.08ng m^{-3} yr^{-1} (R^2=0.64, p<0.01 significance level). Combining the analysis

1107   of potential source contribution function (PSCF), principle component analysis (PCA), and

1108   emission inventory, we found that Yangtze River Delta (YRD) region was the dominant source

1109   region of GEM in Chongming Island and the main source industries included coal-fired power

1110   plants, coal-fired industrial boilers, and cement clinker production. We further quantified the effect

1111   of emission change on the air Hg concentration variations at Chongming Island through a coupled

1112   method of trajectory clusters and air Hg concentrations. It was found that the reduction of domestic

1113   emissions was the main driver of GEM decline in Chongming Island, accounting for 70% of the

1114   total decline. The results indicated that air pollution control policies targeting SO_2, NO_x and

1115   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

1145 observation records of China's air Hg concentrations published (Fu et al., 2015).

1146 China contributes to the largest Hg emissions in the world and will continue to be one significant

1147 Hg emitter for global Hg emissions in the coming future (UNEP, 2013, Wu et al., 2016, Chen et al.,

1148 2018; Pacyna et al., 2016). Large Hg emissions in China have led to the average air Hg

1149 concentrations of 2.86 $\pm$0.95 ng m^{-3} (in the range of 1.60-5.07 ng m^{-3}) at the remote sites in China

1150 (Fu et al., 2015). Such Hg concentration level is approximately 1.3 ng m^{-3} higher than the

1151 background concentration of GEM in Northern Hemisphere (Zhang et al., 2016;Sprovieri et al.,

1152 2017;Fu et al., 2015). In addition, the large Hg emissions in China will also impact the air Hg

1153 concentrations in East Asia and even North America through long-range transport (Sung et al.,

1154 2018;Zhang et al., 2017). Meanwhile, China has a great potential for Hg emission reduction during

1155 implementation of the *Minamata Convention on Mercury* (Chen et al., 2018). Therefore, long-term

1156 atmospheric Hg observations in China are critical to understand the Hg cycling at both regional and

1157 global scale. China's Hg emissions had increased from 147 t yr^{-1} in 1978 to around 538 t yr^{-1} in 2010

1158 due to the dramatic economic development (Zhang L et al., 2015; Wu et al., 2016; Hui et al., 2017).

1159 Atmospheric Hg monitoring that spanned the longest periods (from 2002 to 2010) in Guiyang,

1160 southwestern China witnessed the increase of Hg emissions in China (Fu et al., 2011). However,

1161 recently atmospheric Hg emissions in China have been estimated to decrease since 2012 (Wu et al.,

1162 2016). This decreasing trend needs to be confirmed by atmospheric Hg observations.

1163 In this study, we measured GEM, other air pollutants (eg., PM$_{2.5}$ and NO$_x$), and meteorological

1164 parameters (eg., temperature and wind speed) at a remote marine site of Chongming Island in East

1165 China during 2014-2016. We analyzed annual and seasonal variation of GEM and the potential

1166 impact factors. Combining the analysis of potential source contribution function (PSCF), principle

1167 component analysis (PCA), and emission inventory, the potential source regions and source

1168 industries of atmospheric Hg pollution at the monitoring site are identified. In addition, a coupled

1169 trajectories and air Hg concentration method is developed to assess the effect of Hg emission change

1170 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

1200 were recoated once every two weeks following the procedure developed by Landis et al. (2002).

1201 In our research, random uncertainties of individual measurement had been averaged out and the

1202 systematic uncertainties need to be considered. The overall practically achievable systematic

1203 uncertainty would be 10% considering that the instrument was not in ideal performance (Slemr et

1204 al., 2015; Steffen et al., 2012). For example, slow deactivation of the traps, contamination of the

1205 switching valves and leaks would increase the uncertainties but were difficult to quantify (Slemr et

1206 al., 2015;Steffen et al., 2012). Because of the consistency of instrument and the quality

1207 assurance/quality control have been paid special attention to during the sampling campaign, the

1208 systematic differences of instrument did not affect the huge variation between 2014 and 2016.

1209 During the sampling campaigns, $PM_{2.5}$, O_3, NO_x, CO and SO_2 were also monitored by Thermo

1210 Scientific TEOM 1405D, Model 49i O_3 Analyzer, Model 48i CO Analyzer, Model 42i-TL NOx

1211 Analyzer and Model 43i SO_2 Analyzer, respectively. The detection limits of O_3, SO_2, NO_x, CO and

1212 $PM_{2.5}$ are 1.0, 0.5, 0.4, 0.04 and 0.1 $\mu g\ m^{-3}$, respectively. The meteorological parameters including

1213 air temperature, wind speed, and wind direction are measured by Vantage Pro2 weather station

1214 (Davis Instruments). The instruments are tested and calibrated periodically. All data are hourly

1215 averaged in this study.

1216 **2.3 Sources apportionment of atmospheric Hg pollution**

1217 2.3.1 PSCF model

1218 To identify the source areas for pollutants with a relatively long lifetime such as GEM (Xu and

1219 Akhtar, 2010), the PSCF values for mean GEM concentrations in grid cells in a study domain are

1220 calculated by counting the trajectory segment endpoints that terminate within each cell. The number

1221 of endpoints that fall in the ij-th cell are designated n_{ij}. The number of endpoints for the same cell

1222 having arrival times at the monitoring site corresponding to GEM concentrations higher than a

1223 specific criterion is defined to be m_{ij}. The criterion in this study is set as the average Hg concentration

1224 during our study period. The PSCF value for the ij-th cell is then defined as:

$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}} W_{ij} \tag{1}$$

1226 where W_{ij} is an empirical weight to reduce the effects of grid cells with small n_{ij} values. In this

1227 study, W_{ij} is defined as in the following formula, in which Avg is the mean n_{ij} of all grid cells with

1228 n_{ij} greater than zero:

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

1229

1230   The PSCF value indicates the probability of a grid cell through which polluted events occurs.

1231   More method details can be found in the study of Polissar et al. (Polissar et al., 1999). In this study,

1232   the domain that covered the potential contribution source region (105 °–135 °E, 15 °–45 °N) was

1233   divided into 22500 grid cells with 0.2 °×0.2 °resolution. 72-hour back trajectories were generated

1234   hourly from 1 March, 2014 to 31 December, 2015 and from March 26 to December 31 in 2016 by

1235   TrajStat, a software including HYSPLIT for trajectory calculation with trajectory statistics modules

1236   (Wang et al., 2009). PSCF map was plotted using ArcGIS version 10.1.

1237   2.3.2 Principal component analysis (PCA)

1238   Correlation between Hg and other pollutant concentrations are used to identify source industries.

1239   Strong positive loadings (loading>0.40) with SO_2 and $PM_{2.5}$ typically indicate the impact of coal

1240   combustion, and strong positive loadings with GEM and CO have often been used as an indicator

1241   for regional transport because both pollutants have similar source and stable chemical properties

1242   (Lin et al., 2006; Pirrone et al., 1996). In this study, PCA was applied to infer the possible influencing

1243   factors of GEM in 2014 and 2016. Prior to analysis, each variable was normalized by dividing its

1244   mean, and pollutant concentrations (SO_2, CO, NO_X, $PM_{2.5}$) were averaged to 1-h sampling intervals

1245   to match the hourly Hg monitoring during sampling period. The results in 2016 had no CO data due

1246   to instrument broken. Statistics analyses were carried out by using SPSS 19.0 software.

1247   **2.4 Quantification method of source contribution**

1248   To further quantitatively assess the effect of change in emissions from different regions on air

1249   concentrations variation at a certain monitoring site, a quantitative estimation method which coupled

1250   trajectories with air Hg concentrations was developed. We firstly identified the trajectories by using

1251   the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single-Particle Lagrangian

1252   Integrated Trajectory (HYSPLIT) model. The gridded meteorological data at a horizontal resolution

1253   of 1 °×1 °were obtained from the Global Data Assimilation System (GDAS) (Draxler and Hess,

1254   1998). The starting heights were set to be 500 m above ground level to represent the center height

1255   of boundary layer where pollutants are usually well mixed in boundary layer. Secondly, each

1256   trajectory was assigned with GEM concentration by matching the arriving time in Chongming site.

1257 Third, the backward trajectories which coupled with Hg concentrations were clustered into groups

1258 according to transport patterns by using NOAA HYSPLIT 4.7. Thus, the grouped clusters were

1259 applied to identify the Hg source regions. The Hg average concentration of the cluster j was then

1260 calculated as equation (3). And, the trajectory weighted concentration in the cluster j as equation

1261 (4). At last, the contribution of reduction at a certain region on Hg concentration at monitoring sites

1262 in a certain period can be calculated as equation (5).

1263

1264
$$C_{j,t} = \frac{\sum_{i=1}^{n} C_{i,j,t}}{\sum_{i=1}^{n} N_{i,j,t}} \tag{3}$$

1265
$$TWC_{j,t} = AR \times C_{j,t} \tag{4}$$

1266 where N refers to a certain trajectory. j refers to a certain cluster. t is the studied period, and n is

1267 the number of trajectory. m is the number of cluster. C is the GEM concentration, ng m^{-3}. TWC refers

1268 to the trajectory weighted concentration, ng m^{-3}. In order to reduce the influence of trajectory

1269 changes in different region between calculated years, the average ratio (AR) was used here for

1270 calculating TWC.

1271
$$CR_j = \frac{TWC_{j,t_2} - TWC_{j,t_1}}{\sum_{j=1}^{m} TWC_{j,t_2} - \sum_{j=1}^{m} TWC_{j,t_1}} \tag{5}$$

1272 where CR refers to the contribution of GEM reduction. t_1 and t_2 refers to the two period

1273 participating to comparison, namely year 2014 and 2016 in this study, respectively.

1274 This approach is a simple method to quantify the influence of anthropogenic emissions on GEM

1275 concentration variation. It should be noted that uncertainties always exist in calculating trajectories,

1276 causing uncertainties in all trajectory-based approaches. Trajectory errors vary considerably in

1277 different situation. Draxler (1996) suggested uncertainties might be 10% of the travel distance.

1278 Besides, meteorological conditions were pretty similar in 2014 and 2016 so as to reduce the

1279 interference from meteorology (Table S2).

1280 **2.5 Regional atmospheric Hg emissions**

1281 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.

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

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 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). However, in

1310   2016, the GEM concentrations were similar to the background concentrations in the Northern

1311   Hemisphere. During this period, monthly GEM concentrations showed a significant decrease with

1312   a rate of -0.60 $\pm$ 0.08 ng m^{-3} yr^{-1} (R^2=0.64, p<0.01 significance level, n = 32) (Figure 2a). The

1313   amount of valid data for each mouth was shown in Table S3. From another aspect, the trend

1314   decomposition of the GEM concentration signal (signal = trend + seasonal + random) from March

1315   2014 to December 2016 were performed in Figure 2 (https://anomaly.io/seasonal-trend-

1316   decomposition-in-r/). By using this method, we also observed a pronounced trend (Figure 2b) and

1317   the random was limited in the range of -0.24 – 0.24 ng m^{-3} (Figure 2d).

1318   One potential worry is that the calculated trend will be sensitive to seasonal variation and the

1319   missing data in January and February of 2016 may impact the downward trend. To evaluate the

1320   impact of the missing data, we estimate the Hg concentrations in the missing months based on the

1321   data of the same months in 2015 and 2017 (Figure S1). Combining the estimated data, we re-fit the

1322   Hg concentrations and downward trend still maintained robust and similar to the downward trend

1323   in manuscript (Figure S1). Thus, we assume that the missing data is not very important and will not

1324   impact our main conclusion.

1325   Table S4 showed the Hg variation trends in different regions. Significant decreases of GEM

1326   concentrations in North hemisphere over the past two decades have been well documented (Weigelt

1327   et al., 2015; Cole et al., 2013; Kim et al., 2016). All the stations in Table S4 used Tekran instruments

1328   except for the observation in South Korea. Different instruments could cause potential differences

1329   in the observation, but they were comparable and did not affect the conclusion of comparison in

1330   downward trend (Slemr et al., 2015; Sprovieri et al., 2016). Weigelt et al. (2015) showed that GEM

1331   concentrations decreased from 1.75 ng m^{-3} in 1996 to 1.4 ng m^{-3} in 2009 at Mace Head, Europe.

1332   Ten-year trends of GEM concentrations at six ground-based sites in the Arctic and Canada also

1333   showed a decreasing trend at a rate of 13-35 pg m^{-3} y^{-1} (Cole et al., 2013). In South Korea, the

1334   observed GEM concentration also had significant decrease in recent years (Kim et al., 2016). In

1335   South Africa, annual average GEM concentration at Cape Point decreased from 1.29 ng m^{-3} in

1336   1996 to 1.19 ng m^{-3} in 2004 (Slemr et al., 2008) and were increasing from 0.93 ng m^{-3} in 2007

1337   (Slemr et al., 2015) until 2016 (Martin et al, 2017). However, limited GEM monitoring sites and

1338   relative short-time spans in China restricted the views of long-term trends in atmospheric Hg

1339   concentration in this region. A preliminary assessment indicated that atmospheric Hg concentrations

1340   in China kept increasing before 2012 (Fu et al., 2015). The decreasing trend observed in our study

1341   was accordant with reported data in Mt. Changbai during 2014-2015 cited in the review of Fu et al.

1342   (2015). The atmospheric Hg at Chongming was influenced by and in turn reflected regional Hg

1343   emission and cycle. Although the decline in atmospheric Hg was observed in many sites of the

1344   Northern Hemisphere, much sharper decrease of Hg concentrations was observed at Chongming in

1345   our study. The specific reasons for the Hg concentration decrease in our study will be discussed in

1346   section 3.4.

3.2 Seasonal variation of GEM concentrations

1348   According to the decomposition result (Figure 2c), we observed strong seasonal cycle with

1349   seasonal GEM peak in July and trough in September, so GEM concentrations in the same month

1350   but different years were averaged to discuss the seasonal circle (Figure 3). The average data can

1351   eliminate the effect of downward trend and get result of average seasonal variation. The error bars

1352   in the Figure 3 mean the standard deviation of the monthly average. Observed GEM concentrations

1353   showed an obvious seasonal cycle. The mean GEM concentration in warm season (from April to

1354   September) is 0.29 ng m^{-3} higher than that in cold season. Such seasonal variation trend is also

1355   observed at Nanjing, Miyun, Mt. Ailao, Mt. Waliguan, and Shangri-La (Zhang et al., 2013; Zhang

1356   et al., 2016; Fu et al., 2015; Zhu et al., 2012). On the other hand, the means of GEM at Mt. Gongga,

1357   Mt. Daimei, Mt.Leigong, and Mt. Changbai in China are relatively higher in cold seasons. The

1358   average of atmospheric Hg concentrations in the north hemisphere also have a trough value in

1359   summer (Sprovieri et al., 2016).

1360   Seasonal variations of GEM concentration are generally attributed to the following factors,

1361   including natural and anthropogenic emissions, atmospheric chemical reaction, and air mass

1362   transportation. The higher Hg concentrations in cold seasons in Mt. Leigong were mainly

1363   explained by coal-combustion for urban and residential heating during cold seasons. Whereas,

1364   increasing solar radiation and soil/air temperature dominate the higher Hg concentrations in Mt.

1365   Ailao. In addition, sites in southern, eastern, and northeastern China also impacted from

1366   anthropogenic emissions of GEM from the north and west by the northerly winter monsoon while

1367   the sites located in western, southwestern, and northern China were impacted in the warm season

1368   (Fu et al., 2015). As to most sites in the northern hemisphere, high wet Hg precipitation induced

1369   probably by faster GEM oxidation led to lower Hg concentrations in summer.

1370 Source emission is one significant factor on GEM concentrations in the air. The GEM

1371 concentrations at a remote site are generally regarded under the impact of regional emissions.

1372 Therefore, the emissions in the YRD regions (Anhui, Zhejiang, Jiangsu, and Shanghai) were

1373 calculated. However, the anthropogenic emissions were in the range of 2.5-2.7 t, which is almost

1374 unchanged. Compared to the anthropogenic emissions, we observed almost synchronized trends

1375 between natural emissions and air Hg concentrations in Figure 4. The natural emissions showed a

1376 huge seasonal variation, from -5.4 t to 8.4 t. The largest natural emissions were observed in summer

1377 when the highest GEM concentrations were monitored. In the autumn, the natural emissions

1378 performed as the largest deposition direction amount and the GEM concentrations were the lowest

1379 in the whole year. Therefore, natural emissions instead of anthropogenic were supposed to be one

1380 significant factor of the seasonal cycle of GEM concentrations (Figure 4). The seasonal trend of

1381 natural emissions is closely related with the canopy types in YRD areas, where widely subtropical

1382 forests, paddy field, and dry farming were observed (Figure S2). The high temperature will speed

1383 up decomposition of organic compound in soil, which leads to Hg emissions from farmland and

1384 forest in YRD region (Luo et al., 2016; Yu et al., 2017). In autumn and winter, with the decrease of

1385 temperature (Table S2), the role of soil changed from Hg source to sink, which reduces the Hg

1386 concentrations in the air (Wang et al., 2016). At the same time, the growing vegetation in autumn

1387 also absorbs air Hg, resulting lower Hg concentrations compared to that in winter. Transport also

1388 overall enhanced the observed seasonal variation of GEM concentrations at Chongming Island.

1389 According to the statistics of backward trajectories in section 3.4, the GEM concentrations in the

1390 air mass which did not pass via the YRD regions also showed high GEM concentration in warm

1391 season in 2014 (Figure S3).

1392 From Figure 2, we also observed more pronounced seasonal variation in 2014, which can be

1393 attributed to the lower wet deposition and GEM oxidation. On one aspect, as a costal site, the

1394 Chongming Island is abundant with •OH. The increase of O_3 concentration from the summer of

1395 2014 to 2016 may contribute to a higher oxidation of GEM in 2016. On another aspect, and higher

1396 wet Hg deposition is approximately 6.6 times of that in the winter at Chongming (Zhang et al.,

1397 2010). Meanwhile, the rainfall in 2016 summer (546 mm) was higher than the rainfall in 2014 (426

1398 mm). Therefore, the higher oxidation and wet deposition rate of Hg in the summer of 2016 will

1399 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 invasion 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 °C, 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.

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1489   *Competing interests*. The authors declare that they have no conflict of interest.

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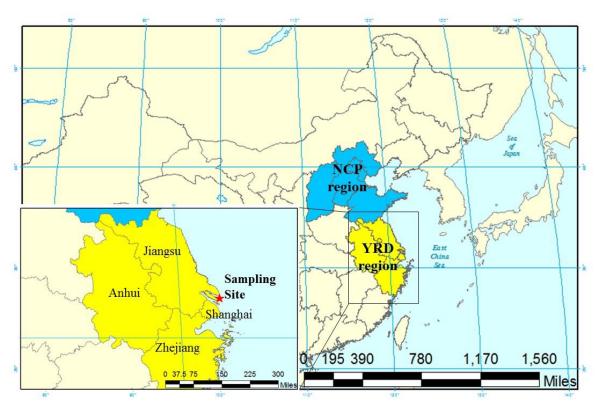
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Figure citation

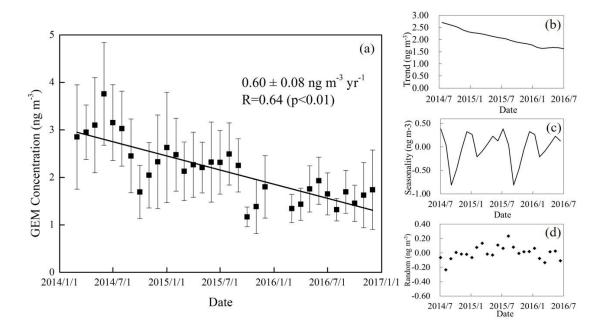**Figure 1.** The location of the Chongming monitoring site in Shanghai, China

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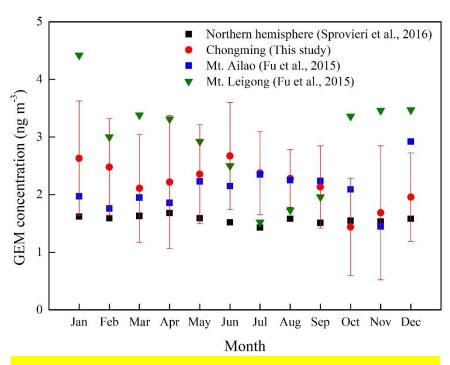
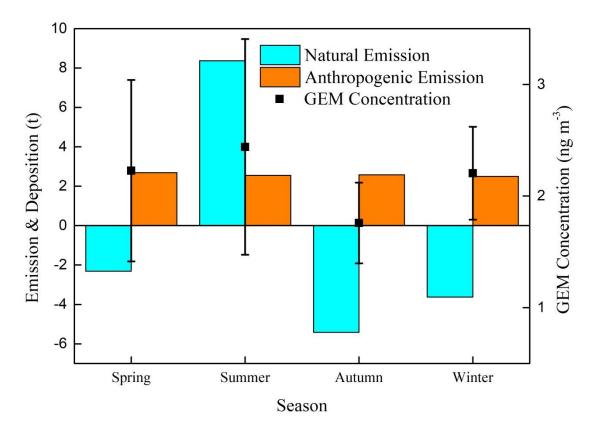**Figure 3.** Monthly variations of GEM concentration at remote sites in China

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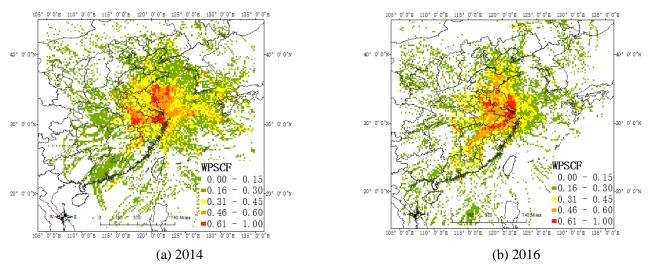
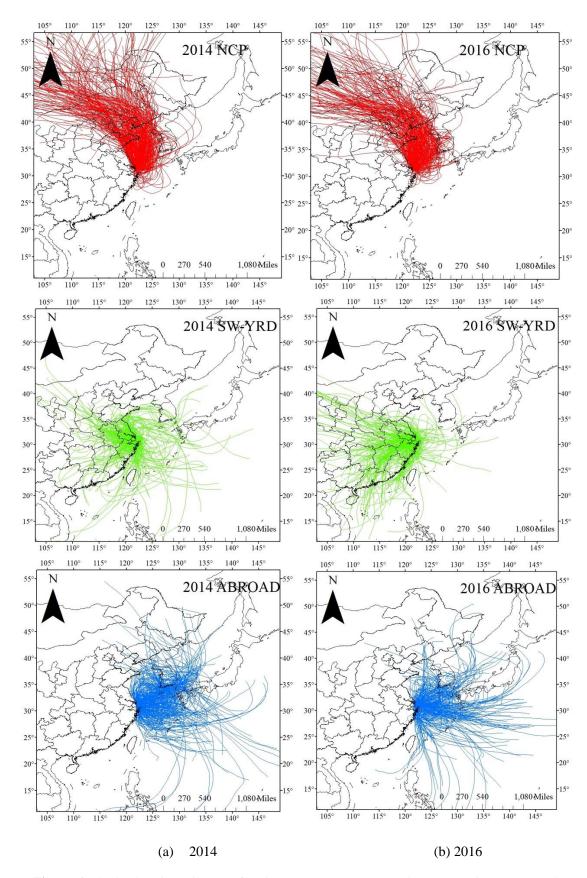**Figure 5.** Source regions of GEM at monitoring site from PSCF model in 2014(a) and 2016(b)

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-pollutants

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

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(a) 2014 (b) 2016

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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	Invasion of air mass from stratosphere	Component	Combustion	Invasion 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* "

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12 pages (including cover page)

4 Figures (S1, S2, S3, S4)

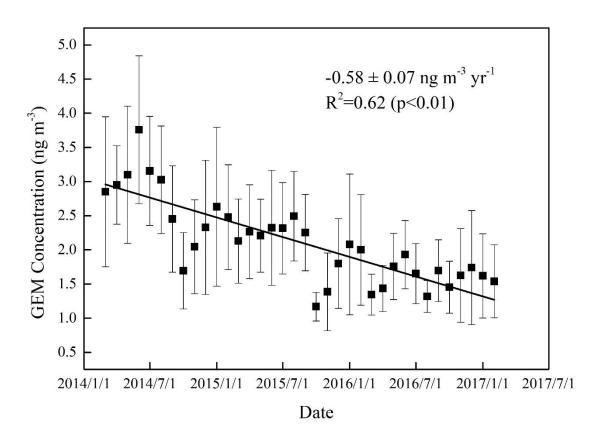**6 Tables (S1, S2, S3, S4, S5, S6)**

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.
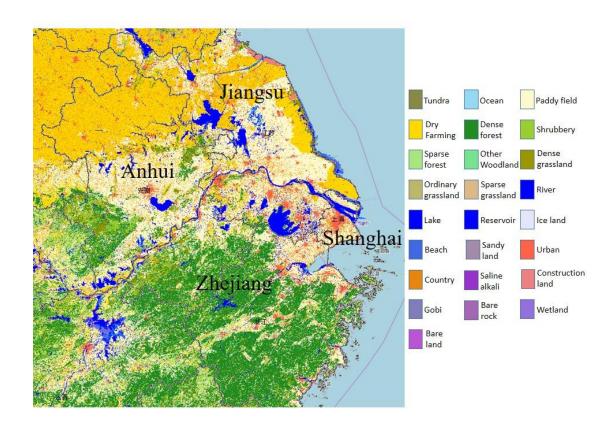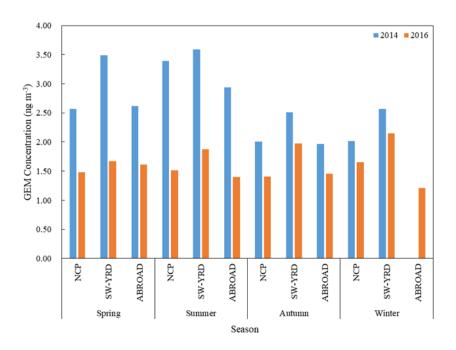
Figure S2. Land use type of YRD region in 2015

Figure S3. The seasonality of GEM concentration in the NCP, SW-YRD and ABROAD region (No trajectory transport though ABROAD in winter of 2014)
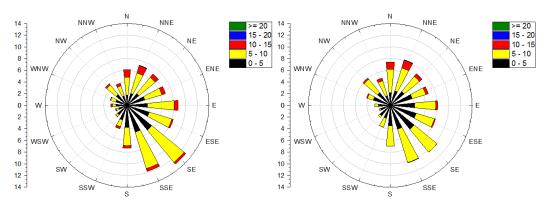
Figure S4. Wind rose at Chongming island in 2014 and 2016

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(°C)	2016(°C)	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