

1 **Reply to Comments from Reviewer #1**

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

5 6 **To reviewer**

7 ***Comment 1:***

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

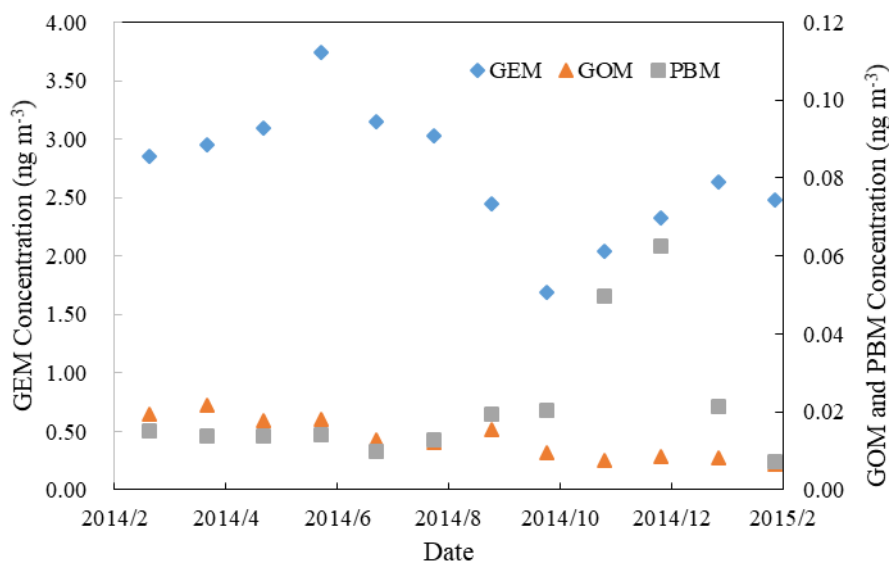
14 ***Response:***

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

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

27 Figure R1 showed the seasonal variation of GOM and PBM from March 2014 to February
28 2015. Considering that the concentrations of GOM and PBM were affected by emissions,
29 weather condition and deposition processes simultaneously, we need more researches to

30 determine the dominant impact factors. So it is hard to get some additional 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 $-0.60 \pm 0.08 \text{ ng m}^{-3} \text{ yr}^{-1}$ ($R^2=0.64$, $p < 0.01$ 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

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

67

68 **Comment 3:**

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

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

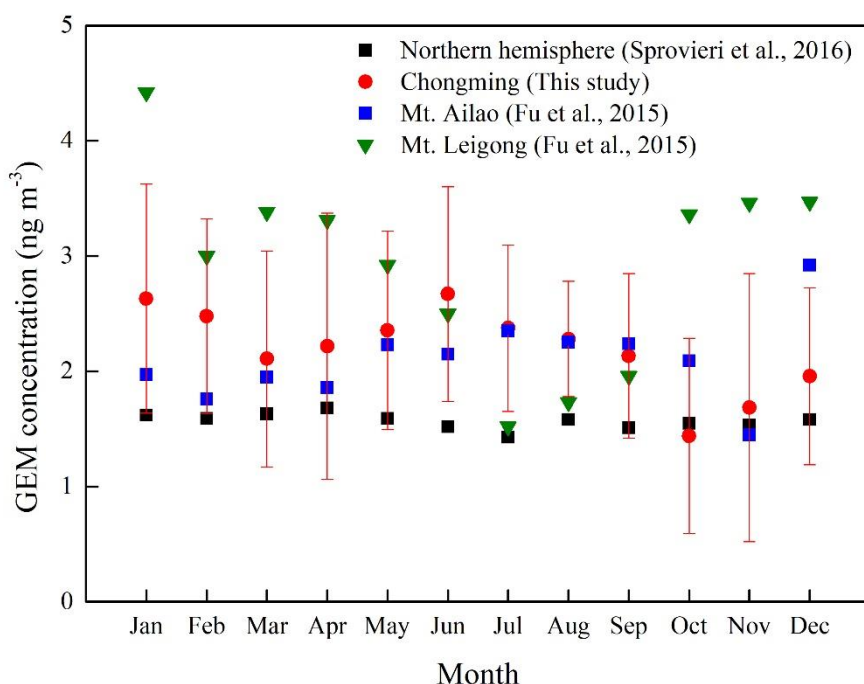
76 average during sampling period. The monthly GEM concentrations were detrended before the
77 average (Figure 2). The standard deviations of the monthly means have been added.

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

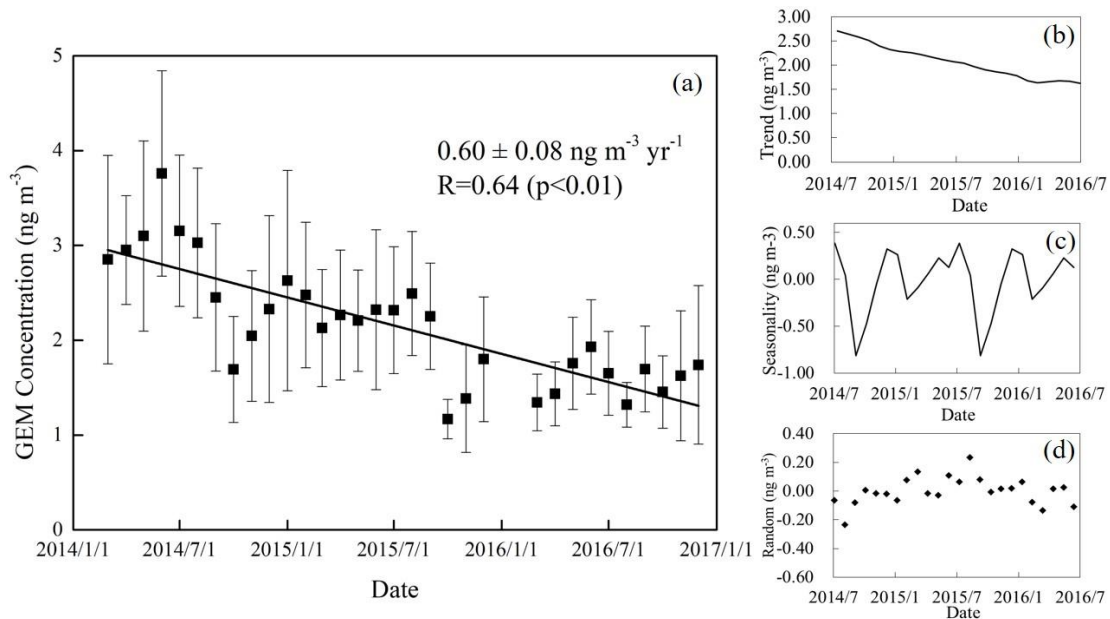
87 **See revised manuscript at Figure 3.**

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

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

99 Note: The observed concentrations during July 2015-April 2016 were TGM concentrations indeed
100 due to the problems of Tekran 1130/1135. However, the GOM concentrations at Chongming island
101 accounted for less than 1% of TGM. Thus, the GEM concentrations were approximated to TGM
102 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 deposits. We have made this concept clear in both text and Figure 4.

120

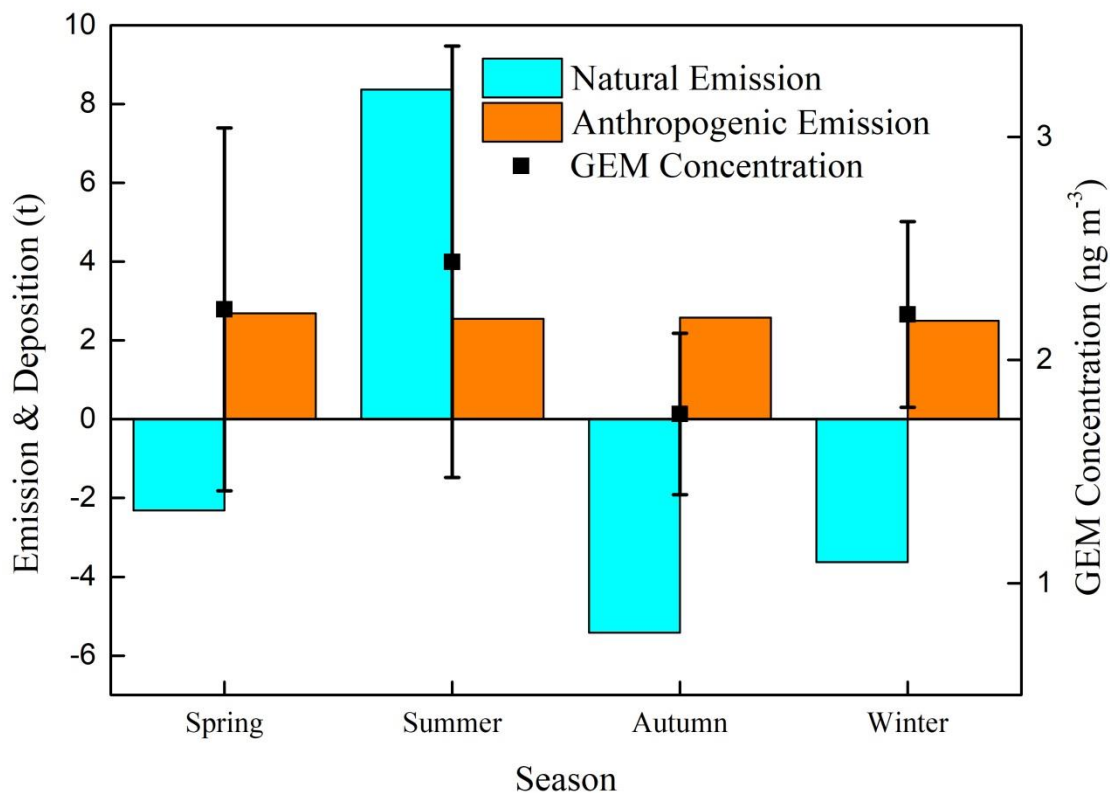
121 “The GEM emissions from natural sources E_N are calculated as followings.

$$122 \quad E_N = \sum_i F_i \times A_i \times t \quad (6)$$

123 Where F_i is a bi-directional Hg flux of canopy i , $\text{ng km}^{-2} \text{yr}^{-1}$; 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

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

137 **See the revised manuscript Figure 4.**

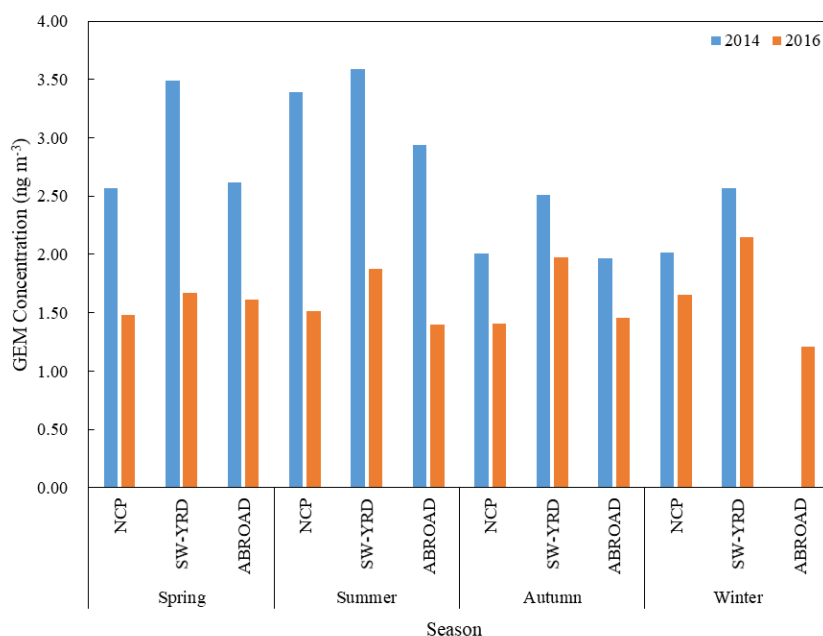
138

139 4.2 This will rise a problem: trajectory analysis in section 3.4 shows large influence of transport
 140 from the NW provinces of China outside of the YRD region. How does this transport influence
 141 the seasonal variation?

142 **Response:**

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

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



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

156 We also revised the manuscript as below.

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

161 **See the revised manuscript at line 314 - 317**

162

163 **Comment 5:**

164 Line 243-245: “The annual emissions from both natural source and anthropogenic source...
 165 was 0.75 and 10.3 t, respectively” – the reader may think that natural emissions make less than
 166 10% of the anthropogenic ones and cannot thus be responsible for the seasonal variation. One
 167 has to look in Fig. 4 to find out that the “natural emissions” are a sum of natural emissions in
 168 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.

198

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

199 where F_i is a bi-directional Hg flux of canopy i , $\text{ng km}^{-2} \text{ yr}^{-1}$; 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 O₃, 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 O₃ concentration. O₃ 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 SO₂ 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
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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 SO₂, 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 SO₂, 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

268 ***Comment 9:***

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

274 ***Response :***

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

278

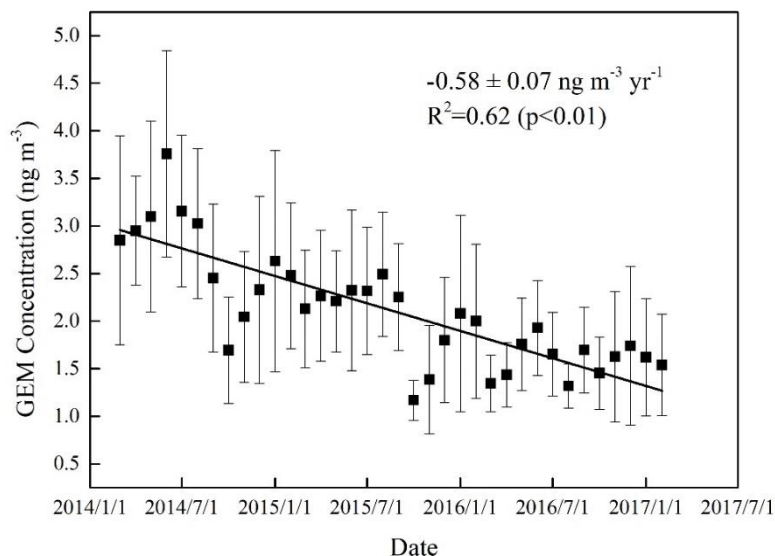
279 ***Comment 10:***

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

283 ***Response :***

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

290 The Figure S1 was revised as below.



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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374 Concentrations Measured at Several Sites in the Southern Hemisphere. *Atmospheric Chemistry and*
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383 Network. *Atmospheric Chemistry and Physics*, 2016, 16(18): 11915-11935.

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

398 **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**

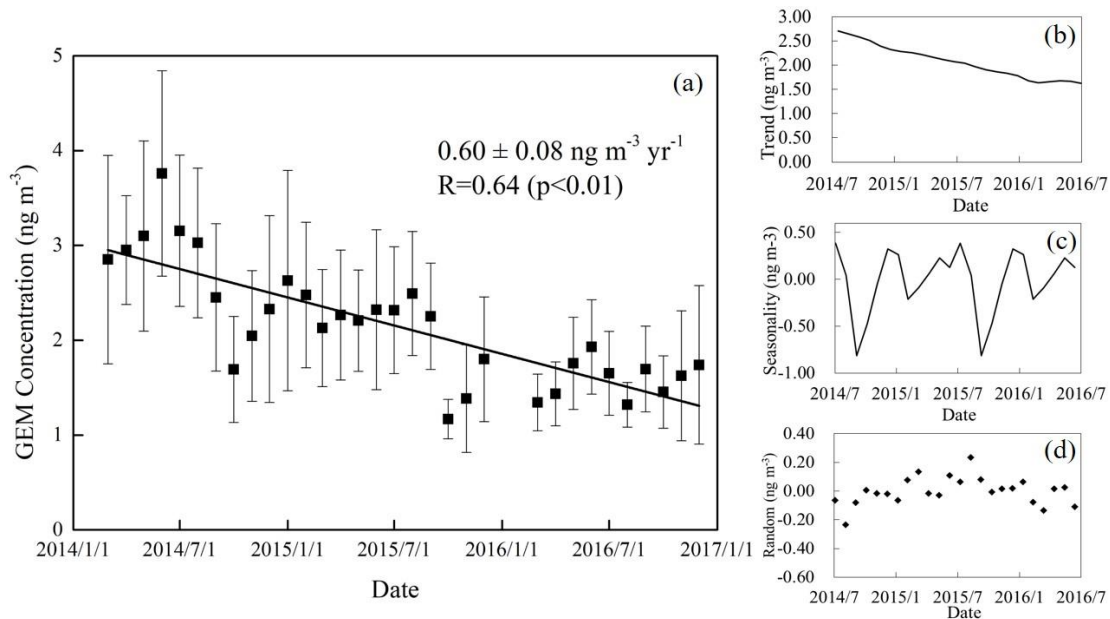
403 ***Major Comment:***

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 1 (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 $\cdot\text{OH}$ 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

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

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

432

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

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

439 **See the revised manuscript at line 238 - 243**

440

441 “From Figure 1, we also observed more pronounced seasonal variation in 2014, which can
 442 be attributed to the lower wet deposition and GEM oxidation. On one aspect, as a costal site,
 443 the Chongming Island is abundant with •OH. The increase of O₃ 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 SO₂, NO₂, 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 SO₂, NO₂, 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 SO₂, NO₂, and PM showed a
462 decreasing trend, which is used to support that "air pollution control policies targeting SO₂,
463 NO₂, 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 SO₂, NO_x, O₃ 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} ($\mu\text{g m}^{-3}$)		71.93	53.05	25.09	60.75	44.75	23.89	-16%	-16%	-5%
SO ₂ ($\mu\text{g m}^{-3}$)		34.52	21.01	1.60	24.37	16.40	1.47	-29%	-22%	-8%
NO ₂ ($\mu\text{g m}^{-3}$)		45.07	34.34	12.62	41.55	34.40	10.84	-8%	0%	-14%
O ₃ ($\mu\text{g m}^{-3}$)		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%

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 help 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 $2.86 \pm 0.95 \text{ ng m}^{-3}$ (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⁻³
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 ± 43.31 pg m⁻³) to 2016 (22.07 ± 30.55 pg m⁻³), which was also consistent with the downwawrd

626 trend of GEM. However, the GOM concentration increased from $(15.41 \pm 16.02 \text{ pg m}^{-3})$ in
627 2014 to $(18.97 \pm 9.28 \text{ pg m}^{-3})$ 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 $14.81 \pm 13.21 \text{ pg m}^{-3}$, 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}, O₃, NO_x, CO and SO₂ were monitored".

685 Why aren't you discussing the data, especially SO₂, NO_x, PM_{2.5} while your main conclusion is

686 that Hg decreasing trend in due to air pollution control policies targeting SO₂, 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 SO₂, NO₂, and PM showed a

694 decreasing trend, which is used to support that "air pollution control policies targeting SO₂,

695 NO₂, 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 SO₂, NO_x, O₃ 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} (µg m ⁻³)		71.93	53.05	25.09	60.75	44.75	23.89	-16%	-16%	-5%
SO ₂		34.52	21.01	1.60	24.37	16.40	1.47	-29%	-22%	-8%

($\mu\text{g m}^{-3}$)									
NO ₂	45.07	34.34	12.62	41.55	34.40	10.84	-8%	0%	-14%
($\mu\text{g m}^{-3}$)									
O ₃	60.29	56.27	41.70	61.84	60.92	44.38	3%	8%	6%
($\mu\text{g m}^{-3}$)									
GEM	No data		2.68	No data		1.60		No data	-40%
(ng m^{-3})									

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 $2.14 \pm 0.82 \text{ ng m}^{-3}$. 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 $2.68 \pm 1.07 \text{ ng m}^{-3}$, $2.14 \pm 0.82 \text{ ng m}^{-3}$, and $1.60 \pm 0.56 \text{ ng m}^{-3}$, 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 ($2.68 \pm 1.07 \text{ ng m}^{-3}$) 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 $-0.60 \text{ ng/m}^3/\text{yr}$ ". Table 1 refers to TGM concentrations, not GEM.
768 Additionally, as mentioned earlier, the rate is $-0.52 \text{ ng/m}^3/\text{yr}$ 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 $14.81 \pm 13.21 \text{ pg m}^{-3}$, 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 ³ in 1996 to 1.19 ng m⁻³ in 2004 (Slemr et al., 2008) and were increasing from 0.93 ng m⁻³ in
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?

819 **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 maintained by the Environment Canada- developed
823 Research Data Management and Quality Assurance System (RDMQ). At Zeppelin, the
824 instrument maintenance were carried out by the protocols of Norwegian mercury monitoring
825 program. Instruments at these places have been maintained 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 instruments 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 manuscript.

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
Kuujuarapik, 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	-	$-1.3 \pm 0.2\% \text{ y}^{-1}$	Rural	2537A	Weigelt et al. 2015
Yong San, South Korea	2004- 2011	No trend ($3.54 \pm 1.46 \text{ ng m}^{-3}$)		Urban	AM-3	Kim et al. 2016
Yong San South Korea	2013- 2014	Decrease to $2.34 \pm 0.73 \text{ ng m}^{-3}$			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

839 **Comment 15:**

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

845 **Response:**

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

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

854 **See the revised manuscript at line 266 - 271**

855

856 **Comment 16:**

857 Line 224: Are you referring to Figure 2?

858 **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

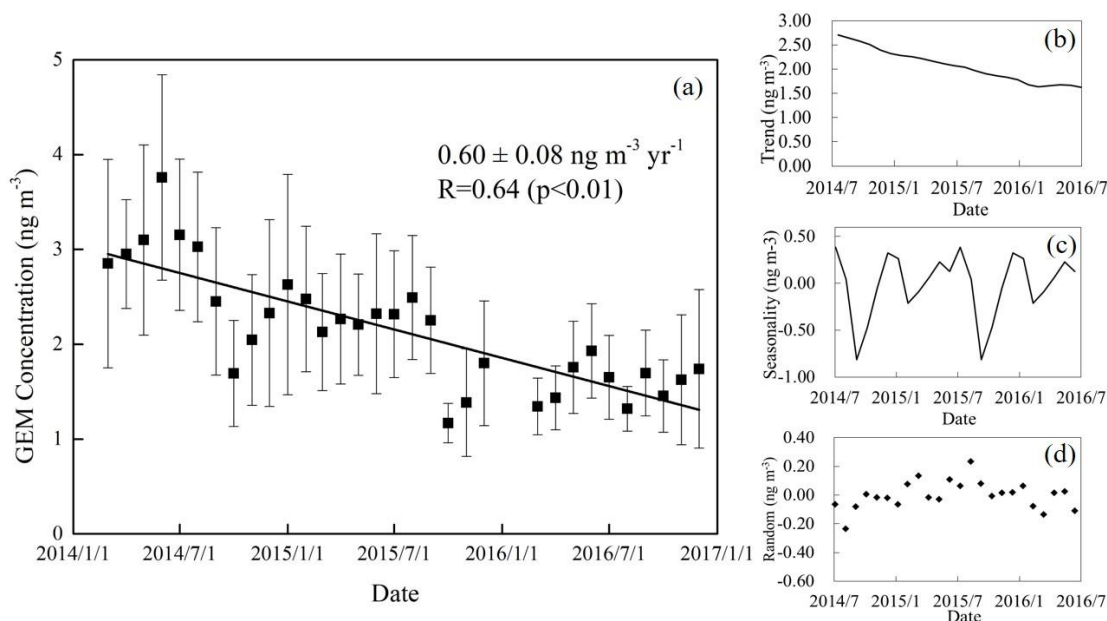
881 **Comment 19:**

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 O₃ 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 1, 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 O₃ 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 Korea
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, NO_x, and PM
937 concentrations?

938 ***Response:***

939 The SO₂, 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, NO_x, and PM_{2.5} due to the following
941 reasons. First, the residential time of SO₂, NO_x, 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 Hg⁰. Second, SO₂, NO_x and PM_{2.5} are more reactive in the
944 atmosphere compared with Hg⁰ (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 $\times 0.52 = 0.182$ 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:**

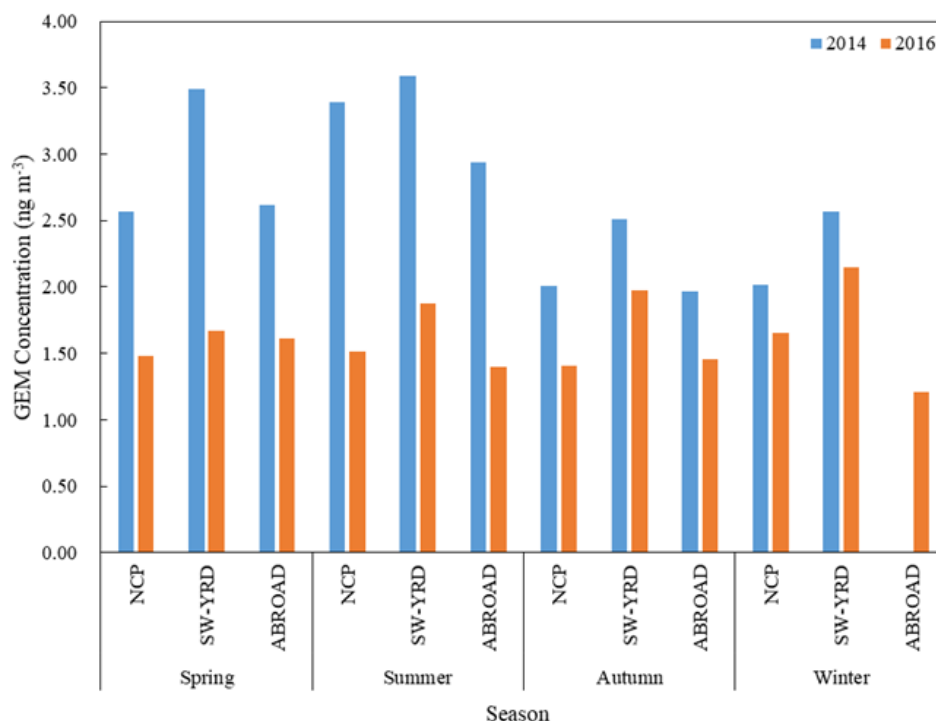
983 The seasonality of GEM concentration in the various clusters was showed in Figure S3. In
984 our revised manuscript, cluster NW, SW and EAST were modified to cluster NCP, SW-YRD
985 and ABROAD. The seasonality of cluster NCP, SW-YRD and ABROAD were similar to the
986 seasonality at Chongming. The 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

1080 **Recent decrease trend of atmospheric mercury concentrations in East China: the**
1081 **influence of anthropogenic emissions**

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1099

1100 **Abstract**

1101 Measurements of gaseous elemental Hg (GEM), other air pollutants including SO₂, NO_x, O₃,
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 (R²=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 SO₂, NO_x and
1115 particulate matter reductions had significant co-benefits on GEM.

1116 **1 Introduction**

1117 Mercury (Hg) is of crucial concern to public health and the global environment for its
1118 neurotoxicity, long-distance transport, and bioaccumulation. The atmosphere is an important
1119 channel for global Hg transport. Once atmospheric Hg deposits to the aquatic system, it can be
1120 transformed into methylmercury (MeHg) which bio-accumulates through the food web and affects
1121 the central nervous system of human beings (Mason et al., 1995). Hg is therefore on the priority list
1122 of several international agreements and conventions dealing with environmental protection,
1123 including the *Minamata Convention on Mercury*.

1124 Atmospheric Hg exists in three operationally defined forms: gaseous elemental mercury (GEM),
1125 gaseous oxidized mercury (GOM), and particulate-bound mercury (PBM). And the sum of GEM
1126 and GOM is known as total gaseous mercury (TGM). In the atmosphere, Hg mainly presents as
1127 GEM, accounting for over 95% of the total in the most observation sites (Fu et al., 2015; Li et al.,
1128 2016; Zhang et al., 2017). GEM is stable and with low solubility in the troposphere with a long
1129 residence time and can be transported at regional and global scale (Lindberg et al., 2007). GEM can
1130 be oxidized through photochemical reaction to GOM, which can be converted to PBM upon
1131 adsorption/absorption on aerosol surfaces. GOM is much soluble than GEM, and PBM can be
1132 quickly scavenged by both dry and wet deposition. Therefore, the residence time of both GOM and
1133 PBM is shorter than that of GEM, generally several days to a few weeks for GOM and 0.5 – 2 year
1134 for GEM (Schroeder and Munthe, 1998).

1135 The atmospheric Hg observation results are important evidences to assess the effect of Hg
1136 emission control. During the past decades, significant decreases of GEM concentrations in Europe
1137 and North America have been observed (Cole et al., 2013; Weigelt et al., 2015). Air Hg
1138 concentrations in the northern hemisphere are reported to decline by 30-40% between 1990 and
1139 2010 (Zhang Y et al., 2016). Such a decrease is consistent with the decrease in anthropogenic Hg
1140 emissions inventory in Europe and North America (Streets et al., 2011). So far, most of the long-
1141 term observations on the ground sites have been carried out in the developed countries. For the
1142 developing countries such as China, limited atmospheric Hg observations have been carried out (Fu
1143 et al., 2008b; Zhang H et al., 2016; Hong et al., 2016) and there is no official national observing
1144 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 $2.86 \pm 0.95 \text{ ng m}^{-3}$ (in the range of $1.60\text{-}5.07 \text{ ng m}^{-3}$) 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., $\text{PM}_{2.5}$ 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.

1171 **2 Materials and methods**

1172 **2.1 Site descriptions**

1173 The monitoring remote site (31°32'13"N, 121°58'04"E, about 10 m above sea level) locates at the
1174 top of weather station in Dongtan Birds National Natural Reserve, Chongming Island, China (Figure
1175 1). As China's third largest island, Chongming Island is located in the east of Yangtze River Delta
1176 region with a typical subtropical monsoon climate. It is rainy, hot, with southern and southeastern
1177 winds in summer and is dry, cold, and with northwestern wind in winter. The dominant surface types
1178 are farmland and wetland. There are no large anthropogenic emission sources in the island and no
1179 habitants within 5 km distance from the site. The downtown Shanghai area is 50 km to the southwest
1180 of the site.

1181 **2.2 Sampling methods and analysis**

1182 During the monitoring period, we used Tekran 2537X/1130/1135 instruments to monitor
1183 speciated Hg in the atmosphere, which was widely used for air Hg observation in the world. The
1184 sampling inlet was 1.5 m above the instrument platform. Continuous 5-minute of GEM was
1185 measured by Tekran 2537X Hg vapor analyzer with the detection limit of 0.1 ng m⁻³ at a sampling
1186 flow rate of 1.0 L min⁻¹ during two campaigns: March 1, 2014 to December 31, 2015 and March 26
1187 to December 31, 2016. From July 5, 2015 to April 30 2016, the Tekran 1130/1135 speciation unit
1188 was damaged by the rainstorm, the Tekran 2537X were operated without speciation units but with
1189 PTFE filter to protect the instrument from particles and sea salt. Therefore, the observed
1190 concentrations during July 2015-April 2016 were TGM concentrations indeed. However, the GOM
1191 concentrations at Chongming Island accounted for less than 1% of TGM (TGM=GOM+GEM).
1192 Thus, the GEM concentrations were approximated to TGM concentrations from July 2015 to April
1193 2016.

1194 The 2537X analyzer was calibrated automatically every 25 h using the internal Hg permeation
1195 source inside the instrument, and the internal permeation source was calibrated every 12 months
1196 with manual injection of Hg by a syringe from an external Hg source (module 2505). Two zero and
1197 two span calibrations were performed for each calibration of gold trap A and B, respectively. The
1198 difference between gold trap A and gold trap B was limited to ±10%. The impactor plates and quartz
1199 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}, O₃, NO_x, CO and SO₂ were also monitored by Thermo
1210 Scientific TEOM 1405D, Model 49i O₃ Analyzer, Model 48i CO Analyzer, Model 42i-TL NO_x
1211 Analyzer and Model 43i SO₂ Analyzer, respectively. The detection limits of O₃, SO₂, NO_x, CO and
1212 PM_{2.5} are 1.0, 0.5, 0.4, 0.04 and 0.1 µg 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:

$$1225 \quad PSCF_{ij} = \frac{m_{ij}}{n_{ij}} W_{ij} \quad (1)$$

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:

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

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 SO₂ 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 (SO₂, 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

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

$$1265 \quad TWC_{j,t} = AR \times C_{j,t} \quad (4)$$

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.

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

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-

1282 based emission factor methods and transformed normal distribution function method. Detailed
1283 introduction of these two methods and the speciation profile of the emitted Hg for each sector are
1284 described in our previous study (Wu et al., 2016). Conventional air pollutant (SO₂, PM_{2.5}, and NO_x)
1285 emissions were calculated following the study of Zhao et al. (2013). The source regions included in
1286 the emission inventory consisted of Shanghai, Jiangsu, Zhejiang, and Anhui provinces according to
1287 the PSCF results (See section 3.3). The studied emission sectors included coal-fired power plants,
1288 coal-fired industrial boilers, residential coal-combustion, cement clinker production, iron and steel
1289 production, mobile oil combustion and other small emission sectors (eg., zinc smelting, lead
1290 smelting, municipal solid incineration, copper smelting, aluminum production, gold production,
1291 other coal combustion, stationary oil combustion, and cremation). The monthly Hg emissions were
1292 mainly distributed according to fuel combustions or products productions by month (Table S1). For
1293 small emission sectors, the annual emissions were equally distributed into monthly emissions. The
1294 GEM emissions from natural sources E_N are calculated as followings.

$$1295 \quad E_N = \sum_i F_i \times A_i \times t \quad (6)$$

1296 where F_i is a bi-directional Hg flux of canopy i , ng km⁻² yr⁻¹; A is the studied area, km²; t is the
1297 studied year, yr. The bi-directional Hg flux was obtained from the study of Wang et al. (2016)
1298 directly. It should be pointed out that the natural emission is a concept of net emission in this
1299 manuscript, which reflected a net effect of two competing processes (Zhang, 2009): total Hg natural
1300 emissions and total Hg deposition. The total natural emissions included primary natural release and
1301 re-emission of legacy Hg stored in the terrestrial and water surface (Wang et al., 2016). When the
1302 value is positive, it means the net effect is Hg emissions to air. Otherwise, Hg deposited.

1303 **3 Results and discussions**

1304 **3.1 Decreasing trends of atmospheric Hg during 2014-2016**

1305 The average concentrations of GEM in 2014 (March to December), 2015 and 2016 (March to
1306 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
1307 GEM concentrations in 2014 were higher (t test, $p < 0.01$) than the Northern Hemisphere back-
1308 ground concentration (about 1.5 ng m⁻³) (Sprovieri et al., 2010) and those measured in other remote
1309 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 $-0.60 \pm 0.08 \text{ ng m}^{-3} \text{ yr}^{-1}$ ($R^2=0.64$, $p<0.01$ significance level, $n = 32$) (Figure 2a). The
1313 amount of valid data for each month 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-](https://anomaly.io/seasonal-trend-decomposition-in-r/)
1316 [decomposition-in-r/](https://anomaly.io/seasonal-trend-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 $-0.24 - 0.24 \text{ ng m}^{-3}$ (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 4** 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 $13\text{-}35 \text{ pg m}^{-3} \text{ y}^{-1}$ (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.

1347 **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 1, 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 O₃ 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

1400 seasonal variation in 2016. Meanwhile, the higher oxidation and wet deposition in 2016 also
1401 contributed to the downward trend of GEM by reducing the seasonality in spring and summer
1402 (Figure S3).

1403 3.3 Source apportionment of atmospheric Hg pollutions

1404 According to the PSCF result, YRD region, including Shanghai, Jiangsu, Anhui, and Zhejiang
1405 provinces, was the dominant source region in both 2014 and 2016 (Figure 5). Therefore, Hg
1406 emissions from these areas would contribute to high proportion of Hg pollution at Chongming Island.
1407 The offshore area mainly around Jiangsu province also has a high PSCF value because some
1408 trajectories from North China, especially Shandong province, transport to Chongming Island
1409 through this area. Compared to the result in 2014, the PSCF value had an obvious decline in East
1410 China Sea in 2016. The decline from the East China Sea may be contributed by the downward trend
1411 of GEM concentrations in South Korea and Japan (Kim et al., 2016; Kim et al., 2013), where the
1412 anthropogenic Hg emissions of Japan and South Korea have been reduced by 13% and 4% during
1413 2010-2015, respectively (UNEP 2013; UNEP 2018). The air mass from Japan and South Korea
1414 would pass through the East China Sea to Chongming.

1415 PCA method was applied to preliminarily identify the source industries. In the studied period,
1416 totally 2 factors were identified in 2014 and 2016, respectively. The factor 1 had strong factor
1417 loadings of GEM, SO₂, NO_x, CO, and PM_{2.5} in both 2014 and 2016 (No CO data in 2016 due to
1418 equipment problems). The factor 1 accounted for 49% variance in 2014 and 50% variance in 2016
1419 (Table 11). The results indicated common significant source sectors of the above five air pollutants,
1420 which can also be proven from emission inventories (Table 2). The dominant source industries
1421 included coal-fired power plants, coal-fired industrial boilers, and cement clinker production. The
1422 PCA results showed that anthropogenic emissions were the main sources of GEM during the
1423 sampling period.

1424 The factor 2 in both 2014 and 2016 had a strong positive loading on O₃ and negative loading on
1425 NO_x. Considering the low loading of CO and high loading of O₃, the factor 2 can be viewed as a
1426 sign of the invasion of air mass from stratosphere (Fishman and Seiler, 1983; Jaffe, 2010). The air
1427 mass from stratosphere will increase the O₃ concentration. O₃ react with NO, which makes a
1428 negative correlation with NO. However, the low loading on GEM of factor 2 indicated that Factor
1429 2 had no relationship with GEM concentrations at Chongming from the aspect of whole year data.

1430 **3.4 The influence of anthropogenic emissions**

1431 To further understand the reason of the downward trend, we firstly compared the meteorological
1432 conditions in both 2014 and 2016. We noted that the difference of annual temperature, solar radiation,
1433 and relative humidity were constrained in the range of 17.13 ± 7.48 °C, 165.55 ± 45.87 W m⁻² and
1434 $75.38\pm 5.82\%$, respectively (Table S2). The coefficient of variation for annual mean of these
1435 meteorological conditions in 2014 and 2016 was 2.6%, 6.7% and 0.2%, respectively. In addition,
1436 the wind rose was similar, and the dominating wind was from SE in both 2014 and 2016 (Figure
1437 S4). The HYSPLIT results also provided similar trajectories in 2014 and 2016 (Figure 6). Therefore,
1438 we assumed that the meteorological condition was not the dominant reason of GEM decline at
1439 Chongming site.

1440 To further quantify the driver of GEM decline, a trajectory-based analysis method was used in
1441 this study. The 72-h air mass back trajectories were calculated using HYSPLIT for every 8 hours
1442 starting at the observation site. Approximately 918 and 832 trajectories were calculated in sampling
1443 period in 2014 (Mar 1 to Dec 31, 2014) and 2016 (Mar 26 to Dec 31, 2016), respectively. The
1444 trajectories were grouped into 3 clusters in each year according to geographical regions (Figure 6).
1445 The first cluster of trajectories mainly passed through the regions (eg., North China) north and
1446 northwest to Chongming Island before arriving to our monitoring site, which was denoted as cluster
1447 NCP. The second cluster mainly passed YRD region to Chongming, which was signed as cluster
1448 SW-YRD. The third type mainly originated from the East China Seas, South Korea, Japan and
1449 Northeast Asia continent, and then arrived to our monitoring sites directly without passing the
1450 mainland China. This type of trajectories was named as cluster ABROAD. Some trajectories
1451 originated from the East China Sea and crossed the mainland China before arriving Chongming
1452 were grouped into cluster NCP or SW-YRD depending on the regions it crossed. The trajectories
1453 for each of the three clusters in 2014 and 2016 were shown in Table 3.

1454 Table 3 showed the detail statistics data of the three classifications. From 2014 to 2016, the whole
1455 China region (NCP, SW-YRD) contributed to 70% of GEM decline at Chongming Island.
1456 Considering downward trend of emission inventory and atmospheric pollutant from 2014 to 2016
1457 in NCP and SW-YRD region (Table S5, Table S6), the reason of downward trend can be attributed
1458 to the effectiveness of existing air pollution control measures in China (SC, 2013; MEP, 2014).
1459 Meanwhile, the cluster NCP, cluster SW-YRD, and cluster ABROAD caused 26%, 44%, and 30%

1460 for GEM decline, respectively (Table 3). The cluster SW-YRD contributed to 44% of reduction,
1461 suggesting that air pollution controls on anthropogenic emissions in YRD region dominated the
1462 recent decrease of GEM concentrations at Chongming site. The largest decline of Hg concentration
1463 (1.32 ng m^{-3}) was also observed in the cluster SW-YRD demonstrated the efficiency of emission
1464 reduction in YRD region (Table S5, Table S6). Moreover, ABROAD region caused 30% of GEM
1465 decline from 2014 to 2016, which implies global effort on atmospheric Hg emission control under
1466 the guidance of *Minamata Convention on Mercury*.

1467 **4 Conclusion**

1468 Atmospheric Hg was continuously measured for three years at a regional background site in the
1469 YRD region. During the sampling period, a downward trend for GEM concentrations (-0.60 ± 0.08
1470 $\text{ng m}^{-3} \text{ y}^{-1}$) at Chongming Island was observed. The seasonal GEM cycle was dominated by the
1471 natural emissions while the annual GEM concentration trend was mainly impacted by anthropogenic
1472 emissions. By using a new approach that considers both cluster frequency and the Hg concentration
1473 associated with each cluster, we quantified that atmospheric Hg from NCP region, SW-YRD region,
1474 and ABROAD region have caused 26%, 44%, and 30% decline of GEM concentrations at
1475 Chongming monitoring site, respectively. The result suggested that reduction of anthropogenic
1476 emissions in mainland China was the main cause of the recent decreasing trend of GEM
1477 concentration at Chongming site. The air pollution control policies in China, especially the pollution
1478 control in the coal-fired power plants, coal-fired industrial boilers, and cement clinker production
1479 in YRD region and Shandong province, have received significant co-benefit of atmospheric Hg
1480 emission reductions. On the other hand, emission reduction from the ABROAD region, where
1481 clusters arrived to Chongming monitoring site directly without passing the mainland China, implies
1482 global effort on atmospheric Hg emission control under the guidance of *Minamata Convention on*
1483 *Mercury*. Considering that the *Minamata Convention on Mercury* had come into force in 2017,
1484 continuous long-term observation of atmospheric Hg in China will be required for the assessment
1485 of policy effectiveness.

1486

1487 *Data availability.* All data are available from the authors upon request.

1488

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

1490

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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

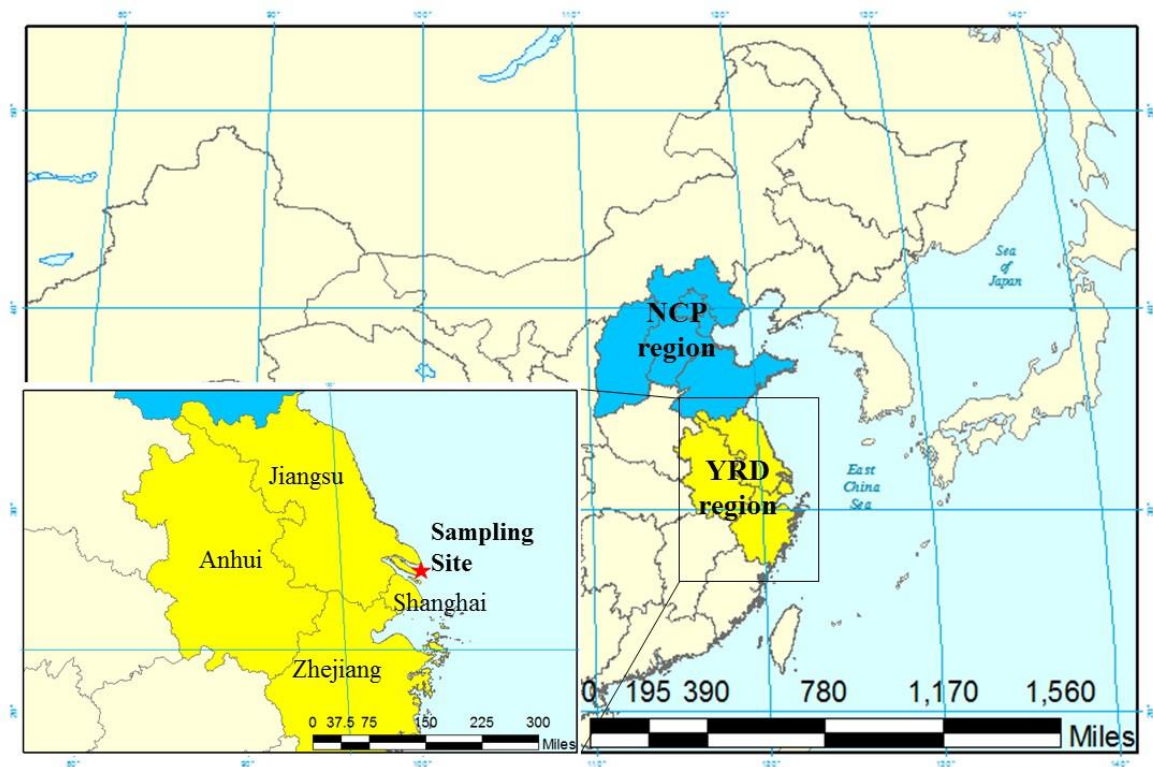


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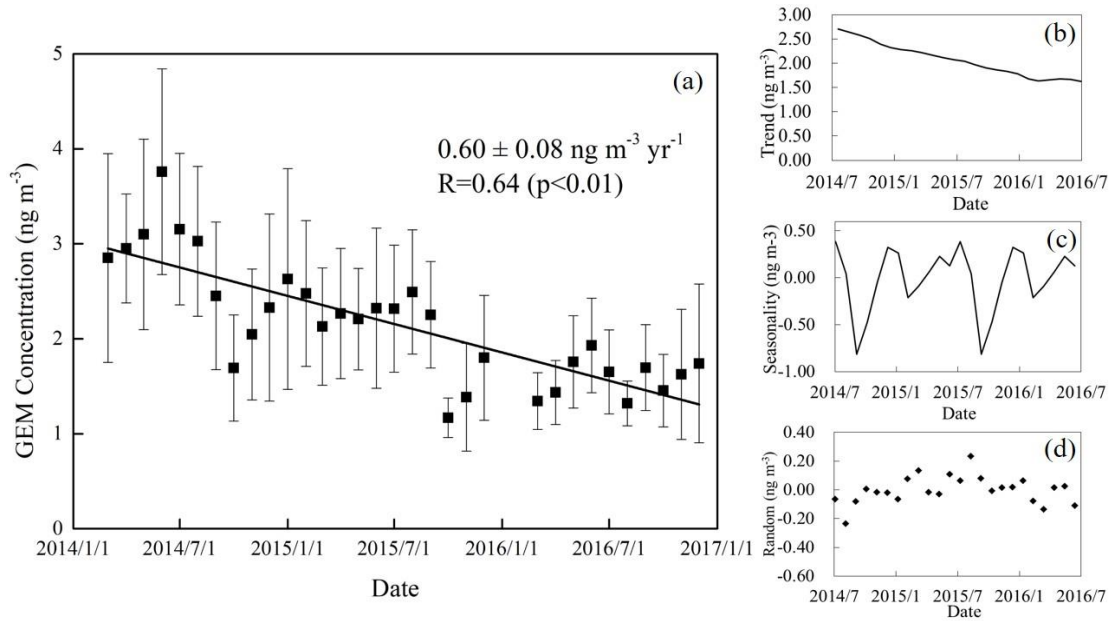


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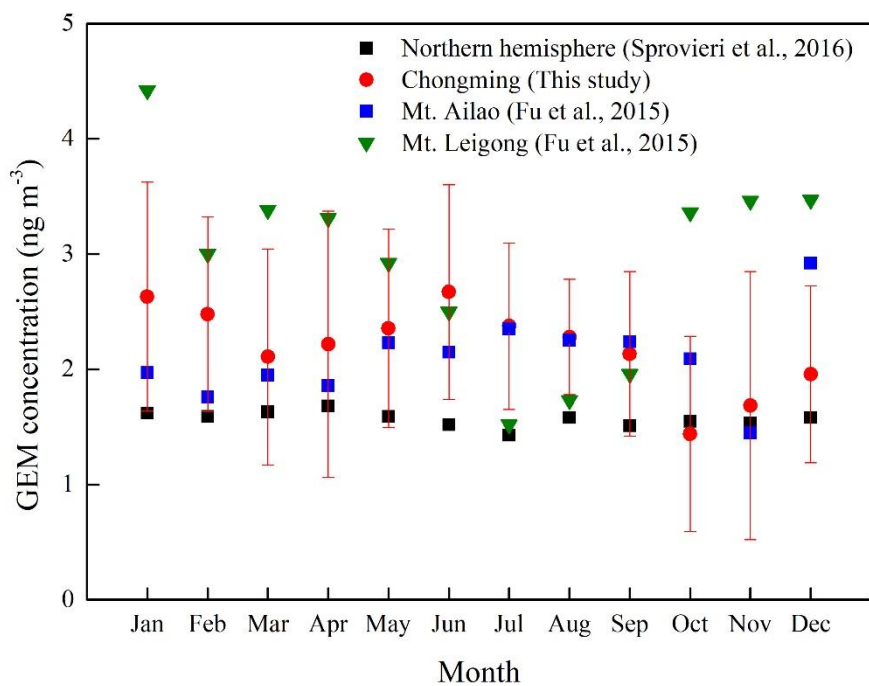


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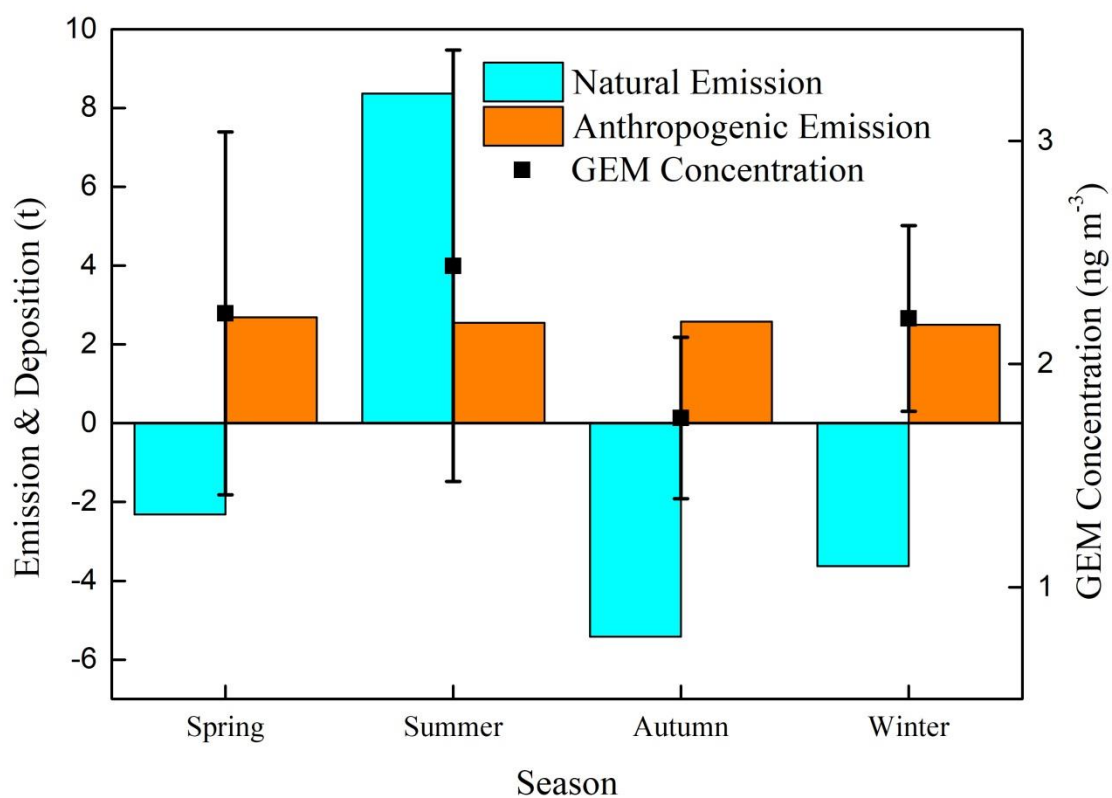
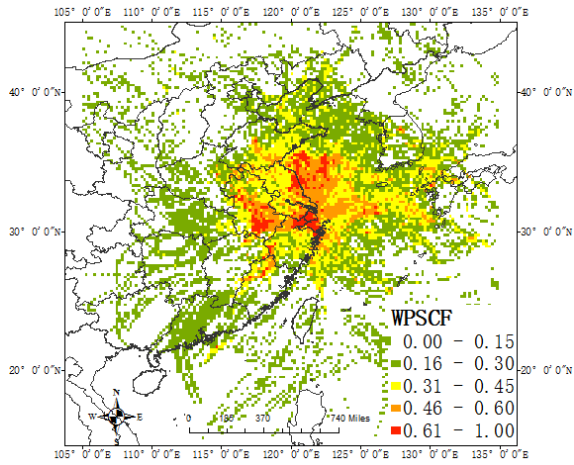
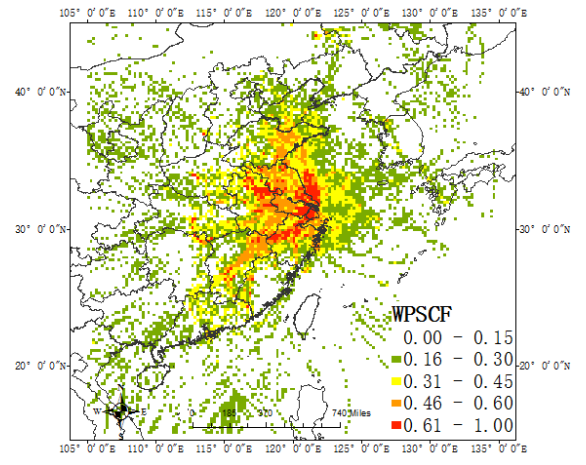


Figure 4. Seasonal cycle of GEM concentrations and natural emissions during 2014-2016. The error bars represent the standard deviation of seasonal average. Positive values of natural emissions represent Hg emitted to air. Otherwise, negative values represent Hg deposition.

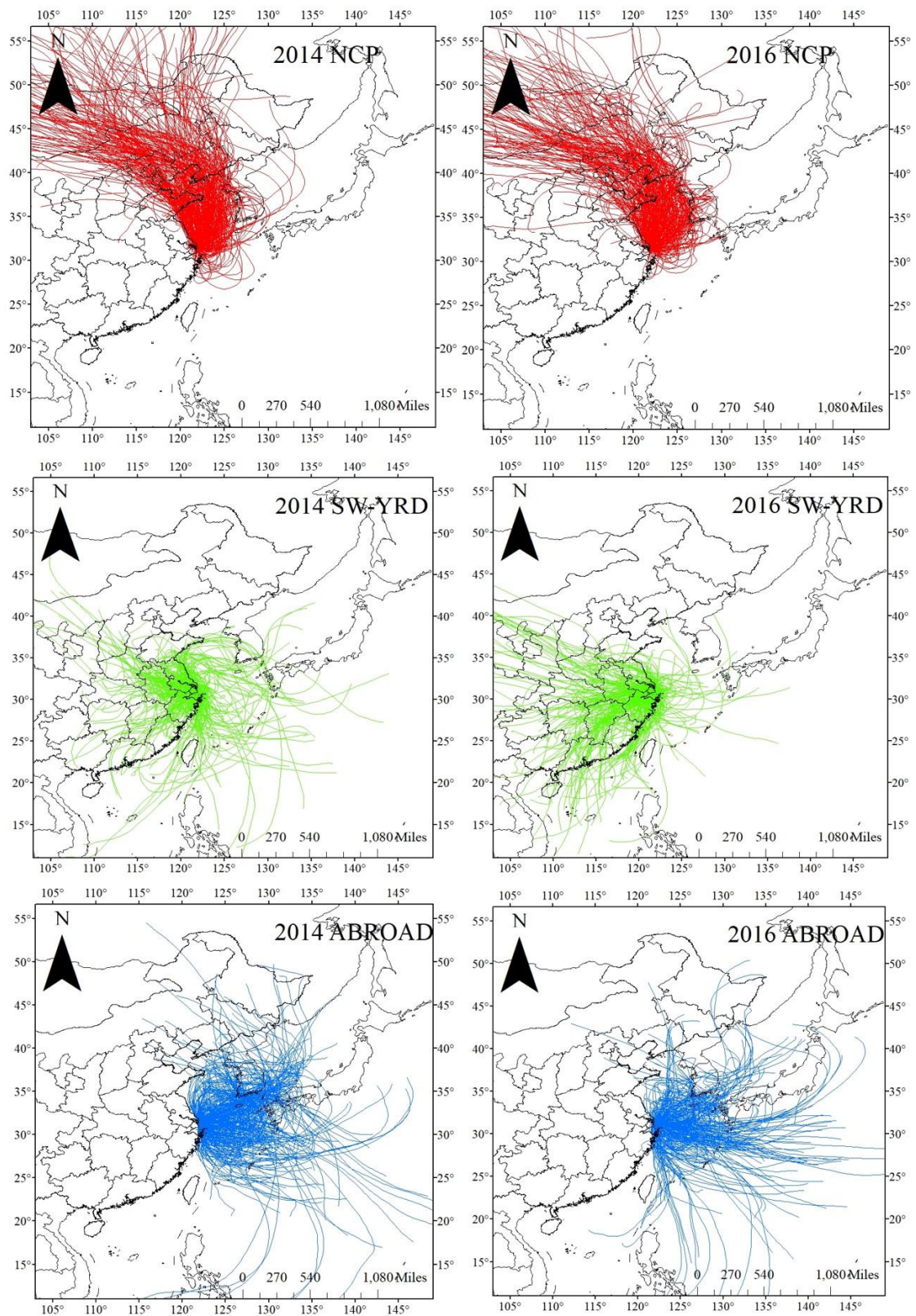


(a) 2014



(b) 2016

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(a) 2014

(b) 2016

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

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

Air pollutants	2014		Air pollutants	2016	
	Factor 1	Factor 2		Factor 1	Factor 2
SO ₂	0.76	0.14	SO ₂	0.82	-0.09
NO _x	0.76	-0.20	NO _x	0.70	-0.52
O ₃	-0.11	0.98	O ₃	-0.41	0.97
PM _{2.5}	0.85	0.05	PM _{2.5}	0.88	0.05
GEM	0.66	0.02	GEM	0.78	-0.19
CO	0.79	0.12			
Component	Combustion	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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7 Tables (S1, S2, S3, S4, S5, S6)

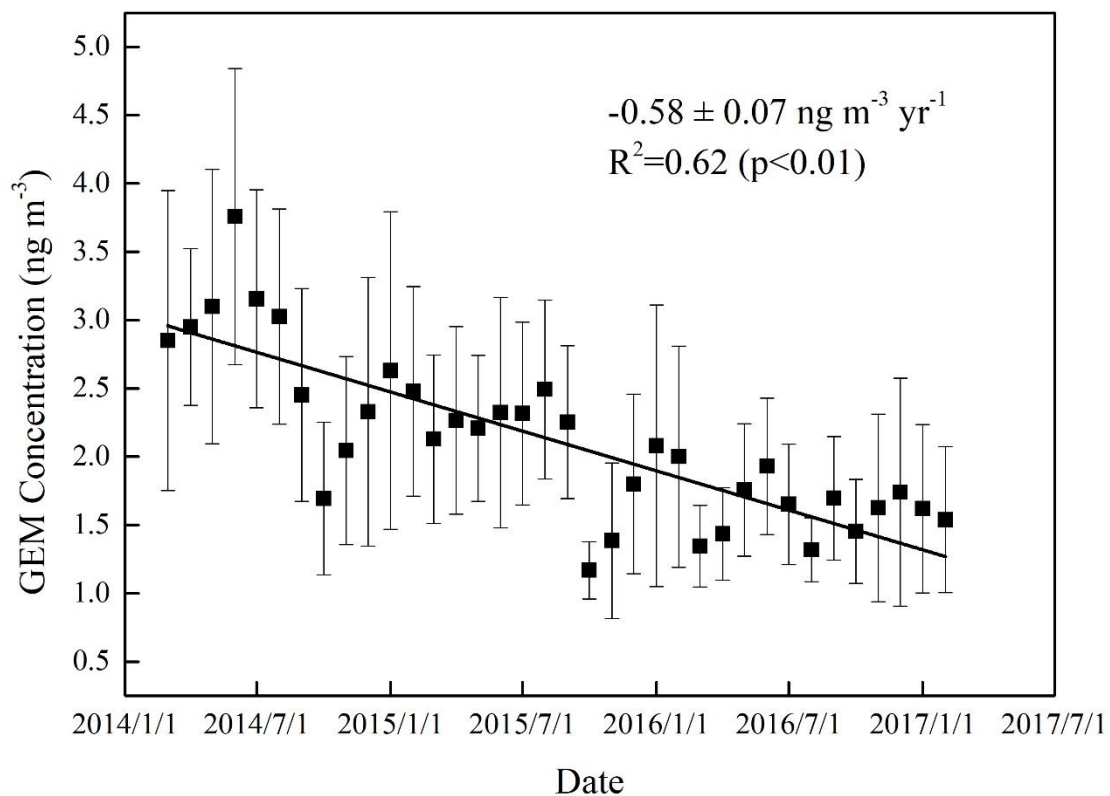


Figure S1. The trend of monthly average GEM concentration from March 2014 to February 2017. The monthly average of GEM in January of 2016 is simulated as the average value that in the January of 2015 and 2017. The same method is used for the data in February of 2016.

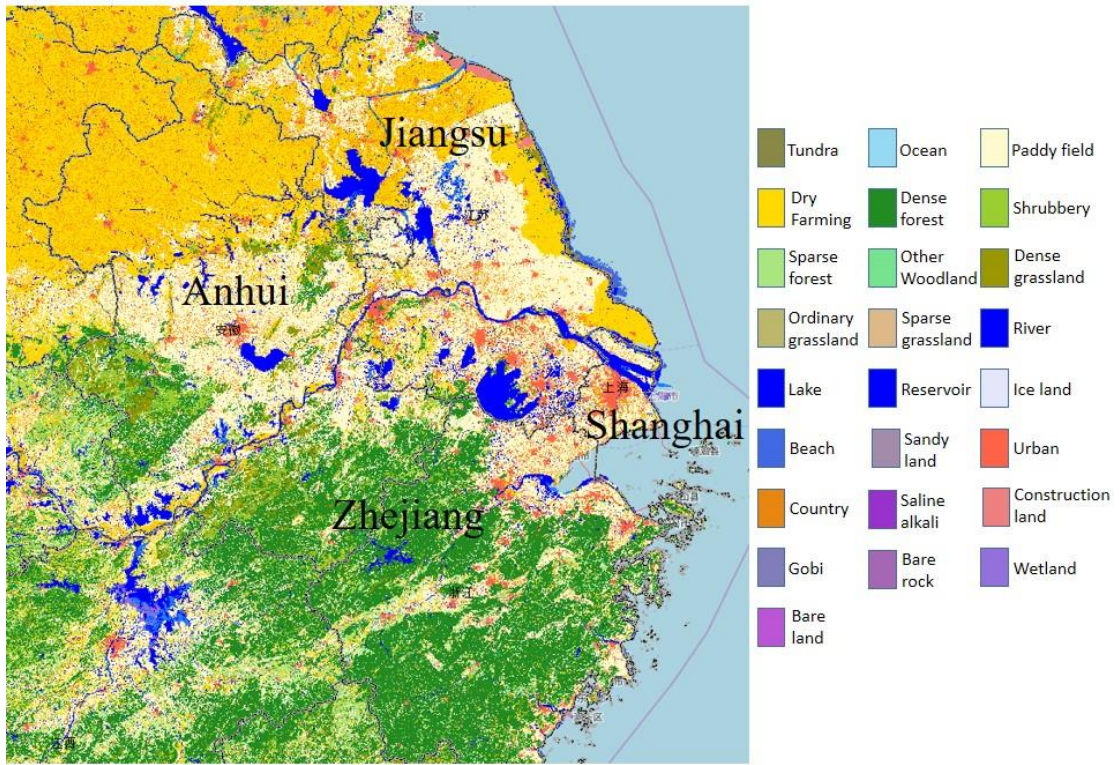


Figure S2. Land use type of YRD region in 2015

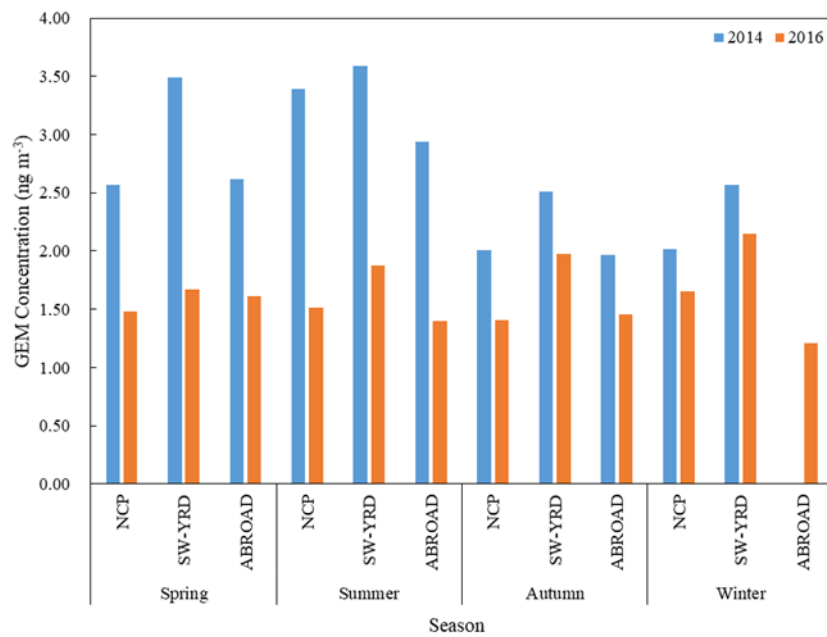


Figure S3. The seasonality of GEM concentration in the NCP, SW-YRD and ABROAD region (No trajectory transport though ABROAD in winter of 2014)

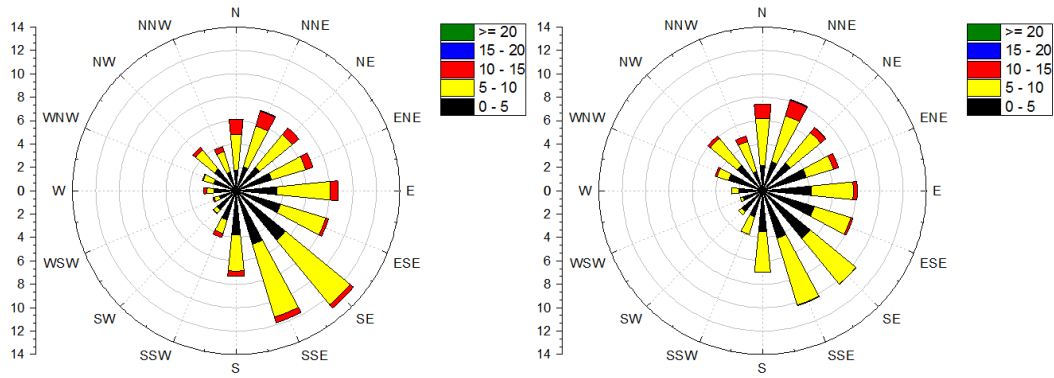


Figure S4. Wind rose at Chongming island in 2014 and 2016

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 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

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

Month	Temperature				Solar Radiation				Relative Humidity			
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
	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
Kuujuarapik, 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 SO₂, NO_x, O₃ 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} (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%

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

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

Air pollutants	2014		2016		Decline proportion	
	NCP	SW-YRD	NCP	SW-YRD	NCP	SW-YRD
PM _{2.5} (kt)	2019	1209	1849	1109	-8%	-8%
NO _x (kt)	5697	4022	5424	3855	-5%	-4%
SO ₂ (kt)	3780	1993	3450	1780	-9%	-11%
GEM (t)	118	72	103	67	-13%	-7%

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