

1 July 17, 2016

2

3 Dear Editor,

4

5 We appreciate the reviewers' suggestions which have considerably improved the manuscript
6 (**acp-2015-939**). Enclosed are point-by-point responses to the reviewers. We hope that with
7 these changes the manuscript will be suitable for publication in "**Atmospheric Chemistry
8 and Physics**"

9

10 Thank you very much.

11 Sincerely,

12 Seung-Muk Yi

13

14 Professor, Dept. of Environmental Health, Graduate School of Public Health

15 Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul 151-742, South Korea

16 Telephone: (82) 2-880-2736, Fax: (82) 2-762-9105, E-mail: yiseung@snu.ac.kr

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18 **Response to Anonymous Referees' Comments**

19

20 ● Journal: ACP

21 ● Title: Characteristics of total gaseous mercury (TGM) concentrations in an industrial
22 complex in southern Korea: Impacts from local sources

23 ● Author(s): Yong-Seok Seo, Seung-Pyo Jeong, Thomas M. Holsen, Young-Ji Han, Eunhwa Choi,
24 Eun Ha Park, Tae Young Kim, Hee-Sang Eum, Dae Gun Park, Eunhye Kim, Soontae Kim, Jeong-
25 Hun Kim, Jaewon Choi, Seung-Muk Yi

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30 ● Special Issue: Data collection, analysis and application of speciated atmospheric mercury

31

32 **Response to Anonymous Referee #1:**

33

34 **Comment 1**

35 Line 195: “The extension to the bivariate case can provide more information on the nature of
36 the sources because different source types such as stack emission sources and ground-level
37 sources can have different wind speed dependencies (prominent at low and high wind speed).”

38 This statement is incorrect because wind speeds are higher at higher elevation than at ground
39 level. It also contradicts a related sentence in lines 439-456: “CBPF shows that the high
40 probabilities from the west occurred under high wind speed ($>3 \text{ m s}^{-1}$) indicative of emissions
41 from stacks as well as low wind speed ($\leq 3 \text{ m s}^{-1}$) indicative of non-buoyant ground level
42 sources”

43

44 **Response 1**

45 The reviewer is correct – we reversed the order in this sentence. The corrected version is
46 shown below (Please see the [Line 225-228](#)).

47

48 *“The extension to the bivariate case can provide more information on the nature of the sources
49 because different source types such as stack emission sources and ground-level sources can
50 have different wind speed dependencies **(prominent at high and low wind speed, respectively)**.”*

51

52

53 **Comment 2**

54 Section 5.1: This paragraph seems out of place. The wind direction analysis should be
55 integrated with the mercury results rather than having its own section. The major result is the
56 mercury analysis rather than the wind direction analysis.

57

58 **Response 2**

59 This section is meant to provide background and context for the Hg results. To improve the
60 presentation we have moved this paragraph to section 2.2 and labeled it: “Meteorological
61 Setting” as shown below (Please see the [Line 180-184](#)):

62

63 *2.2. Meteorological data*

64 *Hourly meteorological data (air temperature, relative humidity, and wind speed and direction)*

65 were obtained from the Automatic Weather Station (AWS) operated by the Korea
66 Meteorological Administration (KMA) (<http://www.kma.go.kr>) (6 km from the site). Hourly
67 concentrations of NO_2 , O_3 , CO , PM_{10} and SO_2 were obtained from the National Air Quality
68 Monitoring Network (NAQMN) (3 km from the site) (Fig. 1).

69 Meteorological Setting. Fig. S1 shows the frequency of counts of measured wind direction
70 occurrence by season during the sampling period. The predominant wind direction at the
71 sampling site was W (20.9%) and WS (19.2%), and calm conditions of wind speed less than 1
72 $m s^{-1}$ occurred 7.6% of the time. Compared to other seasons, however, the prevailing winds in
73 summer were N (17.0%), NE (16.4%), S (16.4%), and SW (15.8%).

74

75

76 **Comment 3**

77 Line 293-294: “however considerably lower than those measured near large Hg sources in
78 Guangzhou, China (Table 1).” This is compared to a much older study. TGM in Guangzhou
79 from a more recent study in Table 1 was 4.6 ng/m^3 , which is similar to the average TGM in
80 this study.

81

82 **Response 3**

83 Thank you for this updated reference. We corrected **Table 1** on **Line 561** and rephrased the
84 sentence as follows on **Line 316-317**.

85

86 “...and those measured in China, in Japan and other locations in Korea, however lower than
87 those measured at Changchun, Gui Yang and Nanjing in China (Table 1).”

88

89

90 **Comment 4**

91 Section 5.5 diurnal variations: The higher daytime than nighttime result is not quite correct. As
92 stated in lines 365-366, “TGM generally showed a consistent diurnal variation with an increase
93 in the early morning (06:00-09:00) and a decrease in the afternoon (14:00-17:00).” The daytime
94 period from 6:00-18:00 includes the morning increase and afternoon decrease; therefore it’s
95 unclear if daytime TGM is really higher. I suggest explaining what caused the early morning
96 increase and afternoon decrease in TGM, instead of the cause for higher daytime TGM in
97 general. There were a few instances where it led to confusing results. E.g. line 384, “the higher

98 concentrations in the daytime than those in nighttime were due to local emission sources
99 because the daytime temperature (14.7 ± 10.0 °C) was statistically significantly higher than that
100 in the nighttime (13.0 ± 9.8 °C) (t-test, $p < 0.05$) and there was a weak but statistically
101 significant negative correlation between TGM concentration and ambient air temperature ($r =$
102 -0.08) ($p < 0.05$).” The negative correlation between TGM and temperature is inconsistent with
103 the higher daytime TGM. Another example in line 406, “TGM concentration was negatively
104 correlated with ambient air temperature ($r = -0.08$) ($p < 0.05$) because high ambient air
105 temperature in the daytime will increase the height of the boundary layer and dilute the TGM,
106 and the relatively lower boundary layer at nighttime could concentrate the TGM in the
107 atmosphere (Li et al., 2011).” This explanation contradicts higher daytime TGM as well. It only
108 explains why TGM is lower in the afternoon, but not the early morning increase.

109

110 **Response 4**

111 To clarify this section the day-night concentration variation and morning-afternoon variation
112 during the day were discussed separately as shown below. Please see the Section 5.4 on [Line](#)
113 [384-458](#).

114

115 *“Diurnal variations of TGM (Fig. 3), co-pollutants concentrations, and meteorological*
116 *data were observed (Fig. S4). TGM, O₃, CO, SO₂, and temperature in the daytime (06:00-*
117 *18:00) were higher than those in the nighttime (18:00-06:00) ($p < 0.05$) except PM₁₀ ($p =$*
118 *0.09) (Fig. S5). However, NO₂ during the nighttime because of relatively lower*
119 *photochemical reactivity with O₃ was higher than that in daytime ($p < 0.05$) (Adame et al.,*
120 *2012).*

121 *The daytime TGM concentration (5.3 ± 4.7 ng m⁻³) was higher than that in the nighttime*
122 *(4.7 ± 4.7 ng m⁻³) ($p < 0.01$), which was similar to several previous studies (Cheng et al.,*
123 *2014; Gabriel et al., 2005; Nakagawa, 1995; Stamenkovic et al., 2007) but different than*
124 *another studies (Lee et al., 1998). Previous studies reported that this different is due to local*
125 *sources close to the sampling site (Cheng et al., 2014; Gabriel et al., 2005), a positive*
126 *correlation between TGM concentration and ambient air temperature (Nakagawa, 1995) and*
127 *increased traffic (Stamenkovic et al., 2007). However, another study suggested that the*
128 *higher TGM concentration during the night was due to the shallowing of the boundary layer,*
129 *which concentrated the TGM near the surface (Lee et al., 1998).*

130 *In a previous study the daytime TGM concentration was relatively lower than that in the*
131 *nighttime because the sea breeze transported air containing low amounts of TGM from the*
132 *ocean during the daytime whereas the land breeze transported air containing relatively high*
133 *concentrations of TGM from an urban area during the nighttime (Kellerhals et al., 2003).*
134 *Although it is possible that the land-sea breeze may affect diurnal variations in TGM*
135 *concentrations since the sampling site was near the ocean and lower TGM were also*
136 *observed during the daytime, the higher concentrations in the daytime than those in nighttime*
137 *were due to local emission sources because the daytime temperature (14.7 ± 10.0 °C) was*
138 *statistically significantly higher than that in the nighttime (13.0 ± 9.8 °C) (*t*-test, $p < 0.05$)*
139 *and there was a weak but statistically significant negative correlation between TGM*
140 *concentration and ambient air temperature ($r = -0.08$) ($p < 0.05$). In addition, there are*
141 *several known Hg sources such as iron and steel manufacturing facilities including electric*
142 *and sintering furnaces using coking between the sampling site and the ocean.*

143 *As shown in Fig. 3 and Fig. S4, there was a weak but negative relationship between the*
144 *TGM concentrations and O₃ concentrations ($r = -0.18$) ($p < 0.01$), suggesting that oxidation*
145 *of GEM in the oxidizing atmosphere during periods of strong atmospheric mixing was*
146 *partially responsible for the diurnal variations of TGM concentrations. In addition, oxidation*
147 *of GEM by bromine species in the coastal area (Obrist et al., 2011) or by chloride radicals in*
148 *marine boundary layer (Laurier et al., 2003) might play a significant role. If oxidation of*
149 *GEM occurred, GOM concentrations would increase. However there are uncertainties on the*
150 *net effects on TGM (the sum of the GEM and the GOM) since we did not measure GOM*
151 *concentrations.*

152 *TGM concentration was negatively correlated with ambient air temperature ($r = -0.08$)*
153 *($p < 0.05$) because high ambient air temperature in the daytime will increase the height of*
154 *the boundary layer and dilute the TGM, and the relatively lower boundary layer at nighttime*
155 *could concentrate the TGM in the atmosphere (Li et al., 2011). Although there was a*
156 *statistically significant negative correlation between the TGM concentration and ambient air*
157 *temperature, there was a rapid increase in TGM concentration between 06:00-09:00 when*
158 *ambient temperatures also increased possibly due to local emissions related to industrial*
159 *activities, increased traffic, and activation of local surface emission sources. Similar patterns*
160 *were found in previous studies (Li et al., 2011; Stamenkovic et al., 2007). Nonparametric*
161 *correlations revealed that there is a weak positive correlation between TGM and ambient air*
162 *temperature ($r_s = 0.11$, $p=0.27$) between 06:00-09:00. The TGM concentration was*

163 negatively correlated with O_3 ($r_s = -0.33$, $p < 0.01$) but positively correlated with NO_2 ($r_s =$
164 0.21 , $p < 0.05$), suggesting that the increased traffic is the main source of TGM during these
165 time periods.

166 Compared to other seasons, significantly different diurnal variations of TGM were
167 observed in fall. The daytime TGM concentrations in fall were similar to those in other
168 seasons, however, the nighttime TGM concentrations in fall were much higher than other
169 seasons. As described earlier in Section 5.2, the high TGM concentrations in fall was
170 possibly due to the relationship between other pollutants and meteorological conditions as
171 well as different wind direction and sources. The nighttime TGM concentrations in fall were
172 simultaneously positively correlated with PM_{10} ($r=0.26$) ($p < 0.05$) and CO ($r=0.21$) ($p < 0.05$)
173 concentrations and wind speed ($r=0.35$) ($p < 0.01$), suggesting that the combustion process is
174 an important source during this period.

175 TGM generally showed a consistent increase in the early morning (06:00-09:00) and a
176 decrease in the afternoon (14:00-17:00), similar to previous studies (Dommergue et al.,
177 2002; Friedli et al., 2011; Li et al., 2011; Liu et al., 2011; Mao et al., 2008; Shon et al.,
178 2005; Song et al., 2009; Stamenkovic et al., 2007). Significantly different diurnal patterns
179 have been observed at many suburban sites with the daily maximum occurring in the
180 afternoon (12:00-15:00), possibly due to local emission sources and transport (Fu et al.,
181 2010; Fu et al., 2008; Kuo et al., 2006; Wan et al., 2009). Other studies in Europe reported
182 that TGM concentrations were relatively higher early in the morning or at night possibly due
183 to mercury emissions from surface sources that accumulated in the nocturnal inversion layer
184 (Lee et al., 1998; Schmolke et al., 1999).

185 Based on the above results, the diurnal variations in TGM concentration are due to a
186 combination of: 1) reactions with an oxidizing atmosphere, 2) changes in ambient temperature
187 and 3) local emissions related to industrial activities. To supplement these conclusions CPF
188 and CBPF were used to identify source directions and TPSCF was used to identify potential
189 source locations.”

190

191

192 **Comment 5**

193 Section 5.6 line 448: “It is difficult to discuss about the different seasonal patterns for CPF and
194 CBPF for TGM concentrations since there were no correlations between TGM and other
195 pollutants in spring, summer and fall except O_3 .” I don’t understand (and not mentioned in the

196 paper) why the correlation results are needed to interpret the seasonal patterns of CPF and
197 CBPF (Fig. S6). It wasn't needed to explain the overall CPF and CBPF results. Also, I'm still
198 skeptical whether CBPF provides more information about sources than CPF. Wind speed
199 dependency is included in CBPF to differentiate between ground level and stack emissions;
200 however, it's not discussed in the results. There should be a more detailed discussion of the
201 local ground level and stack emissions and uncertainties and disadvantages with the CBPF
202 method.

203

204 **Response 5**

205 In order to clarify, we have deleted the following sentence.

206

207 *“It is difficult to discuss about the different seasonal patterns for CPF and CBPF for TGM*
208 *concentrations since there were no correlations between TGM and other pollutants in spring,*
209 *summer and fall except O₃.”*

210

211 **Response to Anonymous Referee #2:**

212

213 **Comment 1**

214 Section 2 through 5 and conclusion are well improved. However, introduction section needs
215 more work. Please re-organize and add recent literature reviews that are related with this
216 research. Each paragraph contains one topic sentence. Please look at the introduction section.
217 What is the topic sentence for each paragraph?

218 **Response 1**

219 Thank you for your comments. As suggested, we carefully revised and edited including adding
220 recent literature reviews that are related with this research as follows on **Line 71-80, Line 91-**
221 **95, Line 109, Line 118-133** as shown below..

222

223 *“Mercury (Hg) is an environmental toxic and bioaccumulative trace metal whose*
224 *emissions to the environment have considerably increased due to anthropogenic activities*
225 *such as mining and combustion processes (Pirrone et al., 2013; Streets et al., 2011). Hg can*
226 *be globally distributed from the sources through atmospheric transport as gaseous elemental*
227 *form (Bullock et al., 1998; Mason and Sheu, 2002). However, the origins of atmospheric*
228 *mercury are local and regional (Choi et al., 2009) as well as hemispherical and global*
229 *(Durnford et al., 2010). In addition to the general background concentration of Hg in the*
230 *global atmosphere, local Hg emissions contribute to the Hg burden and it contribute to the*
231 *background concentration much of which represents anthropogenic releases accumulated*
232 *over the decades (UNEP, 2002).*

233 *Hg in the atmosphere exists in three major inorganic forms including gaseous elemental*
234 *mercury (GEM, Hg⁰), gaseous oxidized mercury (GOM, Hg²⁺) and particulate bound*
235 *mercury (PBM, Hg(p)). GEM which is the dominant form of Hg in ambient air, (>95%) has a*
236 *relatively long residence time (0.5~2 years) due to its low reactivity and solubility (Schroeder*
237 *and Munthe, 1998). However, GOM has high water solubility and relatively strong surface*
238 *adhesion properties (Han et al., 2005), so it has a short atmospheric residence time (~days).*
239 *PBM is associated with airborne particles such as dust, soot, sea-salt aerosols, and ice*
240 *crystals (Lu and Schroeder, 2004) and is likely produced, in part, by adsorption of GOM*
241 *species such as HgCl₂ onto atmospheric particles (Gauchard et al., 2005; Lu and Schroeder,*
242 *2004; Sakata and Marumoto, 2005; Seo et al., 2015).*

243 Atmospheric Hg released from natural (e.g., volcanoes, volatilization from aquatic and
244 terrestrial environments) (Pirrone et al., 2010; Strode et al., 2007) and anthropogenic
245 sources (e.g., coal combustion, cement production, ferrous and non-ferrous metals
246 manufacturing facilities, waste incineration and industrial boilers) (Pacyna et al., 2010;
247 Pacyna et al., 2006; Pacyna et al., 2003; Pirrone et al., 2010; Zhang et al., 2015) when
248 introduced into terrestrial and aquatic ecosystem through wet and dry deposition (Mason and
249 Sheu, 2002) can undergo various physical and chemical transformations before being
250 deposited. Its lifetime in the atmosphere depends on its reactivity and solubility so that,
251 depending on its form, it can have impacts on local, regional and global scales (Lin and
252 Pehkonen, 1999; Lindberg et al., 2007). A portion of the Hg deposited in terrestrial
253 environments through direct industrial discharge or atmospheric deposition is transported to
254 aquatic system through groundwater and surface water runoff (Miller et al., 2013). A
255 previous study also reported that Hg directly released into terrestrial and aquatic ecosystems
256 from industrial effluent has influenced surface water, sediment and biological tissue
257 (Flanders et al., 2010). Significant spatial variations in atmospheric Hg deposition near
258 urban and industrial areas are due to local anthropogenic sources including municipal waste
259 incinerators, medical waste incinerators, electric power generating facilities and cement
260 kilns (Dvonch et al., 1998), ferrous and non-ferrous metal processing, iron and steel
261 manufacturing facilities, oil and coal combustion (Hoyer et al., 1995), and other forms of
262 industrial combustion (Brown et al., 2015). Miller et al. (2013) also reported that local
263 sources of elemental Hg are typically industrial processes including retort facilities used in
264 the mercury mining industry to convert Hg containing minerals to elemental Hg and chlor-
265 alkali facilities.

266 The annual average national anthropogenic Hg emissions from South Korea in 2007 have
267 been estimated to be 12.8 tons (range 6.5 to 20.2 tons); the major emission sources are coal
268 combustion in thermal power plants (25.8%), oil refineries (25.5%), cement kilns (21%),
269 incinerators (19.3%) including sludge incinerators (4.7%), municipal waste incinerators
270 (MWIs) (3%), industrial waste incinerators (IWIs) (2.7%), hospital/medical/infectious waste
271 incinerators (HMIWIs) (8.8%), and iron manufacturing (7%) (Kim et al., 2010). Global
272 anthropogenic Hg emissions were estimated to be 1960 tons in 2010 with East and Southeast
273 Asia responsible for 777 tons (39.7%) (19.6 tons for Japan and 8.0 tons for South Korea)
274 (AMAP/UNEP, 2013). China is the largest Hg emitting country in the world, contributing

275 *more than 800 tons (~ 40%) of the total anthropogenic Hg emissions (UNEP, 2008).*

276 *Background atmospheric Hg concentrations in the northern hemisphere have decreased*
277 *since 1996 (Slemr et al., 2003), as measured at the Global Atmosphere Watch (GAW) station*
278 *at Mace Head, Ireland (Ebinghaus et al., 2011) and at the Canadian Atmospheric Mercury*
279 *Network (CAMNet) (Temme et al., 2007). In urban areas in South Korea atmospheric TGM*
280 *concentrations have also decreased over the last few decades due to the reduced fossil fuel*
281 *(mainly anthracite coal) consumption (Kim et al., 2016; Kim and Kim, 2000). However, this*
282 *decreasing trend is inconsistent with steady or increasing global anthropogenic Hg emissions*
283 *since 1990 in the northern hemisphere (Streets et al., 2011; Weigelt et al., 2015; Wilson et al.,*
284 *2010). A previous study reported that the global anthropogenic Hg emissions are increasing*
285 *with an average of 1.3% annual growth without including the artisanal and small-scale*
286 *production sector (Muntean et al., 2014).*

287 *Receptor models are often used to identify sources of air pollutants and are focused on*
288 *the pollutants behavior in the ambient environment at the point of impact (Hopke, 2003). In*
289 *previous studies, conditional probability function (CPF), which utilizes the local wind*
290 *direction, and potential source contribution function (PSCF), which utilizes longer backward*
291 *trajectories (typically 3-5 days), combined with concentration data were used to identify*
292 *possible transport pathways and source locations (Hopke, 2003). While PSCF has been used*
293 *primarily to identify regional sources, it has also been used to identify local sources (Hsu et*
294 *al., 2003).*

295 *The objectives of this study were to characterize the hourly and seasonal variations of*
296 *atmospheric TGM (the sum of the GEM and the GOM) concentrations, to identify the*
297 *relationships between TGM and co-pollutant concentrations, and to identify likely source*
298 *directions and locations of TGM using CPF, conditional bivariate probability function*
299 *(CBPF) and total PSCF (TPSCF).”*

300

301

302 **Comment 2**

303 The 2nd paragraph (line 81-84) could be moved.

304 **Response 2**

305 We have deleted the paragraph (Line 81-84) and combined with next paragraph as follows on

306 **Line 91-95.**

307

308 “Atmospheric Hg released from natural (e.g., volcanoes, volatilization from aquatic and
309 terrestrial environments) (Pirrone et al., 2010; Strode et al., 2007) and anthropogenic sources
310 (e.g., coal combustion, cement production, ferrous and non-ferrous metals manufacturing
311 facilities, waste incineration and industrial boilers) (Pacyna et al., 2010; Pacyna et al., 2006;
312 Pacyna et al., 2003; Pirrone et al., 2010; Zhang et al., 2015) when introduced into...”

313

314

315 **Comment 3**

316 The 3rd (line 85-92), the 4th (line 93-95), and 5th (line 96-103) paragraphs have same topics
317 so they could be combined into one.

318 **Response 3**

319 As suggested, the sentences were combined into one paragraph on **Line 91-112.**

320

321

322 **Comment 4**

323 In the 6th paragraph (line 104-109), the author talked about the Hg emissions in South Korea.
324 Is this urban areas or both urban and rural areas? Which year? How about other counties near
325 South Korea? (e.g., China, Japan?)

326 **Response 4**

327 Kim et al. (2010) reported that the annual **average national** (including urban and rural areas)
328 anthropogenic Hg emissions from South Korea **in 2007** have been estimated to be 12.8
329 tons **ranged from 6.5 to 20.2 tons.**

330

331 In response to this comment, we have rephrased the sentence as follows on **Line 113-114.**

332

333 “The annual average national anthropogenic Hg emissions from South Korea in 2007 have
334 been estimated to be 12.8 tons (range 6.5 to 20.2 tons);”

335

336 We added the sentence about Hg emissions from China and Japan as follows on **Line 118-122.**

337

338 “Global anthropogenic Hg emissions were estimated to be 1960 tons in 2010 with East and

339 Southeast Asia responsible for 777 tons (39.7%) (19.6 tons for Japan and 8.0 tons for South
340 Korea) (AMAP/UNEP, 2013). China is the largest Hg emitting country in the world,
341 contributing more than 800 tons (~ 40%) of the total anthropogenic Hg emissions (UNEP,
342 2008).”

343

344

345 **Comment 5**

346 Line 117-121: should be one paragraph

347 **Response 5**

348 As suggested, we separated two paragraphs as follows on **Line 142-146**.

349

350 “... While PSCF has been used primarily to identify regional sources, it has also been used to
351 identify local sources (Hsu et al., 2003).

352 The objectives of this study were to characterize the hourly and seasonal variations of
353 atmospheric TGM (the sum of the GEM and the GOM) concentrations, to identify the
354 relationships between TGM and co-pollutant concentrations, and to identify likely source
355 directions and locations of TGM using CPF, conditional bivariate probability function (CBPF)
356 and total PSCF (TPSCF).”

357

1 **Characteristics of total gaseous mercury (TGM) concentrations in an**
2 **industrial complex in southern Korea: Impacts from local sources**

3
4 Yong-Seok Seo^{1,2}, Seung-Pyo Jeong¹, Thomas M. Holsen³, Young-Ji Han⁴, Eunhwa Choi⁵, Eun
5 Ha Park¹, Tae Young Kim¹, Hee-Sang Eum¹, Dae Gun Park¹, Eunhye Kim⁶, Soontae Kim⁶,
6 Jeong-Hun Kim⁷, Jaewon Choi⁸, Seung-Muk Yi^{1,2,*}

7
8 ¹Department of Environmental Health, Graduate School of Public Health, Seoul National
9 University, 1 Gwanak, Gwanak-ro, Gwanak-gu, Seoul 151-742, South Korea

10
11 ²Institute of Health and Environment, Seoul National University, 1 Gwanak, Gwanak-ro,
12 Gwanak-gu, Seoul 151-742, South Korea

13
14 ³Department of Civil and Environmental Engineering, Clarkson University, Potsdam,
15 NY13699, USA

16
17 ⁴Department of Environmental Science, Kangwon National University, 192-1, Hyoja-2-dong,
18 Chuncheon, Kangwondo, 200-701, South Korea

19
20 ⁵Asian Institute for Energy, Environment & Sustainability, Seoul National University, 1
21 Gwanak-ro, Gwanak-gu, Seoul 151-742, South Korea

22
23 ⁶Department of Environmental, Civil and Transportation Engineering, Ajou University,
24 Woncheon-dong, Yeongtong-gu, Suwon, 443-749, South Korea

25
26 ⁷Division of Air Pollution Engineering, Department of Climate and Air Quality Research,
27 National Institute of Environmental Research, Hwangyong-ro 42, Seogu, Incheon, 404-708,
28 South Korea

29
30 ⁸University of Pennsylvania, Philadelphia, PA19104, USA

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32
33
34
35
36
37
38
39
40 *Address correspondence to Dr. Seung-Muk Yi, Graduate School of Public Health, Seoul
41 National University, 1 Gwanak, Gwanak-ro, Gwanak-gu, Seoul 151-742, South Korea
42 E-mail) yiseung@snu.ac.kr
43 Telephone) 82-2-880-2736
44 Fax) 82-2-745-9104

45 **Abstract**

46 Total gaseous mercury (TGM) concentrations were measured every 5 min in Pohang,
47 Gyeongsangbuk-do, Korea during summer (17 August~23 August 2012), fall (9 October~17
48 October 2012), winter (22 January ~29 January 2013), and spring (26 March~3 April 2013)
49 to: 1) characterize the hourly and seasonal variations of atmospheric TGM concentrations, 2)
50 identify the relationships between TGM and co-pollutants, and 3) identify likely source
51 directions and locations of TGM using conditional probability function (CPF), conditional
52 bivariate probability function (CBPF) and total potential source contribution function
53 (TPSCF).

54 The TGM concentration was statistically significantly highest in fall ($6.7 \pm 6.4 \text{ ng m}^{-3}$),
55 followed by spring ($4.8 \pm 4.0 \text{ ng m}^{-3}$), winter ($4.5 \pm 3.2 \text{ ng m}^{-3}$) and summer ($3.8 \pm 3.9 \text{ ng m}^{-3}$).
56 There was a weak but statistically significant negative correlation between the TGM
57 concentration and ambient air temperature ($r = -0.08$) ($p < 0.05$). Although the daytime
58 temperature ($14.7 \pm 10.0 \text{ }^\circ\text{C}$) was statistically significantly higher than that in the nighttime
59 ($13.0 \pm 9.8 \text{ }^\circ\text{C}$) ($p < 0.05$), the daytime TGM concentration ($5.3 \pm 4.7 \text{ ng m}^{-3}$) was statistically
60 significantly higher than those in the nighttime ($4.7 \pm 4.7 \text{ ng m}^{-3}$) ($p < 0.01$), possibly due to
61 local emissions related to industrial activities and activation of local surface emission
62 sources. The observed $\Delta\text{TGM}/\Delta\text{CO}$ was significantly lower than that of Asian long-range
63 transport, but similar to that of local sources in Korea and in US industrial events suggesting
64 that local sources are more important than that of long-range transport. CPF, CBPF and
65 TPSCF indicated that the main sources of TGM were iron and manufacturing facilities, the
66 hazardous waste incinerators and the coastal areas.

67 **Keywords:** Total gaseous mercury (TGM); co-pollutant; conditional probability function
68 (CPF); conditional bivariate probability function (CBPF); total potential source contribution
69 function (TPSCF)

70 1. Introduction

71 Mercury (Hg) is an environmental toxic and bioaccumulative trace metal whose emissions
72 to the environment have considerably increased due to anthropogenic activities such as
73 mining and combustion processes (Pirrone et al., 2013; Streets et al., 2011). Hg can be
74 globally distributed from the sources through atmospheric transport as gaseous elemental
75 form (Bullock et al., 1998; Mason and Sheu, 2002). However, the origins of atmospheric
76 mercury are local and regional (Choi et al., 2009) as well as hemispherical and global
77 (Durnford et al., 2010). In addition to the general background concentration of Hg in the
78 global atmosphere, local Hg emissions contribute to the Hg burden and it contribute to the
79 background concentration much of which represents anthropogenic releases accumulated
80 over the decades (UNEP, 2002).

81 Hg in the atmosphere exists in three major inorganic forms including gaseous elemental
82 mercury (GEM, Hg^0), gaseous oxidized mercury (GOM, Hg^{2+}) and particulate bound
83 mercury (PBM, $\text{Hg}(p)$). GEM which is the dominant form of Hg in ambient air, (>95%) has a
84 relatively long residence time (0.5~2 years) due to its low reactivity and solubility (Schroeder
85 and Munthe, 1998). However, GOM has high water solubility and relatively strong surface
86 adhesion properties (Han et al., 2005), so it has a short atmospheric residence time (~days).
87 PBM is associated with airborne particles such as dust, soot, sea-salt aerosols, and ice crystals
88 (Lu and Schroeder, 2004) and is likely produced, in part, by adsorption of GOM species such
89 as HgCl_2 onto atmospheric particles (Gauchard et al., 2005; Lu and Schroeder, 2004; Sakata
90 and Marumoto, 2005; Seo et al., 2012; Seo et al., 2015).

91 Atmospheric Hg released from natural (e.g., volcanoes, volatilization from aquatic and
92 terrestrial environments) (Pirrone et al., 2010; Strode et al., 2007) and anthropogenic sources
93 (e.g., coal combustion, cement production, ferrous and non-ferrous metals manufacturing

94 facilities, waste incineration and industrial boilers) (Pacyna et al., 2010; Pacyna et al., 2006;
95 Pacyna et al., 2003; Pirrone et al., 2010; Zhang et al., 2015) when introduced into terrestrial
96 and aquatic ecosystem through wet and dry deposition (Mason and Sheu, 2002) can undergo
97 various physical and chemical transformations before being deposited. Its lifetime in the
98 atmosphere depends on its reactivity and solubility so that, depending on its form, it can have
99 impacts on local, regional and global scales (Lin and Pehkonen, 1999; Lindberg et al., 2007).
100 A portion of the Hg deposited in terrestrial environments through direct industrial discharge
101 or atmospheric deposition is transported to aquatic system through groundwater and surface
102 water runoff (Miller et al., 2013). A previous study also reported that Hg directly released
103 into terrestrial and aquatic ecosystems from industrial effluent has influenced surface water,
104 sediment and biological tissue (Flanders et al., 2010). Significant spatial variations in
105 atmospheric Hg deposition near urban and industrial areas are due to local anthropogenic
106 sources including municipal waste incinerators, medical waste incinerators, electric power
107 generating facilities and cement kilns (Dvonch et al., 1998), ferrous and non-ferrous metal
108 processing, iron and steel manufacturing facilities, oil and coal combustion (Hoyer et al.,
109 1995), and other forms of industrial combustion (Brown et al., 2015). Miller et al. (2013) also
110 reported that local sources of elemental Hg are typically industrial processes including retort
111 facilities used in the mercury mining industry to convert Hg containing minerals to elemental
112 Hg and chlor-alkali facilities.

113 The annual average national anthropogenic Hg emissions from South Korea in 2007 have
114 been estimated to be 12.8 tons (range 6.5 to 20.2 tons); the major emission sources are coal
115 combustion in thermal power plants (25.8%), oil refineries (25.5%), cement kilns (21%),
116 incinerators (19.3%) including sludge incinerators (4.7%), municipal waste incinerators
117 (MWIs) (3%), industrial waste incinerators (IWIs) (2.7%), hospital/medical/infectious waste

118 incinerators (HMIWIs) (8.8%), and iron manufacturing (7%) (Kim et al., 2010). Global
119 anthropogenic Hg emissions were estimated to be 1960 tons in 2010 with East and Southeast
120 Asia responsible for 777 tons (39.7%) (19.6 tons for Japan and 8.0 tons for South Korea)
121 (AMAP/UNEP, 2013). China is the largest Hg emitting country in the world, contributing
122 more than 800 tons (~ 40%) of the total anthropogenic Hg emissions (UNEP, 2008).

123 Background atmospheric Hg concentrations in the northern hemisphere have decreased
124 since 1996 (Slemr et al., 2003), as measured at the Global Atmosphere Watch (GAW) station
125 at Mace Head, Ireland (Ebinghaus et al., 2011) and at the Canadian Atmospheric Mercury
126 Network (CAMNet) (Temme et al., 2007). In urban areas in South Korea atmospheric TGM
127 concentrations have also decreased over the last few decades due to the reduced fossil fuel
128 (mainly anthracite coal) consumption (Kim et al., 2016; Kim and Kim, 2000). However, this
129 decreasing trend is inconsistent with steady or increasing global anthropogenic Hg emissions
130 since 1990 in the northern hemisphere (Streets et al., 2011; Weigelt et al., 2015; Wilson et al.,
131 2010). A previous study reported that the global anthropogenic Hg emissions are increasing
132 with an average of 1.3% annual growth without including the artisanal and small-scale
133 production sector (Muntean et al., 2014).

134 Receptor models are often used to identify sources of air pollutants and are focused on the
135 pollutants behavior in the ambient environment at the point of impact (Hopke, 2003). In
136 previous studies, conditional probability function (CPF), which utilizes the local wind
137 direction, and potential source contribution function (PSCF), which utilizes longer backward
138 trajectories (typically 3-5 days), combined with concentration data were used to identify
139 possible transport pathways and source locations (Hopke, 2003). While PSCF has been used
140 primarily to identify regional sources, it has also been used to identify local sources (Hsu et
141 al., 2003).

142 The objectives of this study were to characterize the hourly and seasonal variations of
143 atmospheric TGM (the sum of the GEM and the GOM) concentrations, to identify the
144 relationships between TGM and co-pollutant concentrations, and to identify likely source
145 directions and locations of TGM using CPF, conditional bivariate probability function
146 (CBPF) and total PSCF (TPSCF).

147

148 **2. Materials and methods**

149 *2.1. Sampling and analysis*

150 TGM concentrations were measured on the roof of the Korean Federation of
151 Community Credit Cooperatives (KFCCC) building (latitude: 35.992°, longitude: 129.404°,
152 ~10 m above ground) in Pohang city, in Gyeongsangbuk-do, a province in eastern South
153 Korea. Gyeongsangbuk-do has a population of 2.7 million (5% of the total population and the
154 third most populated province in South Korea) and an area of 19,030 km² (19% of the total
155 area of South Korea and the largest province geographically in South Korea). Pohang city has
156 a population of 500,000 (1% of the total population in South Korea) and an area of 605.4 km²
157 (1.1% of the total area in South Korea). It is heavily industrialized with the third largest steel
158 manufacturing facility in Asia and the fifth largest in the world. There are several iron and
159 steel manufacturing facilities including electric and sintering furnaces using coking in
160 Gyeongsangbuk-do including Pohang. In addition, there are several coke plants around the
161 sampling site. The Hyungsan River divides the city into a residential area and the steel
162 complex. Hg emissions data from iron and steel manufacturing, and a hazardous waste
163 incinerator were estimated based on a previous study (Kim et al., 2010) (Fig. 1).

164 TGM concentrations were measured every 5 min during summer (17 August~23 August
165 2012), fall (9 October~17 October 2012), winter (22 January ~29 January 2013), and spring

166 (26 March~3 April 2013) using a mercury vapor analyzer (Tekran 2537B) which has two
167 gold cartridges that alternately collect and thermally desorb mercury. Ambient air at a flow
168 rate of 1.5 L min^{-1} was transported through a 3 m-long heated sampling line (1/4" OD Teflon)
169 in to the analyzer. The sampling line was heated at about $50 \text{ }^\circ\text{C}$ using heat tape to prevent
170 water condensation in the gold traps because moisture on gold surfaces interferes with the
171 amalgamation of Hg (Keeler and Barres, 1999). Particulate matter was removed from the
172 sampling line by a 47 mm Teflon filter.

173

174 2.2. Meteorological data

175 Hourly meteorological data (air temperature, relative humidity, and wind speed and
176 direction) were obtained from the Automatic Weather Station (AWS) operated by the Korea
177 Meteorological Administration (KMA) (<http://www.kma.go.kr>) (6 km from the site). Hourly
178 concentrations of NO_2 , O_3 , CO , PM_{10} and SO_2 were obtained from the National Air Quality
179 Monitoring Network (NAQMN) (3 km from the site) (Fig. 1).

180 Meteorological Setting. Fig. S1 shows the frequency of counts of measured wind direction
181 occurrence by season during the sampling period. The predominant wind direction at the
182 sampling site was W (20.9%) and WS (19.2%), and calm conditions of wind speed less than
183 1 m s^{-1} occurred 7.6% of the time. Compared to other seasons, however, the prevailing winds
184 in summer were N (17.0%), NE (16.4%), S (16.4%), and SW (15.8%).

185

186 2.3. QA/QC

187 Automated daily calibrations were carried out for the Tekran 2537B using an internal
188 permeation source. Two-point calibrations (zero and span) were separately performed for
189 each gold cartridge. Manual injections were performed prior to every field sampling

190 campaign to evaluate these automated calibrations using a saturated mercury vapor standard.
 191 The relative percent difference (RPD) between automated calibrations and manual injections
 192 was less than 2%. The recovery measured by directly injecting known amounts of four
 193 mercury vapor standards when the sample line was connected to zero air ranged from 92 to
 194 110% ($99.4 \pm 5.2\%$ in average).

195

196 3. Model descriptions

197 3.1. Conditional Probability Function (CPF)

198 CPF was originally performed to determine which wind directions dominate during high
 199 concentration events to evaluate local source impacts (Ashbaugh et al., 1985). It has been
 200 successfully used in many previous studies (Begum et al., 2004; Kim et al., 2003a; Kim et al.,
 201 2003b; Xie and Berkowitz, 2006; Zhao et al., 2004; Zhou et al., 2004). CPF estimates the
 202 probability that the measured concentration will exceed the threshold criterion for a given
 203 wind direction. The CPF is defined as follows Eq. (1).

204

$$205 \quad CPF_{\Delta\theta} = \frac{m_{\Delta\theta|C \geq x}}{n_{\Delta\theta}} \quad (1)$$

206

207 where, $m_{\Delta\theta}$ is the number of samples from the wind sector θ having concentration C greater
 208 than or equal to a threshold value x , and $n_{\Delta\theta}$ is the total number of samples from wind sector
 209 $\Delta\theta$. In this study, 16 sectors ($\Delta\theta = 22.5^\circ$) were used and calm winds ($\leq 1 \text{ m s}^{-1}$) were excluded
 210 from the analysis. The threshold criterion was set at above the overall average TGM
 211 concentration (5.0 ng m^{-3}). Thus, CPF indicates the potential for winds from a specific
 212 direction to contribute to high air pollution concentrations.

213

214 *3.2. Conditional Bivariate Probability Function (CBPF)*

215 CBPF couples ordinary CPF with wind speed as a third variable, allocating the measured
 216 concentration of pollutant to cells defined by ranges of wind direction and wind speed rather
 217 than to only wind direction sectors.

218 The CBPF is defined as follows Eq. (2).

219

$$220 \quad CBPF_{\Delta\theta,\Delta u} = \frac{m_{\Delta\theta,\Delta u|C \geq x}}{n_{\Delta\theta,\Delta u}} \quad (2)$$

221

222 where, $m_{\Delta\theta,\Delta u}$ is the number of samples in the wind sector $\Delta\theta$ with wind speed interval Δu
 223 having concentration C greater than a threshold value x , and $n_{\Delta\theta,\Delta u}$ is the total number of
 224 samples in that wind direction-speed interval. The threshold criterion was set at above the
 225 overall average TGM concentration (5.0 ng m^{-3}). [The extension to the bivariate case can](#)
 226 [provide more information on the nature of the sources because different source types such as](#)
 227 [stack emission sources and ground-level sources can have different wind speed dependencies](#)
 228 [\(prominent at high and low wind speed, respectively\).](#) More detailed information is described
 229 in a previous study (Uria-Tellaetxe and Carslaw, 2014).

230

231 *3.3. Potential Source Contribution Function (PSCF)*

232 The PSCF model has been extensively and successfully used in the previous studies to
 233 identify the likely source areas (Cheng et al., 1993; Han et al., 2004; Hopke et al., 2005; Lai
 234 et al., 2007; Lim et al., 2001; Poissant, 1999; Zeng and Hopke, 1989). The PSCF is a simple
 235 method that links residence time in upwind areas with high concentrations through a
 236 conditional probability field and was originally developed by Ashbaugh et al. (1985). PSCF_{ij}

237 is the conditional probability that an air parcel that passed through the ij th cell had a high
238 concentration upon arrival at the monitoring site and is defined as the following Eq. (3).

239

$$240 \quad PSCF_{ij} = \frac{m_{ij}}{n_{ij}} \quad (3)$$

241

242 where, n_{ij} is the number of trajectory segment endpoints that fall into the ij -th cell, and m_{ij} is the
243 number of segment endpoints in the same grid cell (ij -th cell) when the concentrations are higher
244 than a criterion value as measured at the sampling site.

245 High PSCF values in those grid cells are regarded as possible source locations. Cells including
246 emission sources can be identified with conditional probabilities close to one if trajectories that
247 have crossed the cells efficiently transport the released pollutant to the receptor site. Therefore,
248 the PSCF model provides a tool to map the source potentials of geographical areas.

249 The criterion value of PSCF for TGM concentration was set at above the overall average
250 concentration (5.0 ng m^{-3}) to identify the emission sources associated with high TGM
251 concentrations and provide a better estimation and resolution of source locations during the
252 sampling periods. The geographic area covered by the computed trajectories was divided into
253 an array of 0.05° latitude by 0.05° longitude grid cells. As will be discussed in Section 5.3, 24
254 h backward trajectories starting at every hour at a height of 10, 50, and 100 m above ground
255 level were computed using the vertical velocity model because local sources are more
256 important than that of long-range transport in this study (It should be noted that PSCF results
257 using 48 h backward trajectories had similar results as the 24 h backward trajectories). Each
258 trajectory was terminated if they exit the model top (5,000m), but advection continues along
259 the surface if trajectories intersect the ground. To generate horizontally highly resolved
260 meteorological inputs for trajectory calculations, the Weather Research and Forecast (WRF)

261 model was used to generate a coarse domain at a resolution of 27 km and a nested domain at
262 a horizontal resolution of 9 km, which geographically covers northeast Asia and the southern
263 part of the Korean Peninsula, respectively. The nested domain has 174 columns in the east-
264 west direction and 114 rows in the north-south direction. PSCF was calculated with 9 km
265 meteorological data.

266 In this study, TPSCF which incorporates probability from above different starting
267 heights was calculated since backward trajectories starting at different heights traverse
268 different distances and pathways, thus providing information that cannot be obtained from a
269 single starting height (Cheng et al., 1993).

270 Previous studies suggest that there are increasing uncertainties as backward trajectory
271 distances increase (Stohl et al., 2002) and that PSCF modeling is prone to the trailing effect is
272 which locations upwind of sources are also identified as potential sources (Han et al., 2004).
273 An alternative to back trajectory calculations in the interpretation of atmospheric trace
274 substance measurements (Stohl et al., 2002) although this technique does not provide much
275 information on source locations.

276 Generally, PSCF results show that the potential sources covered wide areas instead of
277 indicating individual sources due to the trailing effect. The trailing effect appears since PSCF
278 distributes a constant weight along the path of the trajectories. To minimize the effect of
279 small n_{ij} (the number of trajectory segment endpoints that fall into the ij -th cell) values,
280 resulting in high TPSCF values with high uncertainties, an arbitrary weight function $W(n_{ij})$
281 was applied to down-weight the PSCF values for the cell in which the total number of end
282 points was less than three times the average value of the end points (Choi et al., 2011; Heo et
283 al., 2009; Hopke et al., 1995; Polissar et al., 2001). The TPSCF value for a grid cell was
284 defined with following Eq. (4).

285

286

$$P(TPSCF_{ij}) = \frac{P(m_{ij})_{10m} + P(m_{ij})_{50m} + P(m_{ij})_{100m}}{P(n_{ij})_{10m} + P(n_{ij})_{50m} + P(n_{ij})_{100m}} \times W \quad (4)$$

287

288 where,

289

$$W(n_{ij}) = \begin{cases} 1.0, & 3n_{ave} < n_{ij} \\ 0.8, & 2n_{ave} < n_{ij} \leq 3n_{ave} \\ 0.6, & n_{ave} < n_{ij} \leq 2n_{ave} \\ 0.4, & 0.5n_{ave} < n_{ij} \leq n_{ave} \\ 0.2, & n_{ij} \leq 0.5n_{ave} \end{cases}$$

290

291 4. Clean Air Policy Support System (CAPSS) data

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In this study, the Korean National Emission Inventory estimated using Clean Air Policy Support System (CAPSS) data developed by the National Institute of Environmental Research (NIER) were used (<http://airemiss.nier.go.kr/main.jsp> (accessed December 09, 2015)). The CAPSS is the national emission inventory system for the air pollutants (CO, NO_x, SO_x, TSP, PM₁₀, PM_{2.5}, VOCs and NH₃) which utilizes various national, regional and local statistical data collected from about 150 organizations in Korea. In CAPSS, the Source Classification Category (SCC) excluding fugitive dust and biomass burning based on the European Environment Agency's (EEA) CORE Inventory of AIR emissions was classified into the following four levels (EMEP/CORINAIR) (NIER, 2011).

301

(1) The upper level (SCC1): 11 source categories ,

302

(2) The intermediate level (SCC2): 42 source categories and

303

(3) The lower level (SCC3): 173 source categories

304

305 The sectoral contributions of emissions of South Korea, Gyeongsangbuk-do and Pohang
306 for CO, NO_x, SO_x, TSP, PM₁₀, PM_{2.5}, VOC and NH₃ are shown in Fig. S2 (See SI for
307 details).

308 More detailed information about SCCs in CAPSS is described in Table S1.

309

310 5. Results and Discussions

311 5.1. General characteristics of TGM

312 The seasonal distributions of TGM were characterized by large variability during each
313 sampling period (Fig. 2). The average concentration of TGM during the complete sampling
314 period was $5.0 \pm 4.7 \text{ ng m}^{-3}$ (range: 1.0-79.6 ng m^{-3}). This is significantly higher than the
315 Northern Hemisphere background concentration ($\sim 1.5 \text{ ng m}^{-3}$) (Sprovieri et al., 2010) and
316 those measured in China, in Japan and other locations in Korea, however lower than those
317 measured at Changchun, Gui Yang and Nanjing in China (Table 1). The median TGM
318 concentration was 3.6 ng m^{-3} which was much lower than that of the average, suggesting that
319 there were some extreme pollution episodes with very high TGM concentrations.

320 The TGM concentration follows a typical log-normal distribution (Fig. S3). The range of 2
321 to 5 ng m^{-3} dominated the distribution, accounting for more than half of the total number of
322 samples (60.8%). The maximum frequency of 28.1% occurred between 2 and 3 ng m^{-3} .
323 Extremely high TGM concentration events ($>20 \text{ ng m}^{-3}$) were also observed (1.7% of the
324 time).

325

326

327

328 5.2. Seasonal variations

329 The TGM concentration was statistically significantly higher in fall ($6.7 \pm 6.4 \text{ ng m}^{-3}$) ($p <$
330 0.01), followed by spring ($4.8 \pm 4.0 \text{ ng m}^{-3}$), winter ($4.5 \pm 3.2 \text{ ng m}^{-3}$) and summer (3.8 ± 3.9
331 ng m^{-3}) (Table 2). The highest concentrations ($\text{TGM} > 10 \text{ ng m}^{-3}$) were measured more
332 frequently in fall (24.7%), and the lowest concentrations ($\text{TGM} < 3 \text{ ng m}^{-3}$) mainly occurred
333 in summer (49.7%). The low TGM concentration in summer is likely because increased
334 mixing height (Friedli et al., 2011), and gas phase oxidation (Choi et al., 2013; Huang et al.,
335 2010; Lynam and Keeler, 2006) at higher temperatures particularly at this sampling site
336 which is close to the ocean (2 km) where oxidation involving halogens may be enhanced
337 (Holmes et al., 2009; Lin et al., 2006). The high TGM concentrations in fall was due to
338 different wind direction (see Fig. S1), sources, relationships with other pollutants and
339 meteorological conditions. More detailed information can be found in Section 5.4.

340 The average concentrations of NO_2 , O_3 , CO , PM_{10} and SO_2 during the complete sampling
341 period were $23.1 \pm 10.8 \text{ ppbv}$, $24.6 \pm 12.5 \text{ ppbv}$, $673.7 \pm 487.3 \text{ ppbv}$, $55.5 \pm 26.4 \mu\text{g m}^{-3}$ and
342 $6.7 \pm 4.3 \text{ ppbv}$, respectively. NO_2 , O_3 , CO , PM_{10} and SO_2 concentrations were highest in
343 spring (Table 2). There was a statistically significant positive correlation between the TGM
344 and PM_{10} ($r = 0.10$) ($p < 0.01$). However, the TGM concentration was not significantly
345 correlated with NO_2 , CO or SO_2 concentrations, suggesting that combustion associated with
346 space heating was not a significant source of TGM (Choi et al., 2009).

347

348 5.3. Relationship between TGM and CO

349 CO has a significant anthropogenic source and is considered to be an indicator of
350 anthropogenic emissions (Mao et al., 2008). Previous studies reported that TGM and CO

351 have a strong correlation because they have similar emission sources (combustion processes)
352 and similar long atmospheric residence times (Weiss-Penzias et al., 2003).

353 There was a weak positive correlation between TGM and CO in this study ($r = 0.04$) ($p =$
354 0.27). However there was a statistically significant correlation between TGM and CO in
355 winter ($r = 0.25$) ($p < 0.05$), suggesting that TGM and CO were affected by similar, possibly
356 distant, anthropogenic emission sources in winter.

357 On the other hand, there were no statistically significant correlations between TGM and
358 CO in spring ($r = 0.02$) ($p = 0.78$), in summer ($r = 0.13$) ($p = 0.08$), or in fall ($r = -0.03$) ($p =$
359 0.69), indicating that TGM and CO were affected by different anthropogenic emission
360 sources in these seasons.

361 Previous studies identified the long-range transport of mercury using the $\Delta\text{TGM}/\Delta\text{CO}$
362 enhancement ratio (Choi et al., 2009; Jaffe et al., 2005; Kim et al., 2009; Weiss-Penzias et al.,
363 2003; Weiss-Penzias et al., 2006). Kim et al. (2009) and Choi et al. (2009) investigated high
364 concentration events which were defined as at least a 10 h period with hourly average TGM
365 and CO concentrations higher than the average monthly TGM and CO concentrations. They
366 reported that long-range transport events were characterized by high values of TGM/CO ratio
367 ($\Delta\text{TGM}/\Delta\text{CO}$) (0.0052 - $0.0158 \text{ ng m}^{-3} \text{ ppb}^{-1}$) and high correlations ($r^2 > 0.5$), whereas local
368 events showed low $\Delta\text{TGM}/\Delta\text{CO}$ ($0.0005 \text{ ng m}^{-3} \text{ ppb}^{-1}$ in average) and weak correlations ($r^2 <$
369 0.5).

370 The observed $\Delta\text{TGM}/\Delta\text{CO}$ was $0.0001 \text{ ng m}^{-3} \text{ ppb}^{-1}$ in spring, $0.0005 \text{ ng m}^{-3} \text{ ppb}^{-1}$ in
371 summer, $-0.0007 \text{ ng m}^{-3} \text{ ppb}^{-1}$ in fall, $0.0011 \text{ ng m}^{-3} \text{ ppb}^{-1}$ in winter, which are significantly
372 lower than that indicative of Asian long-range transport (0.0046 - $0.0056 \text{ ng m}^{-3} \text{ ppb}^{-1}$) (Friedli

373 et al., 2004; Jaffe et al., 2005; Weiss-Penzias et al., 2006), suggesting that local sources are
374 more important than that of long-range transport in this study. The $\Delta\text{TGM}/\Delta\text{CO}$ in winter
375 ($0.0011 \text{ ng m}^{-3} \text{ ppb}^{-1}$) was similar to that of a site impacted by local sources in Korea (Kim et
376 al., 2009) and in US industrially related events ($0.0011 \text{ ng m}^{-3} \text{ ppb}^{-1}$) (Weiss-Penzias et al.,
377 2007).

378 There are also uncertainties from the potential mixing between Hg associated with long-
379 range transported airflows and local air making it difficult to distinguish between distant and
380 local source impacts. However, it is possible that the one-week sampling period in each
381 season did not capture the long-range transport events, and more can be learned using a larger
382 dataset than just using the one-week sampling period to confirm these results.

383

384 5.4. Diurnal variations

385 Diurnal variations of TGM (Fig. 3), co-pollutants concentrations, and meteorological
386 data were observed (Fig. S4). TGM, O_3 , CO, SO_2 , and temperature in the daytime (06:00-
387 18:00) were higher than those in the nighttime (18:00-06:00) ($p < 0.05$) except PM_{10} ($p =$
388 0.09) (Fig. S5). However, NO_2 during the nighttime because of relatively lower
389 photochemical reactivity with O_3 was higher than that in daytime ($p < 0.05$) (Adame et al.,
390 2012).

391 The daytime TGM concentration ($5.3 \pm 4.7 \text{ ng m}^{-3}$) was higher than that in the nighttime
392 ($4.7 \pm 4.7 \text{ ng m}^{-3}$) ($p < 0.01$), which was similar to several previous studies (Cheng et al.,
393 2014; Gabriel et al., 2005; Nakagawa, 1995; Stamenkovic et al., 2007) but different than
394 another studies (Lee et al., 1998). Previous studies reported that this different is due to local
395 sources close to the sampling site (Cheng et al., 2014; Gabriel et al., 2005), a positive
396 correlation between TGM concentration and ambient air temperature (Nakagawa, 1995) and

397 increased traffic (Stamenkovic et al., 2007). However, another study suggested that the higher
398 TGM concentration during the night was due to the shallowing of the boundary layer, which
399 concentrated the TGM near the surface (Lee et al., 1998).

400 In a previous study the daytime TGM concentration was relatively lower than that in the
401 nighttime because the sea breeze transported air containing low amounts of TGM from the
402 ocean during the daytime whereas the land breeze transported air containing relatively high
403 concentrations of TGM from an urban area during the nighttime (Kellerhals et al., 2003).
404 Although it is possible that the land-sea breeze may affect diurnal variations in TGM
405 concentrations since the sampling site was near the ocean and lower TGM were also observed
406 during the daytime, the higher concentrations in the daytime than those in nighttime were due
407 to local emission sources because the daytime temperature (14.7 ± 10.0 °C) was statistically
408 significantly higher than that in the nighttime (13.0 ± 9.8 °C) (t-test, $p < 0.05$) and there was a
409 weak but statistically significant negative correlation between TGM concentration and
410 ambient air temperature ($r = -0.08$) ($p < 0.05$). In addition, there are several known Hg
411 sources such as iron and steel manufacturing facilities including electric and sintering
412 furnaces using coking between the sampling site and the ocean.

413 As shown in Fig. 3 and Fig. S4, there was a weak but negative relationship between the
414 TGM concentrations and O₃ concentrations ($r = -0.18$) ($p < 0.01$), suggesting that oxidation
415 of GEM in the oxidizing atmosphere during periods of strong atmospheric mixing was
416 partially responsible for the diurnal variations of TGM concentrations. In addition, oxidation
417 of GEM by bromine species in the coastal area (Obrist et al., 2011) or by chloride radicals in
418 marine boundary layer (Laurier et al., 2003) might play a significant role. If oxidation of
419 GEM occurred, GOM concentrations would increase. However there are uncertainties on the

420 net effects on TGM (the sum of the GEM and the GOM) since we did not measure GOM
421 concentrations.

422 TGM concentration was negatively correlated with ambient air temperature ($r = -0.08$)
423 ($p < 0.05$) because high ambient air temperature in the daytime will increase the height of the
424 boundary layer and dilute the TGM, and the relatively lower boundary layer at nighttime
425 could concentrate the TGM in the atmosphere (Li et al., 2011). Although there was a
426 statistically significant negative correlation between the TGM concentration and ambient air
427 temperature, there was a rapid increase in TGM concentration between 06:00-09:00 when
428 ambient temperatures also increased possibly due to local emissions related to industrial
429 activities, increased traffic, and activation of local surface emission sources. Similar patterns
430 were found in previous studies (Li et al., 2011; Stamenkovic et al., 2007). Nonparametric
431 correlations revealed that there is a weak positive correlation between TGM and ambient air
432 temperature ($r_s = 0.11$, $p=0.27$) between 06:00-09:00. The TGM concentration was
433 negatively correlated with O_3 ($r_s = -0.33$, $p<0.01$) but positively correlated with NO_2 ($r_s =$
434 0.21 , $p<0.05$), suggesting that the increased traffic is the main source of TGM during these
435 time periods.

436 Compared to other seasons, significantly different diurnal variations of TGM were
437 observed in fall. The daytime TGM concentrations in fall were similar to those in other
438 seasons, however, the nighttime TGM concentrations in fall were much higher than other
439 seasons. As described earlier in Section 5.2, the high TGM concentrations in fall was
440 possibly due to the relationship between other pollutants and meteorological conditions as
441 well as different wind direction and sources. The nighttime TGM concentrations in fall were
442 simultaneously positively correlated with PM_{10} ($r=0.26$) ($p<0.05$) and CO ($r=0.21$) ($p<0.05$)

443 concentrations and wind speed ($r=0.35$) ($p<0.01$), suggesting that the combustion process is
444 an important source during this period.

445 TGM generally showed a consistent increase in the early morning (06:00-09:00) and a
446 decrease in the afternoon (14:00-17:00), similar to previous studies (Dommergue et al., 2002;
447 Friedli et al., 2011; Li et al., 2011; Liu et al., 2011; Mao et al., 2008; Shon et al., 2005; Song
448 et al., 2009; Stamenkovic et al., 2007). Significantly different diurnal patterns have been
449 observed at many suburban sites with the daily maximum occurring in the afternoon (12:00-
450 15:00), possibly due to local emission sources and transport (Fu et al., 2010; Fu et al., 2008;
451 Kuo et al., 2006; Wan et al., 2009). Other studies in Europe reported that TGM
452 concentrations were relatively higher early in the morning or at night possibly due to mercury
453 emissions from surface sources that accumulated in the nocturnal inversion layer (Lee et al.,
454 1998; Schmolke et al., 1999).

455 Based on the above results, the diurnal variations in TGM concentration are due to a
456 combination of: 1) reactions with an oxidizing atmosphere, 2) changes in ambient
457 temperature and 3) local emissions related to industrial activities. To supplement these
458 conclusions CPF and CBPF were used to identify source directions and TPSCF was used to
459 identify potential source locations.

460

461 *5.5. CPF, CBPF and TPSCF results of TGM*

462 Conventional CPF, CBPF and TPSCF plots for TGM concentrations higher than the
463 average concentration show high source probabilities to the west in the direction of large steel
464 manufacturing facilities and waste incinerators (Fig. 4). The CPF only shows high
465 probabilities from the west and provides no further information, however, the CBPF shows
466 groups of sources with the high probabilities from the west and the northeast. CBPF shows

467 that the high probabilities from the west occurred under high wind speed ($> 3 \text{ m s}^{-1}$)
468 indicative of emissions from stacks as well as low wind speed ($\leq 3 \text{ m s}^{-1}$) indicative of non-
469 buoyant ground level sources (Uria-Tellaetxe and Carslaw, 2014).

470 As described in Section 5.3, correlations between TGM and CO revealed that TGM and
471 CO were affected by similar anthropogenic emission sources in winter but affected by
472 different sources in spring, summer and fall, which is supported by Fig. S6 which shows
473 significantly different seasonal patterns of CPF and CBPF for TGM concentrations.
474 However, compared to Fig. 4, the CPF and CBPF patterns in fall were similar to those during
475 the whole sampling periods. Especially, the nighttime TGM concentration in fall was
476 simultaneously positively correlated with PM_{10} ($r=0.26$) ($p<0.05$) and CO ($r=0.21$) ($p<0.05$)
477 concentrations and wind speed ($r=0.35$) ($p<0.01$), indicating that the combustion process
478 from the west is an important source during this period.

479 Since TGM showed a significant correlation with CO ($r=0.25$) ($p<0.05$) and showed a
480 weak positive correlation with PM_{10} ($r=0.08$) ($p=0.33$) in winter with high wind speed,
481 combustion sources from the west are likely partially responsible for this result.

482 TPSCF identified the likely sources of TGM as the iron and manufacturing facilities and
483 the hazardous waste incinerators which are located to the west from the sampling site. A
484 previous study reported that the waste incinerators (9%) and iron and steel manufacturing
485 (7%) were relatively high Hg emissions sources in Korea (Kim et al., 2010). Waste
486 incinerators emissions were due to the high Hg content in the waste (Lee et al., 2004).
487 Emissions from iron and steel manufacturing are due to the numerous electric and sintering
488 furnaces using coking which emits relatively high mercury concentrations (Lee et al., 2004)
489 in Gyeongsangbuk-do including Pohang. There are several coke plants around the sampling
490 site (http://www.poscoenc.com/upload/W/BUSINESS/PDF/ENG_PLANT_2_1_3_5.pdf)

491 (accessed December 09, 2015)). They are essential parts of the iron and steel manufacturing,
492 and the major source of atmospheric mercury related to the iron and steel manufacturing is
493 from coke production (Pacyna et al., 2006).

494 The coastal areas east of the sampling site where there are large ports were also identified
495 as the likely source areas of TGM. A previous study reported that the emissions of gaseous
496 and particulate pollutants were high during vehicular operations in port areas and from
497 marine vessel and launches (Gupta et al., 2002). Another possibility is that significant amount
498 of GEM are emitted from the ocean surface because of photo-chemically and
499 microbiologically mediated photo-reduction of dissolved GOM (Amyot et al., 1994; Zhang
500 and Lindberg, 2001). The northeast direction including the East Sea was also identified as
501 potential source areas likely because this is an area with lots of domestic passenger ships
502 routes. The south from the sampling site was also identified as a likely source area of TGM
503 where Ulsan Metropolitan City, South Korea's seventh largest metropolis with a population
504 of over 1.1 million is located. It includes a large petrochemical complex known as a TGM
505 source (Jen et al., 2013).

506

507 **Conclusions**

508 During the sampling periods, the average TGM concentration was higher than the Northern
509 Hemisphere background concentration, however, considerably lower than those near urban
510 areas in China and higher than those in Japan and other locations in Korea. The median
511 concentration of TGM was much lower than that of the average, suggesting that there were
512 some extreme pollution episodes with very high TGM concentrations. The TGM
513 concentration was highest in fall, followed by spring, winter and summer. The high TGM
514 concentration in fall is due to transport from different wind directions than during the other
515 periods. The low TGM concentration in summer is likely due to increased mixing height and
516 gas phase oxidation at higher temperatures particularly at this sampling site which is close to
517 the ocean (2 km) where oxidation involving halogens may be enhanced.

518 TGM consistently showed a diurnal variation with a maximum in the early morning
519 (06:00-09:00) and minimum in the afternoon (14:00-17:00). Although there was a statistically
520 significant negative correlation between the TGM concentration and ambient air temperature,
521 the daytime TGM concentration was higher than those in the nighttime, suggesting that local
522 emission sources are important. There was a negative relationship between the TGM
523 concentrations and O₃ concentrations, indicating that the oxidation was partially responsible
524 for the diurnal variations of TGM concentrations. The observed $\Delta\text{TGM}/\Delta\text{CO}$ was
525 significantly lower than that indicative of Asian long-range transport, suggesting that local
526 sources are more important than that of long-range transport. CPF only shows high
527 probabilities to the west from the sampling site where there are large steel manufacturing
528 facilities and waste incinerators. However, CBPF and TPSCF indicated that the dominant
529 sources of TGM were the hazardous waste incinerators and the coastal areas in the northeast

530 as well as the iron and manufacturing facilities in the west. The domestic passenger ships
531 routes in the East Sea were also identified as possible source areas.

532

533 **Author contribution**

534 Yong-Seok Seo conducted a design of the study, the experiments and analysis of data, wrote
535 the initial manuscript, and finally approved the final manuscript. Seung-Pyo Jeong, Eun Ha
536 Park, Tae Young Kim, Hee-Sang Eum, Dae Gun Park, Eunhye Kim, Jaewon Choi and Jeong-
537 Hun Kim conducted the experiments, analysis of data, and finally approved the final
538 manuscript. Thomas M. Holsen, Young-Ji Han and Eunhwa Choi and Soontae Kim
539 conducted interpretation of the results, revision of the initial manuscript, and finally approved
540 the final manuscript. Seung-Muk Yi conducted a design of the study, acquisition of data of the
541 study, interpretation of data, and revision of the initial manuscript, and finally approved the final
542 manuscript.

543

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548 Korea and Korea Ministry of Environment (MOE) as “the Environmental Health Action
549 Program”.

550

551 **Table List**

552 Table 1. Comparison with previous studies for TGM concentrations.

553 Table 2. Summary of atmospheric concentrations of TGM and co-pollutants, and
554 meteorological data.

555

556 **Figure List**557 Fig. 1. The location of sampling site in this study ((a) South Korea, (b) Gyeongsangbuk-do
558 and (c) Pohang).

559 Fig. 2. Time-series of TGM concentrations in this study.

560 Fig. 3. The diurnal variations of TGM concentrations during the sampling periods.

561 Fig. 4. CPF, CBPF and TPSCF plots for TGM higher than average concentration.

562 **Table 1.** Comparison with previous studies for TGM concentrations.

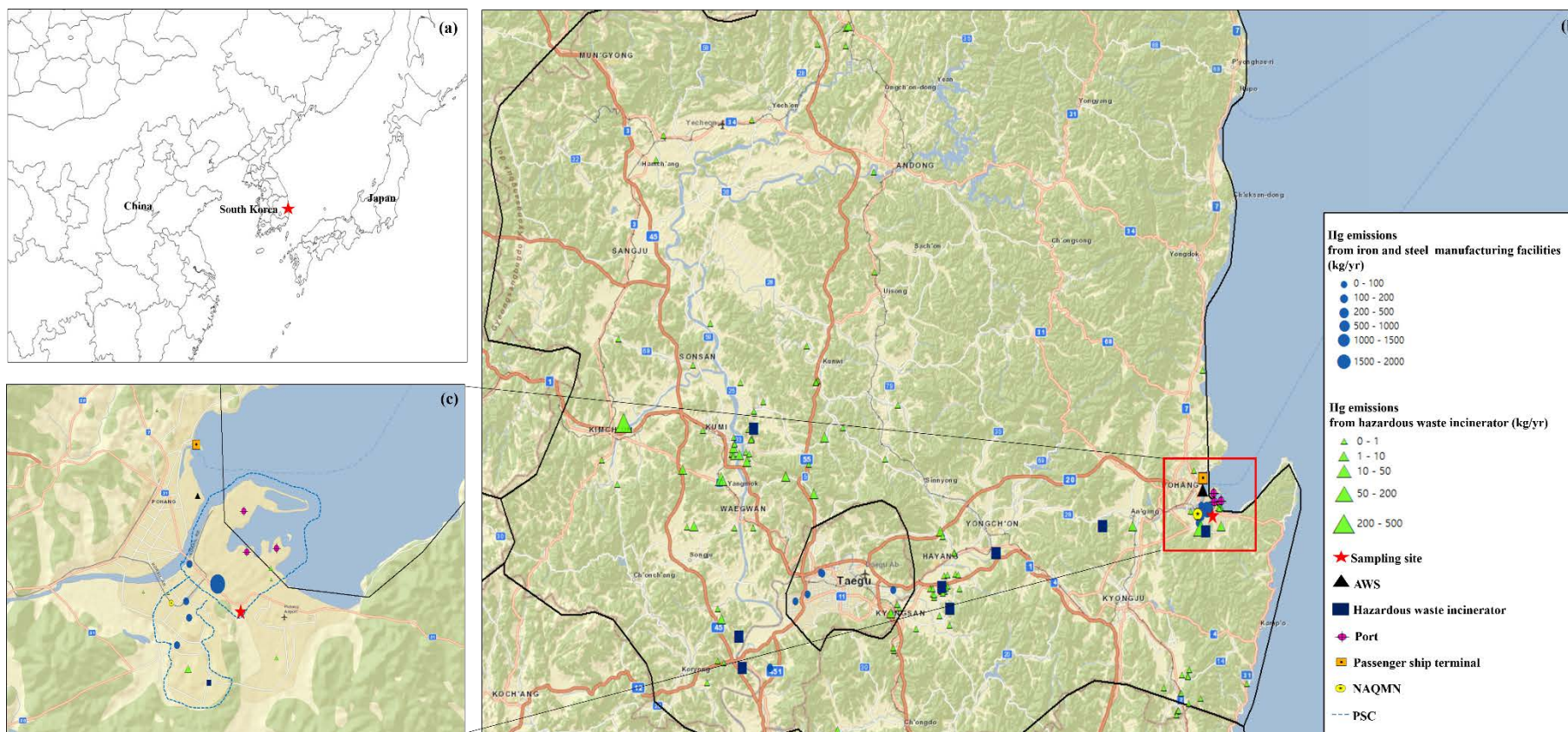
Country	Location	Sampling period	TGM conc. (ng m ⁻³)	Classifications	Reference
China	Mt. Hengduan, Qinghai–Tibet Plateau	Jul. 2010 ~ Oct. 2010	2.5	Remote	Fu et al. (2015)
China	Nanjing, Jiangsu	Jan. 2011 ~ Oct. 2011	7.9	Urban	Hall et al. (2014)
China	Mt. Dinghu, Guangdong	Oct. 2009 ~ Apr. 2010	5.1	Rural	Chen et al. (2013)
China	Guangzhou, Guangdong	Nov. 2010 ~ Nov. 2011	4.6	Urban	Chen et al. (2013)
China	Gui Yang, Guizhou	Jan. 2010 ~ Feb. 2010	8.4	Urban	Feng et al. (2004)
China	Changchun, Jilin	Jul. 1999 ~ Jul. 2000	13.5-25.4	Urban	Fang et al. (2004)
Japan	Fukuoka	Jun. 2012 ~ May 2013	2.33	Urban	Marumoto et al. (2015)
Japan	Tokai-mura	Oct. 2005 ~ Aug. 2006	3.8	Suburban	Osawa et al. (2007)
Japan	Tokyo	Apr. 2000 ~ Mar. 2001	2.7	Urban	Sakata and Marumoto (2002)
Korea	Seoul	1987 ~ 2013	3.7	Urban	Kim et al. (2016)
Korea	Gangwon-do, Chuncheon	2006 ~ 2009	2.1	Rural	Han et al. (2014)
Korea	Seoul	Feb. 2005 ~ Feb. 2006	3.2	Urban	Kim et al. (2009)
Korea	Seoul	Feb. 2005 ~ Dec. 2006	3.4	Urban	Choi et al. (2009)
Korea	Seoul	19 Sep. 1997 ~ 29 Sep. 1997 27 May. 1998 ~ 18 Jun. 1998	3.6	Urban	Kim and Kim (2001)
Korea	Gyeongsangbuk-do, Pohang	17 Aug. 2012 ~ 23 Aug. 2012 9 Oct. 2012 ~ 17 Oct. 2012 22 Jan. 2013 ~ 29 Jan. 2013 26 Mar. 2013 ~ 3 Apr. 2013	5.0	Urban	This study

563

564
565**Table 2.** Summary of atmospheric concentrations of TGM and co-pollutants, and meteorological data. Note that TGM was measured every 5-min, and other pollutants and meteorological data were measured every 1-hour.

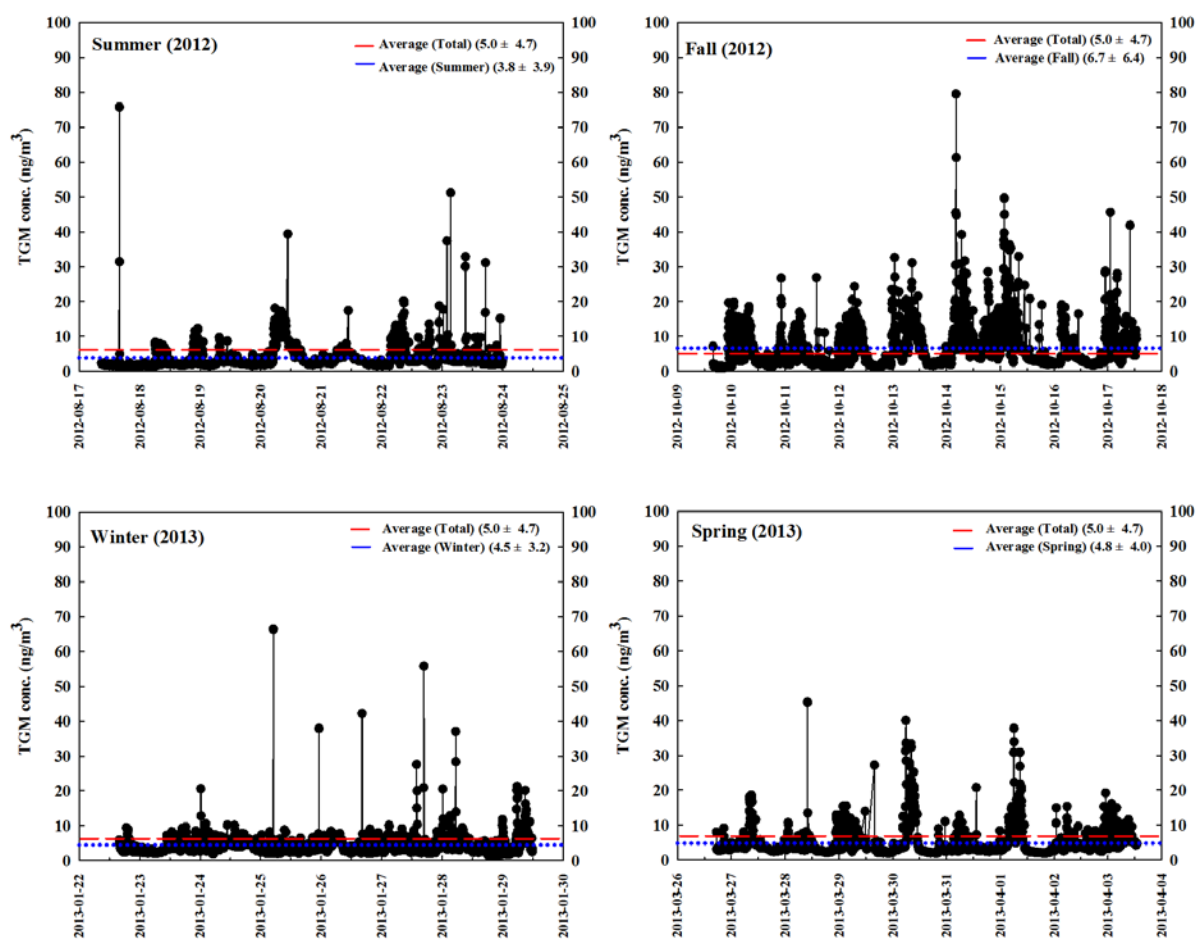
		TGM (ng m ⁻³)	NO ₂ (ppb)	O ₃ (ppb)	CO (ppb)	PM ₁₀ (µg m ⁻³)	SO ₂ (ppb)	Temperature (°C)	Wind speed (m s ⁻¹)	Humidity (%)	Solar radiation (MJ m ⁻²)
Spring	N	2139	189	215	215	215	215	216	216	216	216
	Average	4.8 ± 4.0	25.3 ± 9.0	29.4 ± 14.2	766.5 ± 505.2	70.1 ± 26.0	7.6 ± 3.8	10.5 ± 4.2	2.2 ± 1.2	56.2 ± 16.8	0.82 ± 1.09
	Range	1.9 – 45.3	8 – 55	2 – 58	300 – 3100	28 - 204	5 - 35	1.1 – 21.6	0.4 – 6.2	19.0 – 94.0	0 – 3.44
Summer	N	1863	187	188	187	188	188	186	180	186	141
	Average	3.8 ± 3.9	18.3 ± 9.2	18.9 ± 10.1	697.3 ± 689.7	35.1 ± 15.8	6.5 ± 6.2	26.6 ± 4.2	2.2 ± 1.1	82.5 ± 13.9	0.40 ± 0.69
	Range	1.2 – 75.9	4 – 44	5 – 48	200 – 3300	12 – 87	2 - 27	19.7 – 34.1	0.1 – 6.4	43 - 98	0 – 2.92
Fall	N	2226	212	212	212	212	211	216	216	216	216
	Average	6.7 ± 6.4	25.0 ± 7.8	23.7 ± 13.1	662.7 ± 350.2	58.1 ± 17.8	5.3 ± 3.5	17.4 ± 3.2	2.1 ± 0.8	54.5 ± 14.7	0.62 ± 0.90
	Range	1.0 – 79.6	9 – 53	6 – 69	300 – 2900	20 - 145	3 - 39	11.7 – 25.2	0.5 – 4.5	12 - 79	0 – 2.90
Winter	N	1917	188	187	188	188	186	192	192	192	192
	Average	4.5 ± 3.2	23.5 ± 14.7	26.1 ± 8.7	556.4 ± 298.9	56.3 ± 30.5	7.4 ± 2.5	1.1 ± 4.3	2.8 ± 1.1	46.3 ± 24.5	0.43 ± 0.71
	Range	1.3 – 66.4	5 – 74	1 – 41	200 – 2400	18 – 161	5 – 24	-0.65 – 10.1	0.5 – 6.0	11 - 90	0 – 2.34
Total	N	8145	776	802	802	803	800	810	804	810	765
	Average	5.0 ± 4.7	23.1 ± 10.8	24.6 ± 12.5	673.7 ± 487.3	55.5 ± 26.4	6.7 ± 4.3	13.8 ± 9.9	2.3 ± 1.1	59.4 ± 22.1	0.59 ± 0.90
	Range	1.0 – 79.6	4 – 74	1 – 69	200 – 3300	12 – 204	2 – 39	-6.5 – 34.1	0.1 – 6.4	11 - 98	0 – 3.44

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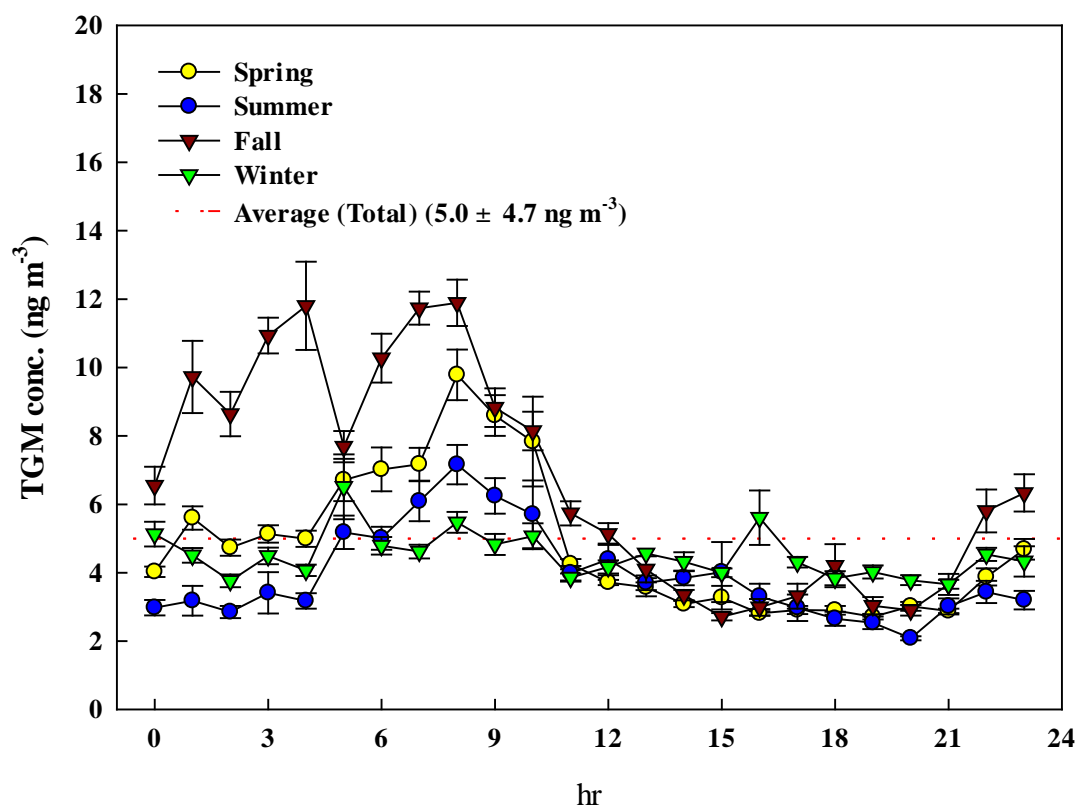
Fig. 1. The location of sampling site in this study ((a) South Korea, (b) Gyeongsangbuk-do and (c) Pohang). AWS, NAQMN and PSC represent Automatic Weather Station, National Air Quality Monitoring Network and Pohang Steel Complex, respectively.



568

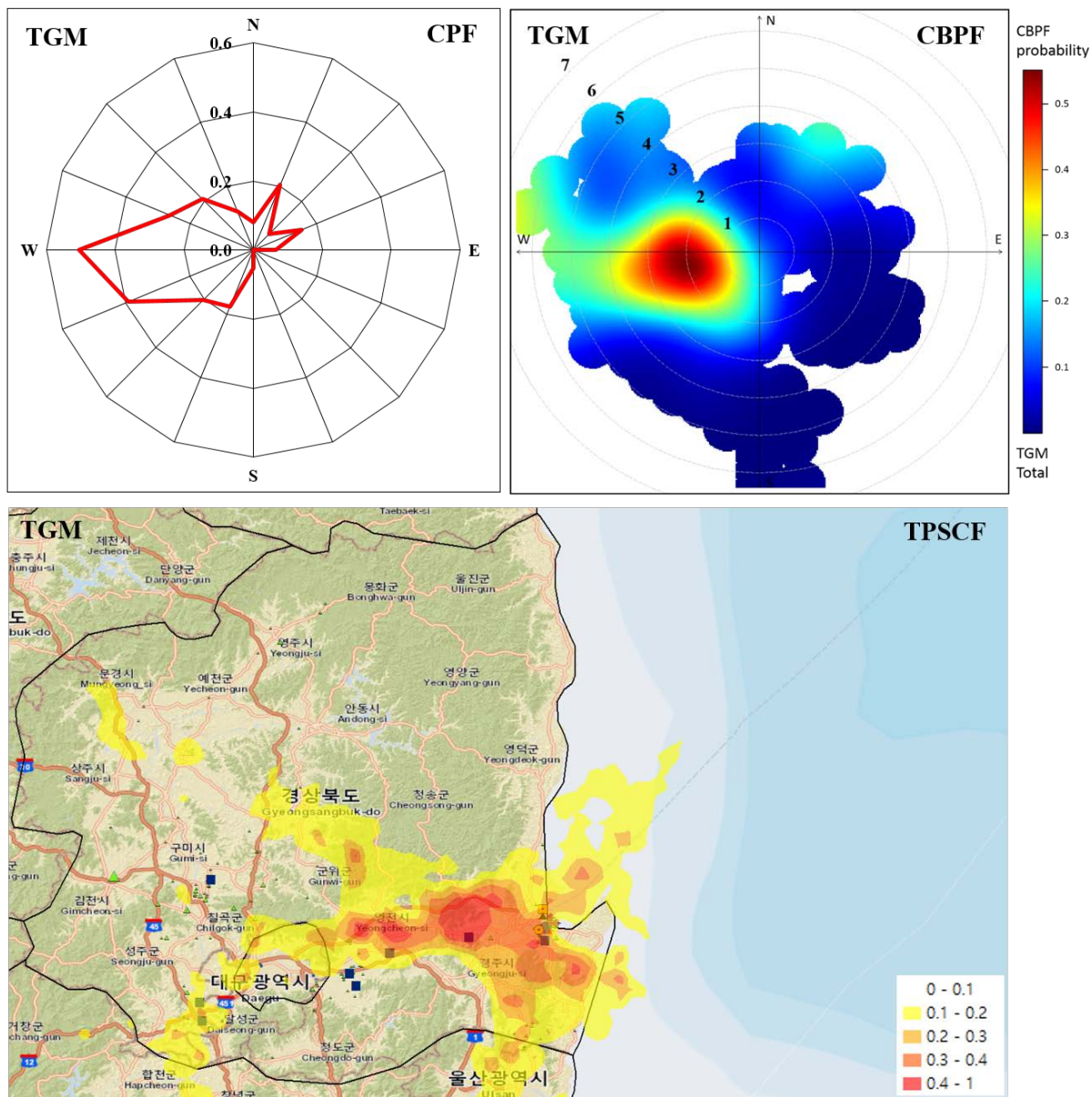
569

Fig. 2. Time-series of TGM concentrations in this study.



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Fig. 3. The diurnal variations of TGM concentrations during the sampling periods. The error bars represent standard error.



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Fig. 4. CPF, CBPF and TPSCF plots for TGM higher than average concentration. The radial axes of CPF and CBPF are the probability and the wind speed (m s^{-1}), respectively.

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