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*Supplement of*

## **Successes and challenges of measuring and modeling atmospheric mercury at the part per quadrillion level: a critical review**

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13 ***Lumex***

14

15 After startup, the Lumex instrument baseline will drift downward asymptotically for 1 to 3  
16 hours before leveling off at a stable value, after which a meaningful zero baseline value can be  
17 set. Laboratory tests show after stabilization, a mean blank concentration of  $-0.13 \pm 0.34 \text{ ng m}^{-3}$   
18 <sup>3</sup> can be maintained for at least 3 hours while operating in the internal zero reference mode,  
19 with no indication of baseline drift (linear regression coefficient of 0.00004). External source  
20 injections of GEM in zero air at calculated concentrations of 240, 120, and 60  $\text{ng m}^{-3}$  resulted in  
21 mean recoveries of 99.7, 97.9, and 95.9 percent, respectively. If the recovery efficiency trend is  
22 extrapolated to 1  $\text{ng m}^{-3}$  the Lumex would under report by about 15%.

23

24 ***Sheu and Mason (2001) methods and detailed results***

25

26 The filter pack consisted of two Teflon filters in front of three cation exchange membranes in-  
27 series. They assumed the first Teflon filter collected PBM and the second was a blank, the first  
28 cation exchange membrane collected GOM, the second was a blank, and the third kept GOM  
29 from diffusing from the pump. They slightly altered the mist chamber from the design of  
30 Lindberg and Stratton (1998).

31

32 The mist chamber collected ~34% more GOM than the cation exchange membrane. Sheu and  
33 Mason suggested this may be an artifact of the mist chamber in marine air. The denuders in  
34 general indicated higher concentrations than the filter pack measurements. They suggested this  
35 was due to an inherent blank problem or due to GEM being oxidized at the KCl surface. They  
36 tested whether there were reactions in the mist chamber when exposed to light and found this  
37 did not occur. PBM was also measured on Teflon filters and concentrations ranged from  $<9$  to  
38  $147 \text{ pg m}^{-3}$ .

39

40 ***Cation exchange membrane***

41

42 The original material used for the cation exchange membrane “ICE 450” has been discontinued  
43 and replaced with a polyethersulfone membrane (“Mustang S”). In laboratory and field tests  
44 collection of GOM in ambient air were not significantly different; however, in the field the ICE  
45 450 in general had higher collection efficiency, although they were not significantly different,  
46 suggesting it may collect some forms of GOM that the Mustang S does not (Huang and Gustin,  
47 submitted). Recent field tests indicated there was no significant loss when surrogate surfaces  
48 and UNR passive sampler for GOM concentrations (box sampler) were deployed for 2 to 4  
49 weeks (Huang and Gustin, submitted). In addition, use of an automated analytical system  
50 (Tekran 2600) for membrane analyses significantly improved the method detection limit (Huang  
51 and Gustin, submitted).

52

### 53 ***TGM vs GEM comparison***

54

55 Limited field work has indicated that the Tekran® 2537 alone does collect all TGM with a soda  
56 lime and particulate filter in-line. Temme et al. (2002) compared data from two Tekran  
57 systems in Antarctica, one measuring TGM using Tekran® 2537 only and the other using the  
58 Tekran® 1130/1135/2537 to measure GEM, GOM and PBM. They suggested that GOM and GEM  
59 were collected as TGM by the 2537. Based on 7 months of data from Mercury Deposition  
60 Network site MDN 98 in Nevada, Gustin (2011) found the sum of the GEM and GOM was more  
61 similar to TGM than GEM alone. This comparison was in an environment with relatively dry air  
62 and similar comparisons in humid air may yield more comparable concentrations since humidity  
63 can reduce GOM collected on surfaces to GEM.

64

### 65 ***Detailed discussion on the potential for different forms in the air in Florida***

66

67 There are several important points not presented in these papers. First in Peterson  
68 et al. (2012), the discrepancy between modeled dry deposition using a multi-resistance  
69 model, and measured deposition varied by location and across different seasons. This  
70 supports the contention that there are different forms in air. At these sites modeled  
71 deposition was weakly correlated with measured deposition ( $r^2 = 0.24$  and  $0.28$   $p < 0.5$ , and  
72  $r^2 = 0.16$   $p < 0.10$ ). This indicates at times there were GOM compounds in the air that the  
73 Tekran® system was more efficiently capturing, and at other times this measurement was  
74 less efficient, or the model was not accurately capturing deposition.

75

76 The constant model parameterization and similar environmental conditions across  
77 all sites (cf. Peterson et al., 2012) provides a basis for comparison of measured deposition  
78 across all sites. In all cases measured deposition was significantly higher (2 to 18 times)  
79 than modeled values (Figure SI 1). At all sites there was a pattern in the ratio of  
80 measured:modeled deposition with a decline in the summer and increase in fall.  
81 Differences between the measured and modeled values were similar for all sites in the  
82 winter. The Davie Site near Fort Lauderdale (DAV) site had the lowest ratio of measured:  
83 modeled deposition during the other seasons, suggesting the denuder was more efficient at  
84 collecting GOM compounds at this location, or that the model was better at capturing  
85 deposition at this site. Ratios were highest at Tampa (TPA) in the spring and fall relative to  
86 the other sites. At Outlying Landing Field (OLF), the most background site, measured:  
modeled values were higher in the spring (time periods when long range transport occurs)

87 and in the fall, a time of year when mobile source pollution would be higher. An alternate  
88 explanation is that the model bias varies seasonally because of errors in meteorology,  
89 surface/land type, PBL height, etc. and this could explain some of the variability in the  
90 measured:modeled ratio. For a detailed discussion of how these data demonstrate there  
91 are different forms of GOM in the air see the SI.

92 Data of Peterson et al (2012), presented in Figure SI 2, shows that the surrogate  
93 surface-derived deposition velocities are highest for the TPA site that is primarily impacted  
94 by mobile sources, and overlap for DVE (local point source impact and mobile source  
95 impacted), and were intermediate for OLF (most background site with a long range  
96 transport impact). Despite the fact these were calculated using the Tekran® GOM data,  
97 since there were similar conditions across the sites, this also indicates there are different  
98 forms in the air at these sites. Dry deposition measured by the surrogate surface will be  
99 influenced by the deposition velocity or the chemical compound. Lin et al. (2006), using the  
100 model of Wesely (1989), reported the deposition velocity of HgO was 2-fold higher than  
101 that for HgCl<sub>2</sub>. Thus, if there are different forms in the air, data collected by the surrogate  
102 surface will vary across space and time.

103  
104 If we assume:

105 1- the cation exchange collects GOM quantitatively then the collection efficiency of KCl-  
106 coated denuder (in a Tekran 1130) for different GOM compounds is HgBr<sub>2</sub> (1.6)>HgSO<sub>4</sub>  
107 (2.3)=HgCl<sub>2</sub> (2.4)>HgO (3.7) >Hg(NO<sub>3</sub>)<sub>2</sub> (12.6) (Values in parenthesis represent concentrations  
108 measured by the cation exchange membrane: concentrations reported by the Tekran® denuder  
109 using the laboratory manifold system in zero air); and

110 2- based on the work presented in Gustin et al. (2012) that deposition associated with  
111 natural background is 0.23 ng m<sup>-2</sup> h<sup>-1</sup> (corrected using the 0.2 ng m<sup>-2</sup> h<sup>-1</sup> factor); deposition  
112 associated with oxidation of GEM due to mobile source emissions at TPA, DAV, and OLF was  
113 0.3, 0.3, and 2.3 ng m<sup>-2</sup> h<sup>-1</sup>, respectively; deposition from long range transport was 2.8 ng m<sup>-2</sup> h<sup>-1</sup>  
114 at all sites in the spring; and deposition at DVE associated with direct emission or formation  
115 due to associated oxidants was 0.3 ng m<sup>-2</sup> h<sup>-1</sup>, there could be different sources associated with  
116 each specific source. For example, natural background could be due to reactions with Br and  
117 perhaps S compounds; deposition associated with mobile sources could be Hg-O or Hg-N  
118 associated compounds; that due to long range transport in the spring could be HgO related or  
119 HgBr<sub>2</sub> compounds; and that associated with local point sources could be, Hg-S, Hg-O or Hg-N  
120 related compounds (cf. Timonen et al., 2013). Since collection efficiency for HgSO<sub>4</sub> is relatively  
121 good, this might explain the better agreement between the measured and modeled deposition  
122 at DVE and this site is impacted by oil electrical generation facilities. These ratios could also be  
123 influenced by ozone and humidity (see Lyman et al., 2010; McClure et al. 2014; Huang and  
124 Gustin, this issue, and discussion below). The atmospheric constituents at all sites are more  
125 complicated than that in the laboratory manifold (discussed below). Recent work by Huang et  
126 al. (this issue) based on thermodesorption of the nylon membranes, at OLF there are different  
127 compounds depending on the time of the year.

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130

131 **Detailed discussion of RAMIX data**

132  
133 Ambient air RM concentrations measured during RAMIX by the DOHGS were  
134 typically higher than the Tekran® derived RM. Although data are noisy, RM was typically  
135 higher as measured by the DOHGS at midnight to late morning (up to 350 pg m<sup>-3</sup>), relative  
136 to the Tekran® system. At 12:00, values measured by the DOHGS compared with the  
137 Tekran® systems were higher by ~75 pg m<sup>-3</sup>. The variation in the discrepancy between  
138 DOHGS and Tekran® RM measured over 24 h points to different GOM compounds in the air  
139 and production of GOM in the manifold.

140 Another piece of evidence for different GOM compounds in the air and points to fast  
141 reactions that were hypothesized to occur in the manifold, is presented in Figure SI 3 from  
142 Gustin et al. (2013). The timing of the hourly peaks in GOM as measured are similar to  
143 those observed for ozone (one at 1200 h and another later in the afternoon). The first is  
144 attributed to mixing of air pollution to the surface from the free troposphere. The second is  
145 due to a reaction with local pollution (see Gustin et al., 2013). This latter could be due to  
146 the reaction suggested by Calvert and Lindberg (2005) for Hg and O<sub>3</sub>:

147  $GEM + O_3 \rightarrow HgO_3 \text{ (unstable)} \rightarrow OHgOO \rightarrow O_2 \text{ and } HgO$ . Equation 2.

148 They also indicated that HgO<sub>3</sub> would deposit on aerosols and other surfaces where it may  
149 form HOHgOH, and this state Hg could be transformed into a variety of Hg (II) compounds  
150 with other ions. For additional discussion of atmospheric chemistry see the discussion  
151 below.

152 Thus, the RAMIX experiment demonstrated that the Tekran® KCl-coated denuder  
153 measurement for GOM was biased low, there were different chemical forms in the air, and  
154 reactions were occurring in the manifold (Gustin et al., 2013). The latter has implications  
155 for reactions that could occur in ambient air. Although there were some limitations with  
156 each method applied during the experiment, the project also demonstrated: 1) using GEM  
157 spikes, measurements made by two carefully calibrated Tekran® 2537 units could  
158 significantly differ and the only way to explain this was a discrepancy between the  
159 instruments; 2) GOM was significantly underestimated by the denuder (17 to 24%  
160 recovery of HgBr<sub>2</sub> spikes in ambient air in Reno); and 3) the DOHGS measured different RM  
161 forms than the Tekran® KCl-coated denuder.

162  
163  
164 **Detailed discussion of atmospheric chemistry**

165  
166 Hynes et al (2009) summarized the laboratory work that has been done on the Hg + O<sub>3</sub>  
167 reaction by Hall (1995), Pal and Aryia (2004a and b), Summer et al.(2005), Rutter et al. (2012).  
168 All of these studies found O<sub>3</sub> to participate in a reaction with GEM. Based on an analyses of the  
169 electronic configuration, Tossell (2003), and Shepler and Peterson (2003) indicated that HgO  
170 is a weakly bound molecule. Hynes et al (2009) calculated a rate coefficient for the following  
171 equation:

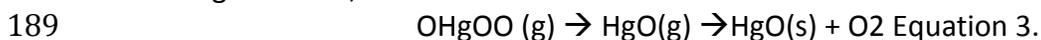


172  
173 They indicated the rate coefficient was unreasonably high and suggested this reaction  
174 was unlikely to occur by homogenous process. Hynes et al (2009) noted that if the Hg + O<sub>3</sub>  
175 reaction does exist it must occur as a weakly bound complex; and "In laboratory experiments

176 such a complex could diffuse to the reactor surface to form solid mercuric monoxide, possibly  
177 by way of oligomer formation. Such a process would be energetically favorable because of the  
178 exothermicity of oligomer formation and the lattice energy associated with formation of the  
179 solid.”

180 Peterson et al (2009) indicated that the rate coefficient for O<sub>3</sub> production developed by  
181 Pal and Ayria (2004a) was sufficient to produce 5 to 10 pg of HgO per hour; and using the rate  
182 coefficient developed by Pal and Ayria (2004b) and a concentration of OH of 1 x 10<sup>-6</sup> molecules  
183 per cm<sup>3</sup> (Seinfeld and Pandis, 1996) approximately 1 to 10 pg of HgO could be produced per  
184 hour.

185 Ariya et al. (2009) suggested that most atmospheric oxidation reactions occur by  
186 multiple steps complicating our ability to experimentally study and calculate reaction rates, and  
187 understand reactions. They proposed after the initial reaction between GEM and O<sub>3</sub> that the  
188 following occurred;



190 The last two steps are irreversible. Additional work by the Ariya group showed that the O<sub>3</sub>  
191 oxidation rate increases in the presence of CO and relative humidity (Snider et al., 2008).

192

### 193 ***PBM detailed information***

194

195 Keeler et al (1995) used Teflon and quartz fiber filters to measure PBM in the Great  
196 Lakes area and showed PBM had a bimodal distribution. A paper by Kim et al. (2012) showed a  
197 bimodal size distribution with the strength of this distribution varying by season and location,  
198 and PBM was measured for 0.18 to 18 μm using an eight-stage MOUDI (Micro Orifice Uniform  
199 Deposit Impactors, Midel M 100R, MSP Corp) with Zeflour filters (Pall corp.) and a cascade  
200 impactor with glass fiber filters. Particles across all size ranges were measured.

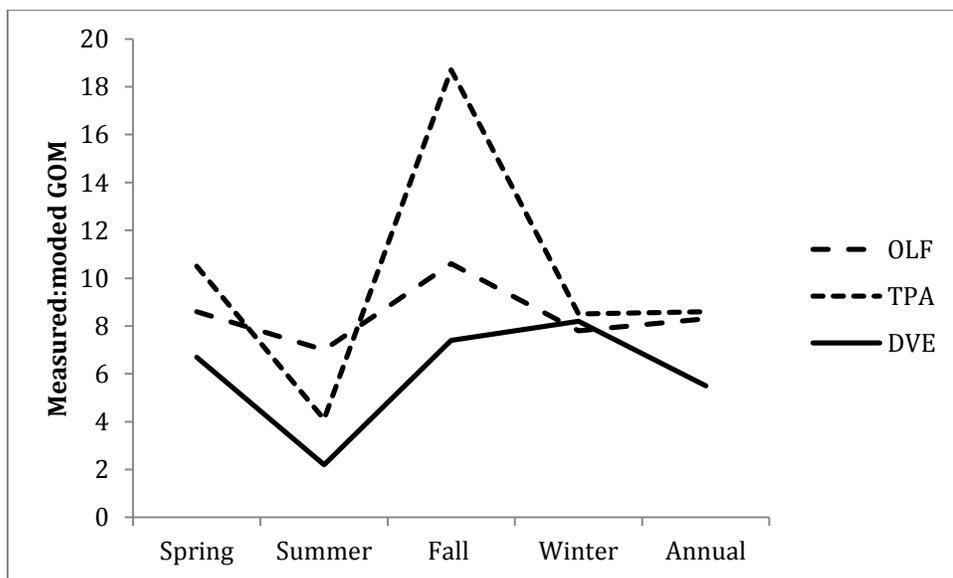
201 Keeler et al. (1995) reported 60-100% of PBM was found in fine particle fraction in  
202 Detroit. A significant fraction of PBM can be found in coarse particle at coastal sites (Engle et al.,  
203 2008; Malcolm et al., 2007). Kim et al. (2012) found PBM in the winter in Chuncheon and Seoul  
204 Korea were in the < 2.5 μm fraction, however, in the summer, in Seoul, there was a constant  
205 distribution across all particulate sizes, and in Chuncheon, concentrations in the summer were  
206 higher in the 2 to 10 μm fraction. This indicates the grain size fraction measured by the  
207 Tekran® is not adequate due to the need for an elutricator to keep particles of > 2.5 μm from  
208 entering the denuder.

209 Rutter et al. (2008) used quartz fiber filters and KCl-coated quartz fiber filters in-series  
210 to collect PBM and GOM and found that concentrations on the first filter were higher, and the  
211 latter comparable to that measured with a Tekran® system. It is possible some GOM was  
212 collected on the quartz fiber filter. PBM measured by the quartz filter was 2.7 times higher  
213 than that measured by the Tekran® 1135. Since RM is semi-volatile (Rutter and Schauer, 2007),  
214 Rutter et al. (2008) suggested that since the particulate module is maintained at 50oC, RM  
215 could be lost. Rutter et al. (2008) suggested that evaporative loss could be augmented by the  
216 presence of other semivolatile compounds such as NH<sub>3</sub>NO<sub>3</sub> or other semivolatile organics.  
217 Malcolm and Keeler (2007) observed less PBM collected on quartz filters for 12 hrs versus 4 hrs  
218 and suggested a negative sampling artifact that could be due to reduction due to reaction with  
219 gases in the air (such as O<sub>3</sub>) or relative humidity.

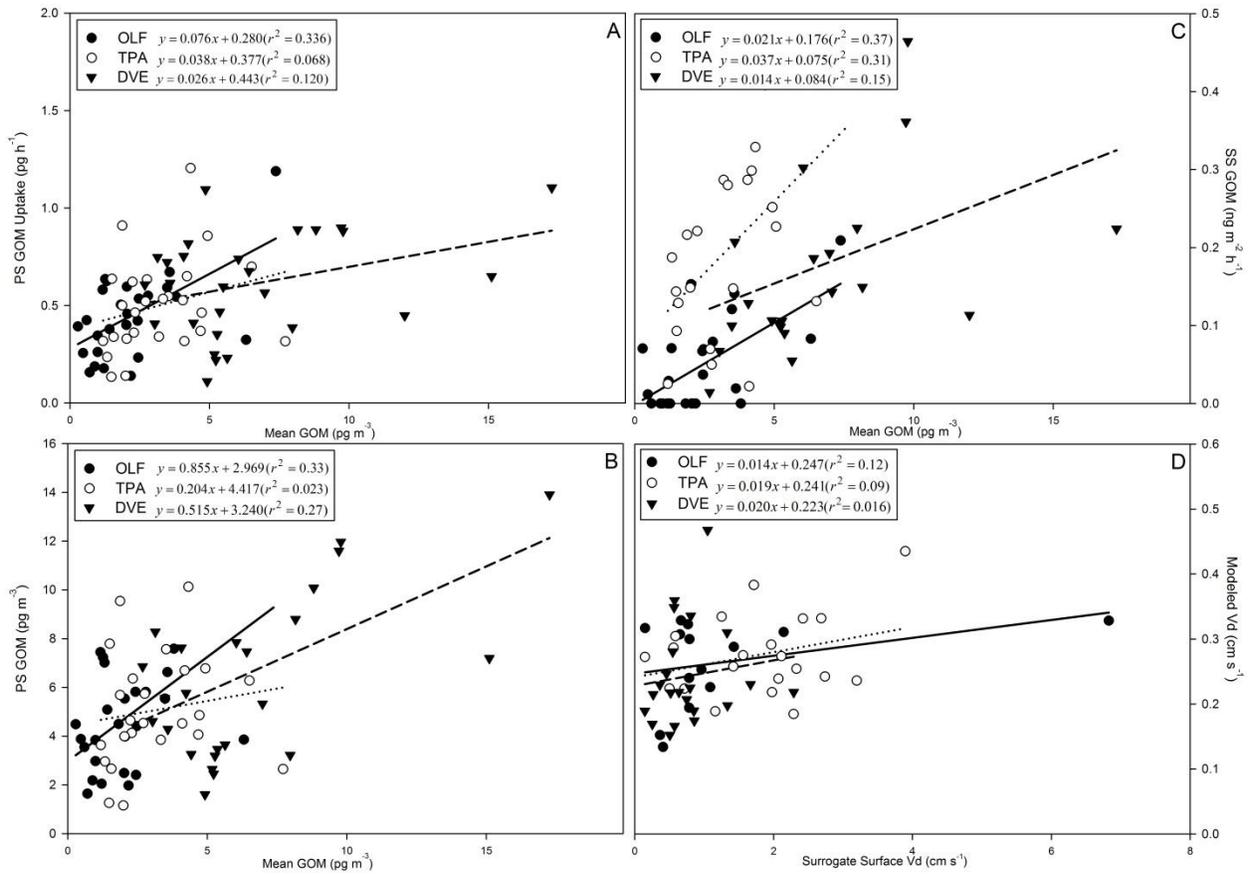
220 Talbot et al. (2011) measured PBM with a Millipore® 1 µm fluoropore membrane to  
 221 compare with data collected using the Tekran® 1135 unit and found the filter method collected  
 222 21% higher PBM and 85% of the data disagreed by 25% or more. In addition, the Tekran®  
 223 measurement had a high percentage of data below the detection limit while the particulate  
 224 filter (average blank concentration 25 pg and blank for the Tekran® measurement was 0 pg m<sup>-3</sup>)  
 225 always had detectable Hg. The filter method was more responsive to pollution episodes  
 226 associated with a combustion source. It is also possible their filters were collecting some GOM.  
 227

228 ***Details on the sampling configuration in Weiss-Penzias et al. 2003***

229  
 230 For this study they describe GEM only concentrations being measured from May 2001  
 231 to May 2002 by a 2537; GOM measurements began in September 2001 by the 1130; and PBM  
 232 was only measured at selected times using filters. For the GEM measurement the instrument  
 233 was housed in a trailer with a black plastic covered line heated to 40°C provided by Tekran and  
 234 a quartz filter to remove PBM and GOM. The quartz filter likely worked for PBM and possibly  
 235 some forms of GOM. Once installed the 1130 was configured to collect a sample over 4 hours.  
 236 Quartz filters were used to collect PBM over 27 hours on 47 mm quartz fiber filters. The  
 237 samplers collected particulate matter < 2.5 µm, and 2.5 to 10 µm.  
 238

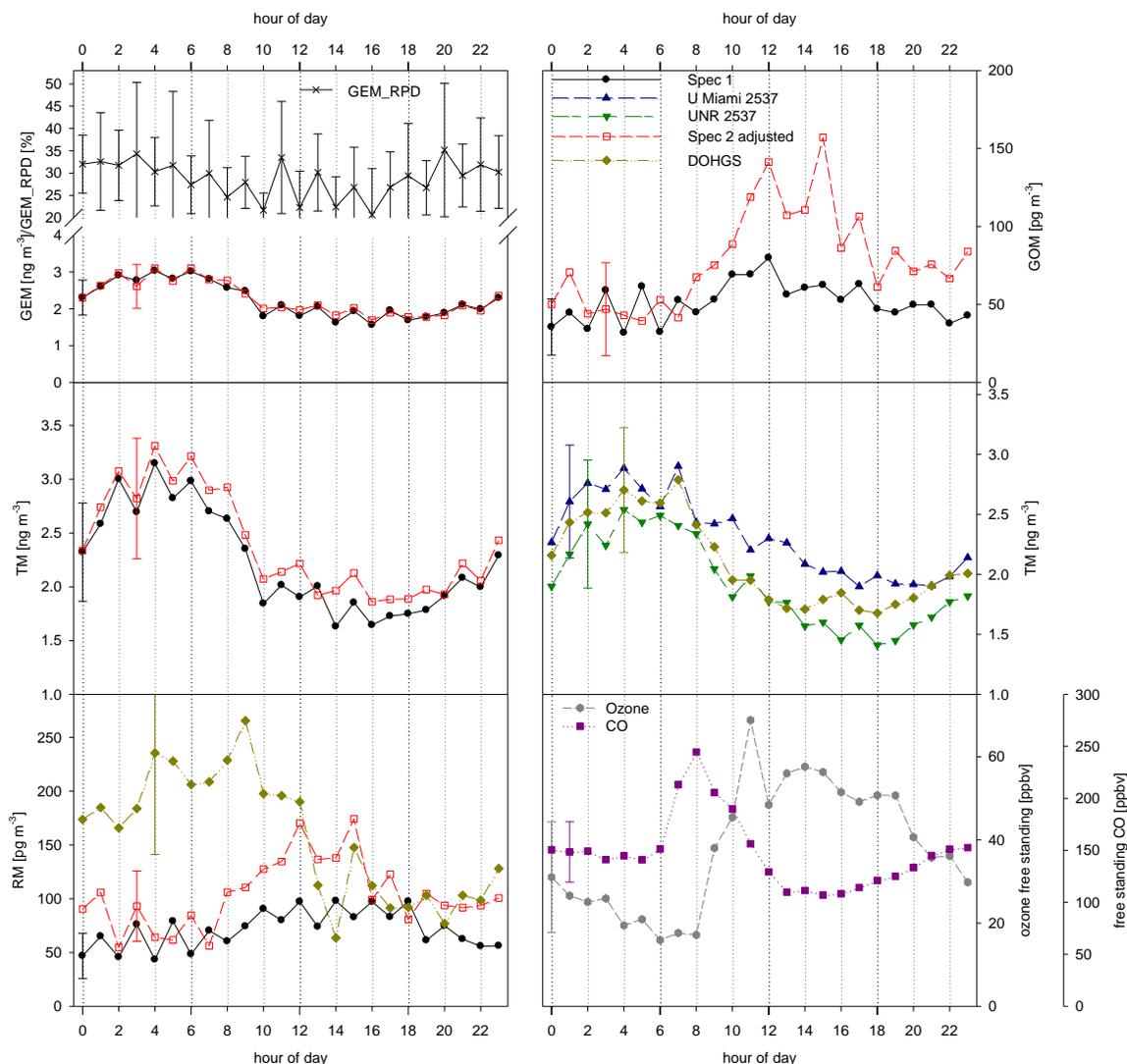


239  
 240  
 241 Figure SI 1. Ratio of surrogate surface measured (uncorrected dry deposition value-see section  
 242 3.2.2) to modeled values of GOM dry deposition for Spring (March-May ), Summer (June-August),  
 243 Fall(September-November ), winter (December-January), and for the year. Data were collected  
 244 from July 2009 to July 2010 at Outlying Landing Field (OLF) near Pensacola, a site just east of  
 245 Tampa (TPA), and one near Fort Lauderdale (DVE) and the town of Davie (Data from Peterson et al.,  
 246 2012).  
 247



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 249  
 250  
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Figure SI 2. Figure 4 from Peterson et al (2012). Reprinted with permission from Peterson et al., 2012, Copyright 1 May 2012, Elsevier Limited.



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254 Figure SI 3. Figure 2 from Gustin et al (2013). Reprinted with permission from Gustin et al., 2013,  
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