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**Total gaseous mercury concentrations at the Cape Point  
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1 **Total gaseous mercury concentrations at the Cape Point GAW station and**  
2 **their seasonality**

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13  
14 **Abstract**

15  
16 Total gaseous mercury (TGM) has been measured at the WMO Global Atmosphere Watch  
17 (GAW) station at Cape Point, South Africa, since September 1995. Annual medians suggest a  
18 small but significant decrease of TGM concentrations from 1.29 ng m<sup>-3</sup> in 1996 to 1.19 ng m<sup>-3</sup>  
19 in 2004. Background TGM concentrations at Cape Point show a pronounced seasonal  
20 variation with a maximum in January and February and a minimum in Juli, August and  
21 September. This seasonal variation is opposite in phase to that of CO at Cape Point, whereas  
22 at Mace Head in the northern hemisphere the seasonal variations of TGM and CO are in  
23 phase. This behavior is in approximate agreement with the seasonal variation of major  
24 mercury sources in both hemispheres, suggesting lesser influence of the seasonality of sinks.  
25 The different seasonal variations of TGM concentrations in the northern and southern  
26 hemispheres may pose an important constraint on the global models of atmospheric mercury.

27

28 **Introduction**

29

30 Mercury, in contrast to other metals, is volatile and its vapor can be transported in air over  
31 large distances because of its low reactivity and solubility. After oxidation to less volatile and  
32 more soluble compounds it is deposited (e.g. Slemr et al, 1985; Lin et al., 2006). Part of the  
33 deposited mercury compounds is then converted to more toxic methyl mercury which  
34 bioaccumulates in the aquatic food chain. The high concentration of methyl mercury in the  
35 predatory fish pose a serious risk for people and animals which depend on a fish diet (e.g.  
36 Mergler et al., 2007; Scheuhammer et al., 2007). Consequently, the research on atmospheric  
37 mercury has intensified during the last decade (Lindberg et al., 2007).

38

39 Despite of the substantial progress, the understanding of the global atmospheric cycle is still  
40 incomplete (e.g. Lin et al., 2006). Natural and anthropogenic mercury emissions are not well  
41 known (e.g. Lindberg et al., 2007). The major knowledge gap, however, concerns the  
42 mechanism of the oxidation of highly volatile and almost insoluble gaseous elemental  
43 mercury (GEM) into less volatile and more soluble oxidized mercury species and their  
44 incorporation into particles (Lin et al., 2006). As the oxidized mercury species are readily  
45 removed from the atmosphere by dry and wet deposition (Lin et al., 2006), the rate of the  
46 GEM oxidation controls the removal of mercury from the atmosphere. Three major oxidation  
47 mechanisms by reactions with  $O_3$ , OH (Lin et al, 2006), and Br/BrO (Holmes et al., 2006)  
48 have been proposed but their relative importance is still unclear (Lin et al., 2006).

49

50 Long term monitoring of mercury can provide valuable constraints on emissions and on the  
51 oxidation mechanism of atmospheric mercury. Mercury is now being monitored at many sites  
52 in the northern hemisphere (NH) but, to the best of our knowledge, only two longer term

53 systematic measurements have been reported from the southern hemisphere (SH) so far.  
54 Ebinghaus et al. (2002a) measured total gaseous mercury (TGM) at the German Antarctic  
55 research station from January 2000 to January 2001. Baker et al. (2002) reported first results  
56 of the long term monitoring of TGM at the Cape Point observatory in South Africa covering a  
57 period from September 1995 to June 1999. The latter measurements have been continued and  
58 here we present the trend and seasonal variation derived from measurements covering almost  
59 a 10 year period since September 1995 until the December 2004.

60

## 61 **Experimental**

62

63 The Cape Point station (34°21'S, 18°29'E) is operated as one of the World Meteorological  
64 Organisation (WMO) Global Atmospheric Watch (GAW) baseline monitoring sites. The  
65 station is located at the southern tip of the Cape Peninsula within the Cape Point National  
66 Park on top of a peak 230 m above sea level and about 60 km south of Cape Town. The  
67 station has been in operation since the end of the 1970s. It receives clean marine air masses  
68 for most of the time and has, therefore, been used to study the trends and seasonal variations  
69 of CO<sub>2</sub>, CO, CH<sub>4</sub>, N<sub>2</sub>O, O<sub>3</sub> and halocarbons at midlatitudes of the SH (e.g. Brunke et al, 1990;  
70 Oltmans et al, 1998).

71

72 As the primary objective of the TGM monitoring at Cape Point was to determine trends and  
73 seasonal variations, only 8 samples were typically taken every other week using a  
74 programmable sequential sampler providing about 200 samples per year. The sampling and  
75 analysis are described in detail by Baker et al. (2002). The technique measures the total  
76 gaseous mercury (TGM) under most conditions and was in excellent agreement with other  
77 TGM techniques during the intercomparison at Mace Head (Ebinghaus et al., 1999).

78

79 **Results and discussion**

80

81 TGM measurements at Cape Point are summarized in Table 1. During three years the  
82 measurements cover only a part of the year: September to December in 1995, January to  
83 March in 1997, and May, July, August and September in 1998. Because of the seasonal  
84 variation discussed later they cannot be regarded as representative for the whole year. The  
85 remaining medians for 1996 and 1999 - 2004 suggest a small but significant decreasing trend  
86 of TGM concentrations of  $0.015 \pm 0.004 \text{ ng m}^{-3} \text{ year}^{-1}$  ( $R^2 = 0.740$ ,  $n = 7$ ). Annual medians  
87 were used because they are less sensitive to extreme values than averages.

88

89 Fig. 1 shows the annual TGM medians for Cape Point together with the TGM medians at the  
90 Neumayer station, Antarctica (Ebinghaus et al., 2002a) and southern hemispheric TGM  
91 measurements during ship cruises on the Atlantic Ocean in 1994, 1996, and 1999 – 2001  
92 (Temme et al., 2003b). An exact comparison of all these measurements is not possible  
93 because of their different spatial and temporal coverage. However, the ship measurements in  
94 1996 and 2000 agree approximately with those from Cape Point while the data from the 1994  
95 are lower for unknown reasons. The TGM concentrations at the Neumayer station in  
96 Antarctica tend to be somewhat lower - possibly because of the mercury depletion events.

97

98 The seasonal variation of TGM and CO concentrations at Cape Point is shown in Fig. 2. For  
99 comparison, seasonal variation of TGM and CO concentrations at Mace Head at the west  
100 coast of Ireland in the NH is also shown. Mace Head is one of the ALE/GAGE/AGAGE  
101 stations used to study trends and seasonal variations of halocarbons,  $\text{CO}_2$ , CO,  $\text{CH}_4$ ,  $\text{H}_2$ ,  $\text{O}_3$   
102 (Simmonds et al., 2005), and mercury (Ebinghaus et al., 2002b). Only baseline data were  
103 taken to eliminate the influence of nearby sources. At Cape Point TGM measurements in air  
104 with  $^{222}\text{Rn}$  concentrations of less than  $600 \text{ mBq m}^{-3}$  were considered to be of baseline quality

105 in contrast to continental air masses, which exceed  $1200 \text{ mBq m}^{-3}$  (Brunke et al., 2004).  
106 Without  $^{222}\text{Rn}$  data, the baseline periods at Mace Head were identified by the NAME  
107 Lagrangian dispersion model (Manning et al., 2003). To eliminate the year to year variation  
108 of background TGM, the monthly means of TGM concentrations were normalized to annual  
109 medians. The TGM points in Fig. 2 then represent an average of relative monthly means over  
110 the period from March 1999 to December 2004, for which the  $^{222}\text{Rn}$  measurements were  
111 available, and the uncertainty bars represent their standard deviation. The TGM points and  
112 their uncertainty bars for Mace Head were derived in the same way as those for Cape Point.  
113 The absolute monthly means of TGM concentrations can be calculated by using the average  
114 annual median TGM concentrations over the period 1999 – 2004 of  $1.203 \pm 0.038 \text{ ng m}^{-3}$  at  
115 Cape Point and  $1.724 \pm 0.060 \text{ ng m}^{-3}$  at Mace Head, respectively. The CO data points and  
116 their uncertainty bars represent the baseline monthly averages of CO concentrations with their  
117 corresponding standard deviations.

118

119 To the best of our knowledge, the seasonal variation of TGM concentration in Fig. 2 is the  
120 first one derived from a long term monitoring in the SH. It shows maximum TGM  
121 concentrations in January and February (austral summer) and a minimum during austral  
122 winter. Although measurements by Ebinghaus et al. (2002a) and Temme et al. (2003a) in the  
123 Antarctic cover a whole year, the seasonal variation of their measurements is obscured by the  
124 frequent mercury depletion events starting in July and ending in February. A preliminary  
125 analysis of Cape Point data from September 1995 to June 1999 suggested a TGM minimum in  
126 March – May and a maximum in June – August (Baker et al., 2002), but no attempt was made  
127 to filter out the baseline data.

128

129 In spite of the occasionally large uncertainty bars for the monthly mean TGM concentrations,  
130 Fig. 2 shows that the seasonal variation of TGM relative to that of CO is opposite in phase in

131 the SH and in phase in the NH. The amplitude of the seasonal variations of 0.134 at Cape  
132 Point appears to be somewhat larger than those for Mace Head (0.097), although this  
133 difference may not be significant in view of the large uncertainties of the monthly means. The  
134 amplitude of the CO concentrations at Cape Point is 32.2 ppb in absolute and 0.49 in relative  
135 terms, at Mace Head 69.4 ppb and 0.58, respectively. The inverse average ratio of relative  
136 TGM (0.116) to CO (0.54) amplitudes of 4.7 suggest a correspondingly longer lifetime of  
137 TGM relative to that of CO, i.e. about 0.83 yr if the lifetime of CO of 0.18 yr is assumed  
138 (Warneck, 1988).

139

140 The seasonal variation of CO concentrations is driven mainly by the seasonal variation of its  
141 major sink due to the reaction with OH. The major source of OH radicals is photolysis of O<sub>3</sub>  
142 leading partly to O(<sup>1</sup>D) atoms and their subsequent reaction with H<sub>2</sub>O. The seasonalities of  
143 solar irradiation (i.e. wavelength < 310 nm) and of water vapor concentrations lead to a  
144 pronounced seasonal variation of OH with a maximum concentration in summer and a  
145 minimum one in winter (Spivakovsky et al., 2000). The CO concentration follows the inverse  
146 pattern with a delay of about 2 months. The similar seasonal behavior of TGM and CO in the  
147 NH was sometimes taken as a sign in favor of OH driven oxidation for TGM (Bergan and  
148 Rodhe, 2001). The opposite behavior in the SH, however, suggests that this may be incorrect.

149

150 We hypothesize that the seasonal variation of TGM in the SH might be driven predominantly  
151 by its emissions. A summary of the currently available information about the seasonality of  
152 mercury emissions is shown in Fig. 3. The seasonality and magnitude of oceanic mercury  
153 emissions were taken from the results of model by Strode et al. (2006). The emissions from  
154 biomass burning were calculated using the seasonal variation of CO emissions given by  
155 Duncan et al. (2003) and Hg/CO emission ratios of 1.89 and  $1.36 \times 10^{-7}$  mol/mol in the  
156 southern and northern hemispheres, respectively (Ebinghaus et al., 2007). The anthropogenic

157 mercury emissions for 2000 (Wilson et al., 2006) were allocated to the hemispheres and to  
158 combustion and other sources. For the combustion sources the hemispheric seasonal  
159 variations given by Rotty (1987) were used to calculate their monthly contributions. The  
160 interhemispheric flux was calculated using the seasonal cycle of the interhemispheric  
161 exchange time derived by Levin and Hessheimer (1996) from  $^{85}\text{Kr}$  measurements and the  
162 seasonally variable north-south TGM gradient. Because of the higher TGM concentrations in  
163 the NH the flux is always negative from the NH perspective and positive for the SH. The total  
164 emissions, including the north to south transport, vary from 152 t/yr in May to 296 t/yr in  
165 December in the SH, and from 105 t/yr in December to 234 t/yr in May in the NH.

166

167 The emissions considered in Fig. 3 yield a total of 4562 t/yr, representing 69% of the total  
168 emissions estimated by Mason and Sheu (2002). The remaining 1605 t/yr are broadly ascribed  
169 to land emissions (Mason and Sheu, 2002) including volcanic emissions of 110 – 700 t/yr  
170 (Nriagu and Becker, 2003; Pyle and Mather, 2003), primary emissions from mercury deposits  
171 of about 500 t/yr (Lindquist et al., 1991), and reemissions of previously deposited mercury.  
172 The latter two are known to vary with temperature, solar radiation, and precipitation (Gustin  
173 et al., 1997) leading to a seasonal variation with maximum emissions in summer and a  
174 minimum in winter. As the contributions of volcanic emissions, primary emissions, and  
175 reemissions are not well known, the seasonality of the land emissions cannot be estimated.  
176 But it will be in phase with the emissions in the SH and thus increase the seasonal amplitude  
177 of the overall emissions there. In the NH, it will be out of phase with the emissions and will  
178 thus reduce the amplitude of the emission seasonality.

179

180 In summary, the seasonality of TGM emissions in Fig. 3 is in reasonable agreement with the  
181 baseline TGM observations at Cape Point and Mace Head suggesting a lesser influence of the  
182 sink seasonality. Of the three potential sinks due to reactions with OH, O<sub>3</sub>, and Br (Lin et al,



183 2006; Holmes et al., 2006) the most pronounced seasonality can be expected for OH. The  
184 resemblance of the TGM seasonality to the seasonality of the sources is thus more consistent  
185 with O<sub>3</sub> and Br oxidation of elemental mercury than with oxidation by OH.

186

## 187 **Conclusion**

188

189 The measurements of total gaseous mercury (TGM) made at the WMO Global Atmosphere  
190 Watch (GAW) station at Cape Point, South Africa, since September 1995 suggest a small but  
191 significant decrease of annual median TGM concentrations from 1.29 ng m<sup>-3</sup> in 1996 to 1.19  
192 ng m<sup>-3</sup> in 2004.

193

194 Baseline TGM concentrations at Cape Point show a pronounced seasonal variation with a  
195 maximum in austral summer and a minimum in austral winter. This seasonal variation is  
196 opposite in phase to that of CO at Cape Point, whereas in the NH at Mace Head the seasonal  
197 variations of TGM and CO are in phase. This behavior seems to be dominated by the seasonal  
198 variation of major mercury sources in both hemispheres, suggesting lesser influence of the  
199 sink seasonality. We are aware, that this explanation is speculative in view of the combined  
200 uncertainties of the emission estimates. But, if the seasonality observed at Cape Point is  
201 confirmed at other sites in the SH, the different seasonalities in the northern and southern  
202 hemispheres of TGM concentrations may pose an important constraint on the global models  
203 of atmospheric mercury.

204

## 205 **Acknowledgments**

206

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210 established with the financial help of the Deutsche Forschungsgemeinschaft.

211

## 212 **References**

213

214 Baker, P.G.L., E.-G. Brunke, F. Slemr, A.M. Crouch, Atmospheric mercury measurements at  
215 Cape Point, South Africa, *Atmos. Environ.* 36, 2459-24565, 2002.

216

217 Bergan, T., and H. Rodhe, Oxidation of elemental mercury in the atmosphere: Constraints  
218 imposed by global scale modeling, *J. Atm. Chem.* 40, 191-212, 2001.

219

220 Brunke, E.-G., H.E. Scheel, W. Seiler, Trends of tropospheric CO, N<sub>2</sub>O and CH<sub>4</sub> as observed  
221 at Cape Point, South Africa, *J. Atmos. Chem.* 11, 197-210, 1990.

222

223 Brunke, E.-G., C.Labuschagne, B. Parker, H.E. Scheel, S. Whittlestone, Baseline air mass  
224 selection at Cape Point, South Africa: application of <sup>222</sup>Rn and other filter criteria to CO<sub>2</sub>,  
225 *Atmos. Environ.* 38, 5693-5702, 2004.

226

227 Duncan, B.N., R.V. Martin, A.C. Staudt, R. Yevich, J.A. Logan, Interannual and seasonal  
228 variability of biomass burning emissions constrained by satellite observations, *J. Geophys.*  
229 *Res.* 108, doi:10.1029/2002JD002378, 2003.

230

231 Ebinghaus, R., F. Slemr, C.A.M. Brenninkmeijer, P. van Velthoven, A. Zahn, M. Hermann,  
232 Emissions of gaseous mercury from biomass burning in South America in 2005 observed  
233 during the CARIBIC flights, *Geophys. Res. Lett.* 34, L08813, doi:10.1029/2006GL028866,  
234 2007.

235

236 Ebinghaus, R., S.G. Jennings, W.H. Schroeder, T. Berg, T. Donaghy, J. Guentzel, C. Kenny,  
237 H.H. Kock, K. Kvietkus, W. Landing, T. Mühleck, J. Munthe, E.M. Prestbo, D.  
238 Schneeberger, F. Slemr, J. Sommar, A. Urba, D. Wallschläger, Z. Xiao, International field  
239 intercomparison measurements of atmospheric Hg species at Mace Head, Ireland, *Atmos.*  
240 *Environ.* 33, 3063-3073, 1999.

241

242 Ebinghaus, R., H.H. Kock, C. Temme, J.W. Einax, A.G. Löwe, A. Richter, J.P. Burrows,  
243 W.H. Schroeder, Antarctic springtime depletion of atmospheric mercury, *Environ. Sci.*  
244 *Technol.* 36, 1238-1244, 2002a.

245

246 Ebinghaus, R., H.H. Kock, A.M. Coggins, T.G. Spain, S.G. Jennings, Ch. Temme, Long-term  
247 measurements of atmospheric mercury at Mace Head, Irish west coast, between 1995 and  
248 2001, *Atmos. Environ.* 36, 5267-5276, 2002b.

249

250 Gustin, M.S., G.E. Taylor Jr., R.A. Maxey, Effect of temperature and air movement on the  
251 flux of elemental mercury from substrate to the atmosphere, *J. Geophys. Res.* 102, 3891-3898,  
252 1997.

253

254 Holmes, C.D., D.J. Jacob, X. Yang, Global lifetime of elemental mercury against oxidation by  
255 atomic bromine in the free troposphere, *Geophys. Res. Lett.* 33, doi:10.1029/2006GL027176,  
256 2006.

257

258 Levin, I., V. Hessheimer, Refining of atmospheric transport model entries by the globally  
259 observed passive tracer distributions of <sup>85</sup>krypton and sulfur hexafluoride (SF<sub>6</sub>), *J. Geophys.*  
260 *Res.* 101, 16745-16755, 1996.

261

262 Lin, C.-J., P. Pongprueksa, S.E. Lindberg, S.O. Pehkonen, D. Byun, C. Jang, Scientific  
263 uncertainties in atmospheric mercury models I: Model science evaluation, *Atmos. Environ.*  
264 *40*, 2911-2928, 2006.

265

266 Lindberg, S., R. Bullock, R. Ebinghaus, D. Engstrom, X. Feng, W. Fitzgerald, N. Pirrone, E.  
267 Prestbo, C. Seigneur, A synthesis of progress and uncertainties in attributing the sources of  
268 mercury in deposition, *Ambio* *36*, 19-32, 2007.

269

270 Lindqvist, O. (ed.), Hg in the Swedish environment: recent research on causes, consequences  
271 and corrective methods, *Water Air Pollut.* *55*, 1-261, 1991.

272

273 Manning, A.J., D.B. Ryall, R.G. Derwent, P.G. Simmonds, S.O. O'Doherty, Estimating  
274 European emissions of ozone-depleting and greenhouse gases using observations and a  
275 modeling back-attribution technique, *J. Geophys. Res.* *108*, doi:10.1029/2002JD002312,  
276 2003.

277

278 Mason, R.P. and G.-R. Sheu, Role of the ocean in the global mercury cycle, *Global*  
279 *Biogeochem. Cycles* *16*, doi:10.1029/2001GB001440, 2002.

280

281 Mergler, D., H.A. Anderson, L.H.M. Chan, K.R. Mahaffey, M. Murray, M. Sakamoto, A.H.  
282 Stern, Methylmercury exposure and health effects in humans: a worldwide concern, *Ambio*  
283 *36*, 3-11, 2007

284

285 Nriagu, J. and C. Becker, Volcanic emissions of mercury to the atmosphere: Global and  
286 regional inventories, *Sci. Total. Environ.* *304*, 3-12, 2003.

287

288 Oltmans, S.J., A.S. Lefohn, H.E. Scheel, J.M. Harris, H. Levy II, I.E. Galbally, E.G. Brunke,  
289 C.P. Meyer, J.A. Lathrop, B.J. Johnson, D.S. Shadwick, E. Cuevas, F.J. Schmidlin, D.W.  
290 Tarasick, H. Claude, J.B. Kerr, O. Uchino, Trends of ozone in the troposphere, *Geophys. Res.*  
291 *Lett.* 25, 139-142, 1998.

292

293 Pyle, D.M. and T.A. Mather, The importance of volcanic emissions for the global  
294 atmospheric mercury cycle, *Atmos. Environ.* 37, 5115-5124, 2003.

295

296 Rotty, R.M., Estimates of seasonal variation in fossil fuel CO<sub>2</sub> emission, *Tellus* 39B, 184-202,  
297 1987.

298

299 Scheuhammer, A.M., M.W. Meyer, M.B. Sandheinrich, M.W. Murray, Effects of  
300 environmental methylmercury on the health of wild birds, mammals, and fish, *Ambio* 36, 12-  
301 18, 2007.

302

303 Simmonds, P.G., A.J. Manning, R.G. Derwent, P. Ciais, M. Ramonet, V. Kazan, D. Ryall, A  
304 burning question. Can recent growth rate anomalies in the greenhouse gases be attributed to  
305 large-scale biomass burning events?, *Atmos. Environ.* 39, 2513-2517, 2005.

306

307 Slemr, F., G. Schuster, W. Seiler, Distribution, speciation, and budget of atmospheric  
308 mercury, *J. Atmos. Chem.* 3, 407-434, 1985.

309

310 Spivakovsky, C.M., J.A. Logan, S.A. Montzka, Y.J. Balkanski, M. Foreman-Fowler, D.B.A.  
311 Jones, L.W. Horowitz, A.C. Fusco, C.A.M. Brenninkmeijer, M.J. Prather, S.C. Wofsy, M.B.

312 McElroy, Three-dimensional climatological distribution of tropospheric OH: Update and  
313 evaluation, *J. Geophys. Res.* 105, 8931-8980, 2000.

314

315 Strode, S.A., L. Jaeglé, N.E. Selin, D.J. Jacob, R.J. Park, R.M. Yantosca, R.P. Mason, F.  
316 Slemr, Air-sea exchange in the global mercury cycle, *Global Biogeochem. Cycles* 21,  
317 GB1017, doi:10.1029/2006GB002766, 2007.

318

319 Temme, Ch., J.W. Einax, R. Ebinghaus, W.H. Schroeder, Measurements of atmospheric  
320 mercury species at a coastal site in the Antarctic and over the South Atlantic Ocean during  
321 polar summer, *Environ. Sci. Technol.* 37, 22-31, 2003a.

322

323 Temme, Ch., F. Slemr, R. Ebinghaus, J.W. Einax, Distribution of mercury over the Atlantic  
324 Ocean in 1996 and 1999-2001, *Atmos. Environ.* 37, 1889-1897, 2003b.

325

326 Warneck, P., *Chemistry of the Natural Atmosphere*, Academic Press, San Diego, 1988.

327

328 Wilson, S.J., F. Steenhuisen, J.M. Pacyna, E.G. Pacyna, Mapping the spatial distribution of  
329 global anthropogenic mercury atmospheric emission inventories, *Atmos. Environ.* 40, 4621-  
330 4632, 2006.

331

### 332 **Figure Labels**

333

334 Figure 1: TGM concentrations measured at Cape Point, Neumayer station (Antarctica) and  
335 onboard ships over the Atlantic Ocean in the southern hemisphere. The concentrations are  
336 given in ng/m<sup>3</sup> (STP, i.e. 273.2 K, 1013 mbar). The points and bars represent the annual  
337 medians (cruise medians for ship cruises) and their 95% confidence intervals.

338

339 Figure 2: Seasonal variation of TGM (relative monthly averages and their standard deviation,  
340 see text) and CO (absolute monthly averages and their standard deviations) at Cape Point and  
341 Mace Head. Only baseline data are considered in this diagram, see text.

342

343 Figure 3: Seasonal variation of the emissions from oceans, biomass burning, anthropogenic  
344 activities, and of the interhemispheric flux. Details of calculations are given in the text.

345

### 346 **Tables**

347

348 Table 1: Summary of the TGM measurements at Cape Point.

349

Year	Range [ng m <sup>-3</sup> ]	Average ± Standard deviation [ng m <sup>-3</sup> ]	Median [ng m <sup>-3</sup> ]	95% confidence interval of median [ng m <sup>-3</sup> ]	Number of measurements
1995	1.23 – 3.16	1.518 ± 0.372	1.42	1.34 - 1.54	26
1996	0.93 – 1.88	1.297 ± 0.136	1.29	1.26 – 1.31	212
1997	0.73 – 1.60	1.182 ± 0.174	1.17	1.11 – 1.22	50
1998	0.95 – 1.82	1.308 ± 0.167	1.28	1.24 – 1.37	38
1999	0.78 – 1.61	1.244 ± 0.139	1.25	1.23 – 1.27	163
2000	0.49 – 2.18	1.227 ± 0.174	1.21	1.18 – 1.22	173
2001	0.84 – 6.78	1.452 ± 0.817	1.24	1.21 – 1.28	152
2002	0.92 – 1.64	1.200 ± 0.132	1.18	1.14 – 1.20	143
2003	0.93 – 1.60	1.176 ± 0.110	1.15	1.14 – 1.18	125
2004	0.78 – 2.37	1.261 ± 0.247	1.19	1.17 – 1.22	155

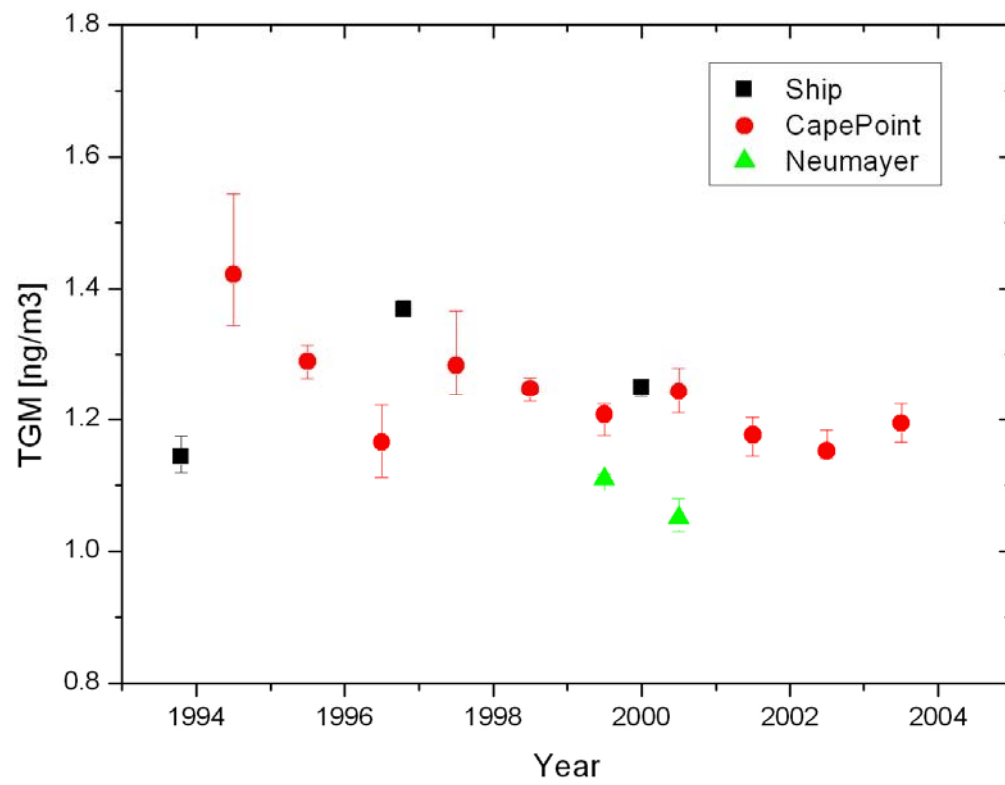
350

351 **Figures**

352

353 Fig. 1:

354



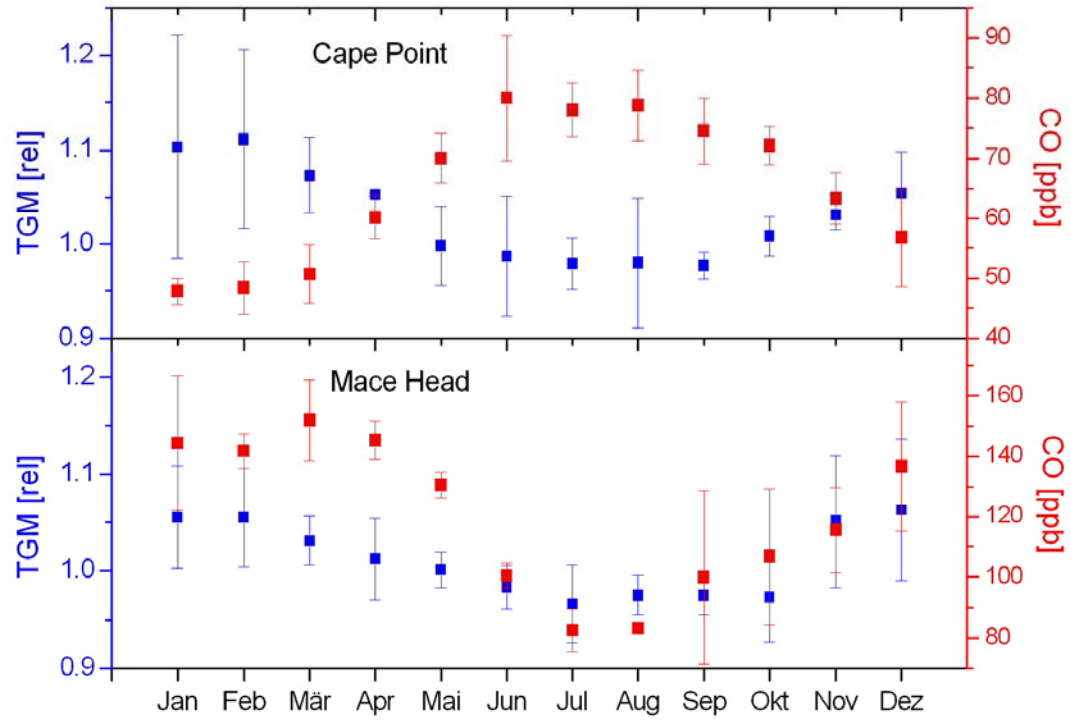
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356

357 Fig. 2:

358

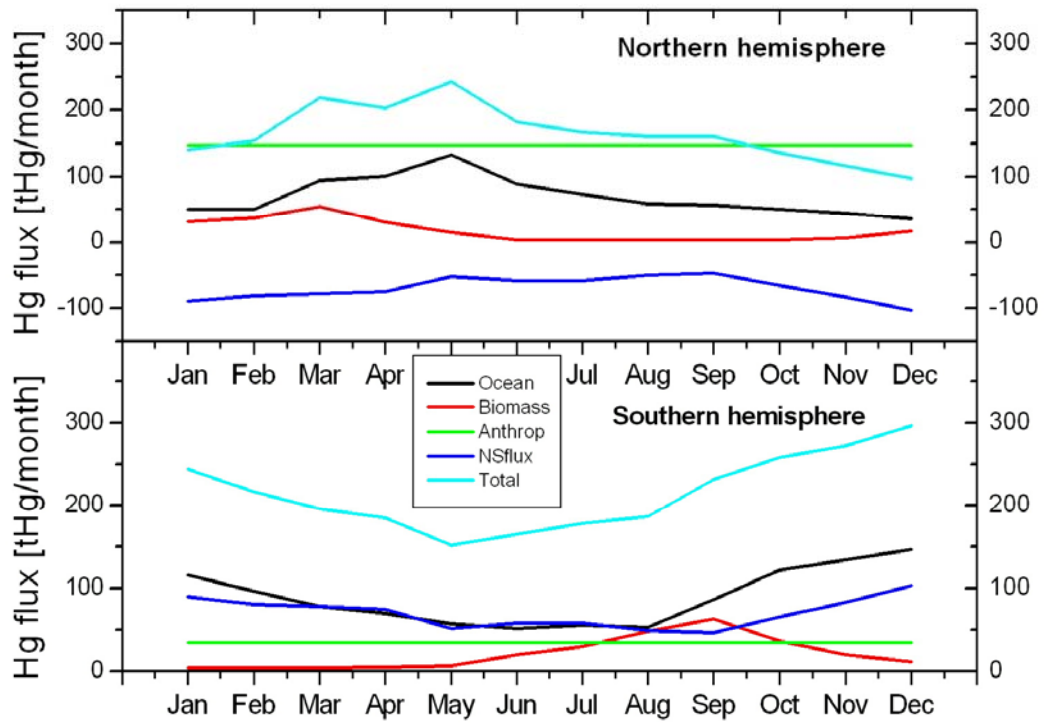


359

360

361 Fig. 3:

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363

364