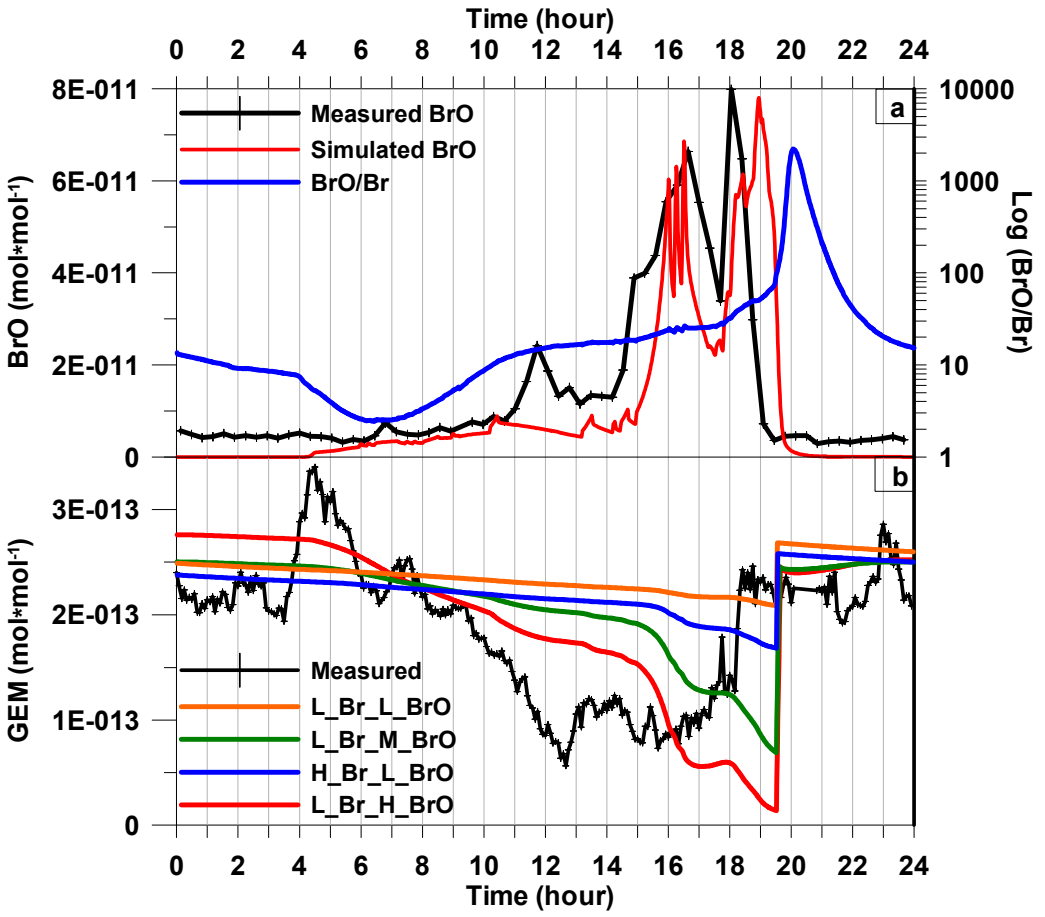
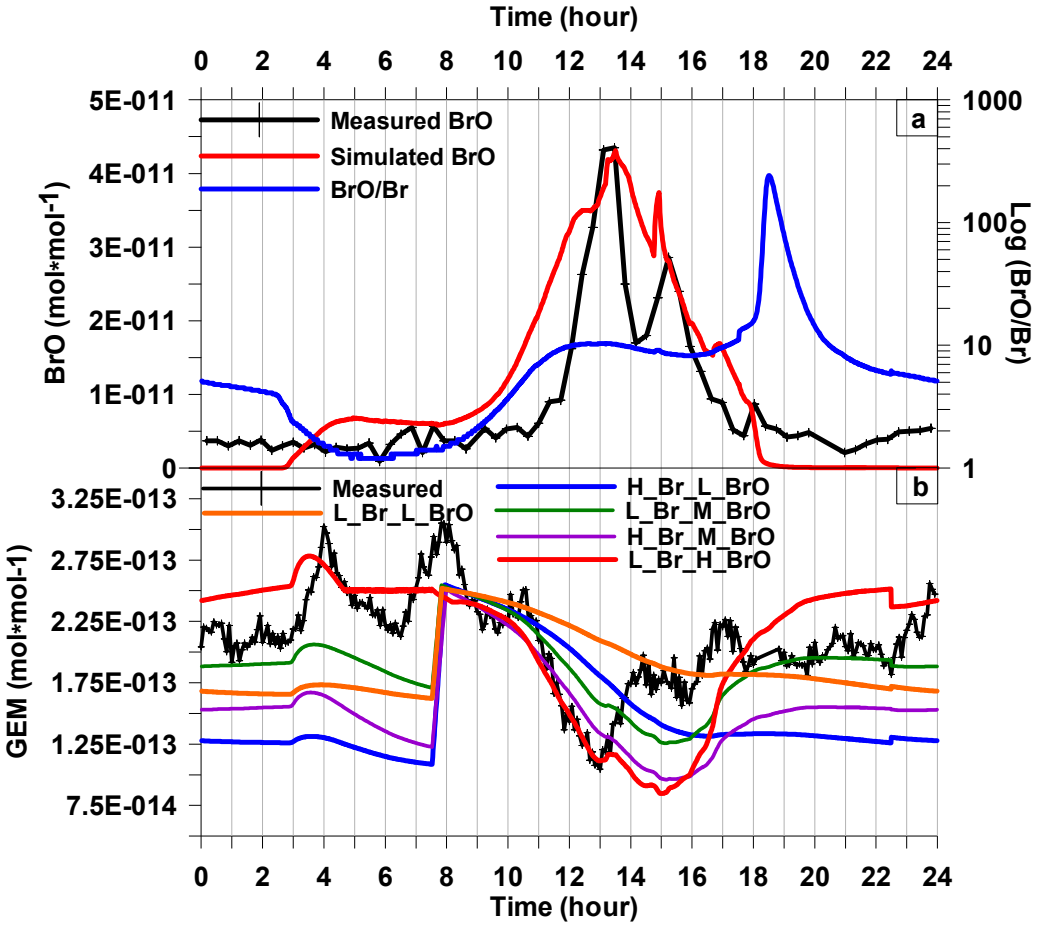
Supplementary Information



Supplementary Figure 1. Simulated vs. measured diurnal profiles of GEM and BrO. (a) Measured and simulated diurnal profiles of BrO, and simulated diurnal profile of [BrO]/[Br] for Julian day 198. (b) Different combinations of values for $k_{\_Hg+BrO}$ and $k_{\_Hg+Br}$ were used to evaluate the simulated GEM diurnal profiles (Sect. 2.3).



Supplementary Figure 2. Simulated vs. measured diurnal profiles of GEM and BrO. (a) Measured and simulated diurnal profiles of BrO as well as simulated diurnal profile of [BrO]/[Br] for Julian day 197. (b) Different combinations of values for $k_{\_Hg+BrO}$ and $k_{\_Hg+Br}$ were used to evaluate the simulated GEM diurnal profiles (Sect. 2.3).

Reaction		k [$cm^3 molecule^{-1} s^{-1}$] or s^{-1}	Reference
G1	$Hg+O_3 \rightarrow HgO+O_2$	$8.43E-17 \times \exp(-1407K/T)$	A
G2	$Hg+OH \rightarrow HgO+H$	$3.55E-14 \times \exp(294K/T)$	A
G3	$Hg+NO_3 \rightarrow HgO+NO_2$	$4.0E-15$	B
G4	$Hg + Br \rightarrow HgBr$	$2.7E-13$	C
G5	$HgBr \rightarrow Hg + Br$	$1.2 \times E10 \times \exp(-8357/T)\ s^{-1}$	D
G6	$HgBr + BrO \rightarrow BrHgOBr$	$3.0E-12$	E
G7	$Hg + BrO \rightarrow HgO + Br$	$1.5E-13$	F
G8	$Hg+H_2O_2 \rightarrow HgO+H_2O$	$8.5E-19$	G
G9	$Hg + H_2O_2 \rightarrow Hg(OH)_2$	$6.1E-19$	H
G10	$Hg + Br_2 \rightarrow HgBr_2$	$9.0E-17$	I
G11	$HgBr \rightarrow Hg + Br$	$6.09E-4$	F
G23	$HgBr + Br \rightarrow HgBr_2$	$3.0E-12$	J
G13	$HgBr+Cl \rightarrow ClHgBr$	$3.0E-12$	K
G14	$HgCl+Br \rightarrow ClHgBr$	$3.0E-12$	K
G15	$Hg + HCl \rightarrow HgCl_2$	$1.0E-19$	H
G16	$Hg + Cl \rightarrow HgCl$	$1.0E-11$	I
G17	$Hg + Cl_2 \rightarrow HgCl_2$	$2.6E-18$	I
G18	$Hg + ClO \rightarrow HgOCl$	$1.9E-14$	L
G19	$Hg + I_2 \rightarrow HgI2$	1.27E-19	M
G20	$Hg + I \rightarrow HgI$	$4.0E-13 \times (T/298K)^{-2.38}$	D

Supplementary Table 1. Gas phase reactions. A. Pal and Ariya (2004); B. Sommar et al. (1997); C. Donohoue et al. (2006); D. Goodsite et al. (2004); E. Shon et al. (2005); F. Raofie and Ariya (2003); G. Tokos et al. (1998); H. Xie et al. (2008); I. Ariya et al. (2002); J. Hedgecock et al. (2005); J. Calvert and Lindberg (2003). L. Byun et al. (2010); M. Raofie et al. (2008).

Reaction		K ($M^{-1}s^{-1}$) / s^{-1}	Reference
A1	$Hg+O_3 \rightarrow HgO+O_2$	$4.7E7$	A
A2	$HgO+H^+ \rightarrow Hg^{2+}+OH^-$	$1.0E10$	B
A3	$Hg+OH \rightarrow Hg^{+}+OH^-$	$2.0E9$	C
A4	$Hg^{+}+OH \rightarrow Hg^{++}+OH^-$	$1.0E10$	C
A5	$Hg^{2+}+HO_2 \rightarrow Hg^{+}+O_2+H^+$	$1.1E4$	D
A6	$Hg^{+}+HO_2 \rightarrow Hg+O_2+H^+$	$1.0E10$	E
A7	$Hg+HOCl \rightarrow Hg^{2+}+Cl^-+OH^-$	$2.09E6$	F
A8	$Hg+ClO^- +H^+ \rightarrow Hg^{2+}+Cl^-+OH^-$	$1.99E6$	F
A9	$Hg+HOBr^- \rightarrow Hg_2^{+}+Br^-+OH^-$	0.279	G
A10	$Hg^{++}+O_2^- \rightarrow Hg^{+}+O_2$	$1.1E4$	D
A11	$Hg+Br_2 \rightarrow Hg^{2+}+2Br^-$	0.196	G
A12	$HgSO_3+H2O \rightarrow Hg+HSO_4^- +H^+$	0.0106	H
A13	$Hg(OH)_2 \xrightarrow{h\nu} Hg+2\ OH$	$3E-7\ s^{-1}$	I

Supplementary Table 2. Aqueous phase reactions. A. Munthe (1992); B. Pleijel and Munthe (1995); C. Lin and Pehkonen (1997); D. Pehkonen and Lin (1997); E. Xie et al. (2008); F. Lin and Pehkonen (1999); G. Wang and Pehkonen (2004); H. van Loon et al. (2000); I. Xiao et al. (1994).

Reaction		K [M^{-1}]	Reference
EQ1	$Hg(II)+OH^- \leftrightarrow Hg(OH)^+$	3.9*E*10	A
EQ2	$Hg(OH)^+ + OH^- \leftrightarrow Hg(OH)2$	1.6*E*11	A
EQ3	$Hg^{2+} + SO_3^{2-} \leftrightarrow HgSO_3$	2.1*E*13	B
EQ4	$HgSO_3 + SO_3^{2-} \leftrightarrow Hg(SO_3)2^{2-}$	1.0*E*10	B
EQ5	$Hg(OH)^+ + Cl^- \leftrightarrow HgOHCl$	2.7*E*7	A
EQ6	$Hg(II)+Cl^- \leftrightarrow HgCl^+$	2.0*E*7	C
EQ7	$HgCl^+ + Cl^- \leftrightarrow HgCl_2$	2.5*E*6	A
EQ8	$HgCl_2 + Cl^- \leftrightarrow HgCl_3^-$	6.7*E*0	D
EQ9	$HgCl_3 - + Cl^- \leftrightarrow HgCl4_2^-$	1.3*E*1	D
EQ10	$Hg(II)+Br^- \leftrightarrow HgBr^+$	1.1*E*9	D
EQ11	$HgBr^+ + Br^- \leftrightarrow HgBr_2$	2.5*E*8	D
EQ12	$HgBr_2 + Br^- \leftrightarrow HgBr_3^-$	1.5*E*2	D
EQ13	$HgBr_3^- + Br^- \leftrightarrow HgBr_4^{2-}$	2.3*E*1	D

Supplementary Table 3. Aqueous-phase equilibria. A. Pleijel and Munthe (1995); B. van Loon et al. (2001); C. Smith and Martell (1976); D. Clever et al. (1985).

Reaction		K_H[M/atm]	Reference
H1	$Hg \rightarrow Hg_{(aq)}$	$3.2E-1$	A
H2	$HgO_{(g)} \rightarrow HgO_{(aq)}$	$2.69E12$	B
H3	$HgCl_2 \rightarrow$ HgCl2(aq)	$2.75E6$	C
H4	$HgBr_2 \rightarrow HgBr_2(aq)$	$2.75E6$	D
H5	$Hg(OH)_2 \rightarrow Hg(OH)_{2(aq)}$	$1.2E4$	E
H6	$(CH3)_2Hg \rightarrow (CH_3)_2Hg_{(aq)}$	$1.3E-1$	E
H7	$CH_3HgCl \rightarrow CH_3HgCl_{(aq)}$	$2.2E3$	E
H8	$CH_3HgOH \rightarrow CH_3HgOH_{(aq)}$	$1.5E5$	F
H9	$BrHGOBr \rightarrow BrHgOBr_{(aq)}$	$2.75E6$	G

Supplementary Table 4. Heterogeneous Reactions. A. Schroeder et al. (1991); B. Schroeder and Munthe (1998); C. Hedgecock et al. (2005); D. Hedgecock and Pirrone. (2004); E. Seigneur (1994); F. Petersen (1998). G. K_H assumed to be as $HgCl_2$ (Xie et al., 2008).