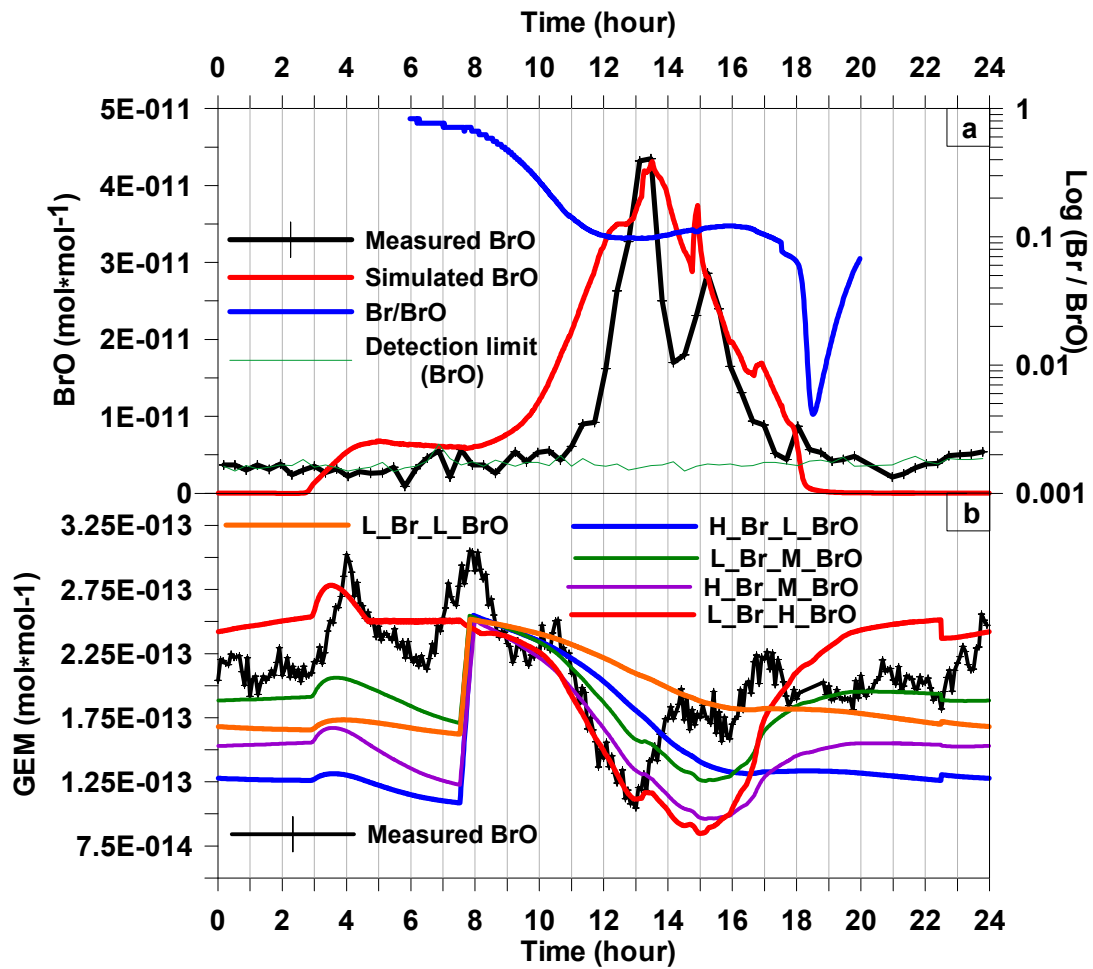


Supplementary Information



Supplementary Figure 1. Simulated vs. measured diurnal profiles of GEM and BrO. (a) Measured and simulated diurnal profiles of BrO as well as simulated diurnal profile of $[\text{Br}]/[\text{BrO}]$ for Julian day 197. (b) Different combinations of values for $k_{\text{Hg}+\text{BrO}}$ and $k_{\text{Hg}+\text{Br}}$ were used to evaluate the simulated GEM diurnal profiles (Sect. 2.3).

Reaction	k [$\text{cm}^3 \text{molecule}^{-1} \text{s}^{-1}$] or s^{-1}	Reference
G1	$\text{Hg} + \text{O}_3 \rightarrow \text{HgO} + \text{O}_2$	$8.43E - 17 \times \exp(-1407K/T)$ A
G2	$\text{Hg} + \text{OH} \rightarrow \text{HgO} + \text{H}$	$3.55E - 14 \times \exp(294K/T)$ A
G3	$\text{Hg} + \text{NO}_3 \rightarrow \text{HgO} + \text{NO}_2$	$4.0E - 15$ B
G4	$\text{Hg} + \text{Br} \rightarrow \text{HgBr}$	$2.7E - 13$ C
G5	$\text{HgBr} \rightarrow \text{Hg} + \text{Br}$	$1.2 \times E10 \times \exp(-8357/T) \text{ s}^{-1}$ D
G6	$\text{HgBr} + \text{BrO} \rightarrow \text{BrHgOBr}$	$3.0E - 12$ E
G7	$\text{Hg} + \text{BrO} \rightarrow \text{HgO} + \text{Br}$	$1.5E - 13$ F
G8	$\text{Hg} + \text{H}_2\text{O}_2 \rightarrow \text{HgO} + \text{H}_2\text{O}$	$8.5E - 19$ G
G9	$\text{Hg} + \text{H}_2\text{O}_2 \rightarrow \text{Hg}(\text{OH})_2$	$6.1E - 19$ H
G10	$\text{Hg} + \text{Br}_2 \rightarrow \text{HgBr}_2$	$9.0E - 17$ I
G11	$\text{HgBr} \rightarrow \text{Hg} + \text{Br}$	$6.09E - 4$ F
G23	$\text{HgBr} + \text{Br} \rightarrow \text{HgBr}_2$	$3.0E - 12$ J
G13	$\text{HgBr} + \text{Cl} \rightarrow \text{ClHgBr}$	$3.0E - 12$ *
G14	$\text{HgCl} + \text{Br} \rightarrow \text{ClHgBr}$	$3.0E - 12$ *
G15	$\text{Hg} + \text{HCl} \rightarrow \text{HgCl}_2$	$1.0E - 19$ K
G16	$\text{Hg} + \text{Cl} \rightarrow \text{HgCl}$	$1.0E - 11$ I
G17	$\text{Hg} + \text{Cl}_2 \rightarrow \text{HgCl}_2$	$2.6E - 18$ I
G18	$\text{Hg} + \text{ClO} \rightarrow \text{HgOCl}$	$1.9E - 14$ L
G19	$\text{Hg} + \text{I}_2 \rightarrow \text{HgI}_2$	$1.27E - 19$ M
G20	$\text{Hg} + \text{I} \rightarrow \text{HgI}$	$4.0E - 13 \times (T / 298K)^{-2.38}$ 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). K. Hall and Bloom (1993). L. Byun et al. (2010); M. Raofie et al. (2008).

* Since the kinetics of these reactions have is not known, we have used the value $k(\text{HgBr} + \text{Br})$ as assumed by Calvert and Lindberg (2003)(see Xie et al., 2008).

Reaction No.		$K (M^{-1}s^{-1}) / s^{-1}$	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	$HgSO_3+H_2O \rightarrow Hg+HSO_4^-+H^+$	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 ⁻ ↔ Hg(OH) ⁺	3.9E10	A
EQ2	Hg(OH) ⁺ +OH ⁻ ↔ Hg(OH) ₂	1.6E11	A
EQ3	Hg ²⁺ +SO ₃ ²⁻ ↔ HgSO ₃	2.1E13	B
EQ4	HgSO ₃ + SO ₃ ²⁻ ↔ Hg(SO ₃) ₂ ²⁻	1.0E10	B
EQ5	Hg(OH) ⁺ +Cl ⁻ ↔ HgOHCl	2.7E7	A
EQ6	Hg(II)+Cl ⁻ ↔ HgCl ⁺	2.0E7	C
EQ7	HgCl ⁺ +Cl ⁻ ↔ HgCl ₂	2.5E6	A
EQ8	HgCl ₂ +Cl ⁻ ↔ HgCl ₃ ⁻	6.7E0	D
EQ9	HgCl ₃ ⁻ +Cl ⁻ ↔ HgCl ₄ ²⁻	1.3E1	D
EQ10	Hg(II)+Br ⁻ ↔ HgBr ⁺	1.1E9	D
EQ11	HgBr ⁺ + Br ⁻ ↔ HgBr ₂	2.5E8	D
EQ12	HgBr ₂ +Br ⁻ ↔ HgBr ₃ ⁻	1.5E2	D
EQ13	HgBr ₃ ⁻ +Br ⁻ ↔ HgBr ₄ ²⁻	2.3E1	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).

Reference		K_H [M/atm]	Reference
H1	$\text{Hg} \rightarrow \text{Hg}_{(\text{aq})}$	$3.2E-1$	A
H2	$\text{HgO}_{(\text{g})} \rightarrow \text{HgO}_{(\text{aq})}$	$2.69E12$	B
H3	$\text{HgCl}_2 \rightarrow \text{HgCl}_{2(\text{aq})}$	$2.75E6$	C
H4	$\text{HgBr}_2 \rightarrow \text{HgBr}_{2(\text{aq})}$	$2.75E6$	D
H5	$\text{Hg}(\text{OH})_2 \rightarrow \text{Hg}(\text{OH})_{2(\text{aq})}$	$1.2E4$	E
H6	$(\text{CH}_3)_2\text{Hg} \rightarrow (\text{CH}_3)_2\text{Hg}_{(\text{aq})}$	$1.3E-1$	E
H7	$\text{CH}_3\text{HgCl} \rightarrow \text{CH}_3\text{HgCl}_{(\text{aq})}$	$2.2E3$	E
H8	$\text{CH}_3\text{HgOH} \rightarrow \text{CH}_3\text{HgOH}_{(\text{aq})}$	$1.5E5$	F
H9	$\text{BrHgOBr} \rightarrow \text{BrHgOBr}_{(\text{aq})}$	$2.75E6$	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).