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*Supplement of*

## **Current understanding of the driving mechanisms for spatiotemporal variations of atmospheric speciated mercury: a review**

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Table S1. Marine boundary layer TGM or GEM (ng m<sup>-3</sup>; units are noted otherwise). Units were converted in STP. The units for time resolution are hours (denoted as h), minutes (denoted as min), and seconds (denoted as s). GEM measurements are indicated with a superscript G.

	Platform	Latitude	Sampling Period	Time Resolution	Range	Mean ( $\pm\sigma$ )	Reference
Atlantic	Ship	54°N, 10°E - 20°N, 67°W	Oct – Nov 1973	2 – 4-h	1 – 10	2.8	Seiler et al. (1980)
	Ship	35°S – 11° N	1977 - 1980	40-min	0.8 – 2.09	1.31 $\pm$ 0.21	Slemr et al. (1981, 1985)
	Ship	11°N – 54°N	1977 - 1980	20 – 40-min	1.0 – 3.41	1.96 $\pm$ 0.35	Slemr et al. (1981, 1985)
	Ship	33.36°S – 8.41°N, 20.23°W – 51.55°W	16 May – 20 Jun 1996	24-h	1.17 – 1.99	1.61 $\pm$ 0.09	Lamborg et al. (1999)
	Ground <sup>G</sup>	Chesapeake Biol. Lab	1997 – 1999	6 – 24-h	1.03 – 2.5	1.83 $\pm$ 0.43	Sheu and Mason (2001)
	Ship	Near Bermuda (31° 40' N, 64°10'W)	Sep, Dec 1999, Mar 2000	N/A	6.5 – 10.95 pmol m <sup>-3</sup> (1.3-2.2)	9.91 $\pm$ 2 pmol m <sup>-3</sup> (2 $\pm$ 0.4)	Mason et al. (2001)
	Ground	Mace Head (53°20'N, 9°54'W)	Sep 1995 - Dec 2001 1996 – 2013	5-min		1.72 1.663 $\pm$ 0.16	Ebinghaus et al. (2002) Weilgelt et al. (2015)
	Ship	67°N – 71°S Neumayer – Punta Arenas	Oct – Nov 1996 Dec 1999 – Jan 2000 Feb 2001	30-min 15-min 5-min	0.76 – 1.84 0.27 – 2.34	1.26 $\pm$ 01 1.08 $\pm$ 0.29 1.1 $\pm$ 0.2	Temme et al. (2003a) Temme et al. (2003b)
	Ground <sup>G</sup>	Pompano Beach, FL	Jun 2000	5-min	<1.8	1.6 $\pm$ 0.06	Malcom et al. (2003)
	Ship <sup>G</sup>	54°N – 85°N 78°N – 85°N	16 Jun – 29 Aug 2004	5-min	1.15 – 1.95 1.17 – 2.72	1.53 $\pm$ 0.12 1.82 $\pm$ 0.24	Aspmo et al. (2006)
	Ship <sup>G</sup>	Norfolk, VA to Bermuda to Barbados	15 Aug - 8 Sep 2003	5-min	1.55 – 1.9	1.63 $\pm$ 0.08	Laurier et al. (2007)
	Ground	Cape Point , South Africa	1996 – 2004 Mar 2007 – Dec 2007	15-min	0.49 – 6.78 <0.05 – 5.44 <sup>G</sup>	1.18 $\pm$ 0.11 ~1.52 $\pm$ 0.37 0.94 $\pm$ 0.16	Slemr et al. (2008) Brunke et al. (2010)
	Ship <sup>G</sup>	58°N – 67°N 43°N – 59°N 23°N – 45°N Sargasso Sea 39°S – 3°S South Africa	16 Aug – 1 Sep 2006 15 – 24 Apr 2007 30 Mar – 11 Apr 2007 8 – 21 Oct 2006	5-min		1.32 $\pm$ 0.16 2.26 $\pm$ 0.26 2.86 $\pm$ 0.17 1.36 $\pm$ 0.24	Soerensen et al. (2010)
	Ground	Appledore Island, ME, US	Jun 2007 – Aug 2010	5-min	< 3251 ppqv (~29)	147 $\pm$ 29 ppqv (1.31 $\pm$ 0.26)	Mao et al. (2012)
	Indian	Ship	39°S – 33°S 22°S – 17°S West Australia	22 – 29 Oct 2006 3 – 6 Nov 2006	5-min		1.11 $\pm$ 0.19 1.03 $\pm$ 0.16
Ship		Eq . – Prydz Bay, Antarctica	24 Nov – 11 Dec 2007	5-min	0.3 – 4.496	1.471 $\pm$ 0.842	Xia et al. (2010)
Ship		9° S - 21° S	Nov 2007	15 – 30-min	1.05 – 1.51	1.2	Witt et al. (2010)
Ground		Nieuw Nickerie 5°56'N, 56°59' W	2007	N/A	N/A	NH: 1.450 $\pm$ 0.268 SH: 1.324 $\pm$ 0.225	Müller et al. (2012)
	Ground <sup>G</sup>	Amsterdam Island 37°48'S, 77°34' E	Jan 2012 to Dec 2013	5-min	0.72 – 1.55	1.03 $\pm$ 0.03	Angot et al. (2014)
Pacific	Ground	San Francisco Bay	1965 -1966	9-min	1 – 2		Williston (1968)

	Airborne	60°N – 55°S, 8 – 10 km altitudes	1973	2 – 4-h	0.6 – 2.5	1.5	Seiler et al. (1980)
	Ship	15°N – 20°S, ~160°W	Oct 1980	N/A	1.0 – 2.25 4°N – 10°S: 1.5 – 2.25	N. of 10°N: 1.56 S. 10°S: 1.0	Fitzgerald et al. (1984)
	Ship	4.5°N – 4.5°S at 85°W; 155°W – 93°W, in the equ.	8 June – 3 July 1984	N/A	0.85 – 1.13	1.02±0.08	Kim & Fitzgerald (1986)
	Ground	48.3 N, 124.6 W, 480 m asl	May 2001 – May 2002	5-min	N/A	Spr: 1.54±0.16 Sum: 1.61±0.13 Fall: 1.54±0.09 Win: 1.51±0.12	Weiss-Penzias et al. (2003)
	Ship <sup>G</sup>	Osaka, Japan - Honolulu, Hawaii	1 May 2002 – 4 Jun 2002	5-min	1.6 – 4.7	2.5	Laurier et al. (2003)
	Airborne <sup>G</sup>	35°N – 55°N, 0 – 8 km alt.	Spring 2002	2.5-min	0.5 – 1.2	N/A	Radke et al. (2007)
	Ground <sup>G</sup>	26.8°N, 128.2°E, 60m amsl	23 Mar – 2 May 2004	5-min	1.5 - 5	2.04±0.38	Chand et al. (2008)
	Ship	Shanghai, China – Eq.	12 Nov – 17 Dec 2007	5-min	1.075- 3.698	1.746±0.513	Xia et al. (2010)
	Ship <sup>G</sup>	44°S – 26°S E. Australia 27°S – 7°S Coral Sea 56°S – 26°S New Zealand 58°S – 33°S Chilean Coast	23 Nov – 15 Dec 2006 16 Dec 2006 – 3 Jan 2007 3 – 14 Jan 2007 31 Jan – 8 Feb 2007	5-min		1.33±0.24 1.21±0.18 1.19±0.17 1.11±0.11	Soerensen et al. (2010)
	Ground <sup>G</sup>	Mt Bachelor, Oregon, USA, 43.981° N 121.691°W 2.7 km a.s.l.	2005 – 2011	5-min	1.2 – 1.4	N/A	Timonen et al. (2013)
	Ship	30°N – 60°N	Jul – Sep 2008	5-min	0.30 – 6.02	1.52±0.68	Kang&Xie (2011)
	Ship <sup>G</sup>	San Diego – San Francisco, CA	14 May – 8 Jun 2010	5-min	<7.21	1.41	Weiss-Penzias et al. (2013)
	Ground <sup>G</sup>	Puerto Villamil, Isabela Island (0°57' S, 90°58' W)  Puerto Baquerizo Moreno, San Cristóbal Island, 0°54'S, 89°36' W	Feb – Jun 2011  Jul – Oct 2011	5-min	0.58 – 2	1.08±0.17	Wang et al. (2014)
	Ship <sup>G</sup>	20°N to 15°S Samoa - HW	1 – 24 October 2011	5-min	N/A	14-20°N 1.32±0.05 ITCZ 5-14°N 1.27±0.10 Equ. 1°S-5°N 1.18±0.05 1-15°S 1.15±0.05	Soerensen et al. (2014)
Japan Sea	Ship	34°N – 39°N	13-15 Jul 18-20 Sep 2008	5-min	0.30 – 1.59 0.70 – 2.24	0.80±0.36 1.14±0.23	Kang&Xie (2011)
Okhotsk Sea	Ship	45°N – 56°N	15-18 Jul 13 – 18 Sep 2008	5-min	0.37 – 1.19 0.52 – 2.88	0.74±0.15 1.71±0.43	Kang&Xie (2011)
Bering Sea	Ship	58°N – 66°N	18 Jul – 1 Aug 9 – 13 Sep 2008	5-min	0.61 – 6.02 0.55 – 3.07	1.94±0.85 1.60±0.46	Kang&Xie (2011)

South China Sea	Ship	Northern SCS	10-28 Aug 2007	5-min	1.04 – 6.57	2.62	Fu et al. (2010b)
Yellow Sea	Ship	Incheon - Jeju Incheon – Qingdao Incheon - Weihai	10-14 Sep 2007 16-20 Oct 2007 28 Apr – 01 May 2008	30 s	1.56 – 2.97 1.29 – 2.36 1.36 – 1.99	2.43±0.59 1.82±0.51 2.03±0.66	Nyugen et al. (2011)
	Ship <sup>G</sup>	Western and central	9-18 Jul 2010	1 min	1.68 – 4.34	2.61±0.5	Ci et al. (2011)
	Ship <sup>G</sup>		May 2003 – Dec 2005 Winter Summer Spring Fall	15-min – hours	4 – 6 2 – 3 N/A N/A	5.7±0.2 2.8±0.2 3.3±0.9 3.8±0.3	Tseng et al. (2012)
Mediterranean Sea	Ground	Palma de Mallorca	13 Nov – 6 Dec 1998	5-min (Tekran)	2.4 – 6.19	3.67±0.87	Pirrone et al. (2003)
	Ship	Western and eastern sectors	14 Jul – 9 Aug 2000	5-min	0.4 – 11.2	1.9±1.02	Sprovieri et al. (2003)
Dead Sea	Ground <sup>G</sup>	Ein Bokek, Israel 31.20° N, 35.37°E, 423 bsl	29 Jun – 28 Jul 2009; 29 Dec 2009 – 17 Jan 2010	5-min	22 – 450 ppqv	N/A	Obrist et al. (2011); Moore et al. (2013)
Adriatic	Ship <sup>G</sup>	Gulf of Venice, Gulf of Trieste	26 Oct – 12 Nov 2004	5-min	0.8 – 3.3	1.6±0.4	Sprovieri & Pirrone (2008)
	Ship <sup>G</sup>	Rome - Messina	26 Oct - 15 Nov 2004 15 Jun – 5 Jul 2005	5-min	N/A	1.6 2.0	Sprovieri et al. (2010)
Augusta Basin	Ship <sup>G</sup>	Across the basin	Nov 2011 Jul – Jun 2012	5 s (20 km)	0.9 – 3.1 1.1 – 3.1	1.5±0.4 2.1±0.98	Bagnato et al. (2013)
Arctic	Ground <sup>G</sup>	Barrow, Alaska	1998 - 2001	5-min	<0.2 – 3.7	1.5 – 2	Lindberg et al. (2002)
	Ship <sup>G</sup>	Göteborg – Barrow Barrow -Chukchi Peninsula - Wrangel Island – Barrow Barrow -Longyearbyen	Jul – Sep 2005	5-min	1 – 5.24	1.73±0.36 Sea ice: 1.81 ± 0.41 Sea w/o ice: 1.55±0.21	Sommar et al. (2010)
	Ship	Central Arctic	Jul – Sep 2012	5-min	0.15 – 4.58	1.23±0.61	Yu et al. (2014)
	Ground <sup>G</sup>	Beaufort Sea	14 – 26 Mar 2009	5-min	0.01–1.51	0.59	Steffen et al. (2013)
Antarctic	Ground	Ross Island	1987 – 1989	N/A	0.02 – 1.85	0.55±0.28	De More et al. (1993)
	Ground	Neumayer 70°39'S, 8°15'W	Jan 2000 – Jan 2001 Jan-Feb 2000, Dec-Feb 2001 Mar – Jul 2000 Aug –Nov 2000	5-min	N/A	1.063±0.235 1.043±0.284  1.146±0.075 0.968±0.278	Ebinghaus et al. (2002)
	Ground <sup>G</sup>	Terra Nova Bay, Antarctic	Nov 2000 – Jan 2001	5-min	0.29 – 2.3	0.9±0.3	Sprovieri et al. (2002)
	Ground	Neumayer, Antarctic	Dec 2000 – Feb 2001	5 – 15-min	<0.3 – 2.34	1.08±0.29	Temme et al. (2003b)
	Ship <sup>G</sup>	65°S – 55°S 65°S – 63°S Antarctica	14 – 24 Jan 2007 25 – 28 Jan	5-min	N/A	1.30±0.16 1.55±0.38	Soerensen et al. (2010)
	Ground <sup>G</sup>	TRS, Queen Maud Land	Feb 2007 – Jun 2010	5-min	0.02 – 3.04	0.93±0.19	Pfaffhuber et al. (2012)

Table S2. Marine boundary layer GOM ( $\text{pg m}^{-3}$ , units noted otherwise). Units were converted in STP. The units for time resolution are hours (denoted as h) and minutes (denoted as min).

	Platform	Latitude	Sampling Period	Time Resolution	Range	Average $\pm\sigma$	Reference
Atlantic	Ship <sup>DDM</sup>	35°S – 54° N	1977	40-min	0.2 – 15.3 $\text{ng m}^{-3}$	N/A	Slemr et al. (1981);
	Ship <sup>DDM</sup>	35°S – 54°N	1978 - 1980	20 – 40-min	<0.02 (<2%) (Median) MMC: <0.1 (<5%)	N/A	Slemr et al. (1985)
	Ground	Chesapeake Biol. Lab	1997 – 1999	6 – 24-h	N/A	40 $\pm$ 50	Sheu&Mason (2001)
	Ground	Near Bermuda (31° 40' N, 64°10'W)	Sep & Dec 1999; Mar 2000	N/A	<0.05 – 6.86 $\text{pmol m}^{-3}$ (<11 – 1500)	1.38 $\pm$ 1.3 $\text{pmol m}^{-3}$ (300 $\pm$ 280)	Mason et al. (2001)
	Ground	Florida ~26°N	Jun 2000	1-h	<6.9	1.6 $\pm$ 1.5	Malcom et al. (2003)
	Ship	22°N – 48°N	May – Jun 2002		2.8 – 92.4	13.8 $\pm$ 13.1	Laurier et al. (2003)
	Ship	54°N – 85°N 78°N – 85°N	16 Jun – 29 Aug 2004	2-h (auto) 2 – 12-h (man)	0 – 22 0 – 21	2.4 $\pm$ 2 2.6 $\pm$ 2.3	Aspmo et al. (2006)
	Ship	Neumayer – Punta Arenas	Feb 2001	1-h	1 – 30	N/A	Temme et al. (2003b)
	Ship	Norfolk, VA to Bermuda, to Barbados	15 Aug - 8 Sep 2003	2-h	<3(LOD) - 27	5.9	Laurier et al. (2007)
	Ground	Appledore Island 42.97° N, 70.62° W	May 2007 – Oct 2007 May 2007 – Oct 2010	90-min	0 – 8.8 ppqv (0 – 79) 0 – 11.6 ppqv (0 – 104)	0.76 $\pm$ 0.88 ppqv (7 $\pm$ 8) 0.4 $\pm$ 0.8 ppqv (4 $\pm$ 7)	Sigler et al. (2009) Mao & Talbot (2012)
	Ship	58°N – 67°N 43°N – 59°N 23°N – 45°N Sargasso Sea 39°S – 3°S South Africa	16 Aug – 1 Sep 2006 15 – 24 Apr 2007 30 Mar – 11 Apr 2007 8 – 21 Oct 2006	40-min	N/A	0.4 $\pm$ 3 0.1 $\pm$ 1 1.2 $\pm$ 2 3.4 $\pm$ 424	Soerensen et al. (2010)
Indian	Ship	39°S – 33°S 22°S – 17°S W. Australia	22 – 29 Oct 2006 3 – 6 Nov 2006	40-min	N/A	4.6 $\pm$ 5 5.0 $\pm$ 6	Soerensen et al. (2010)
	Ground	Amsterdam Island 37°48'S, 77°34' E	Jan 2012 to Dec 2013	4-h	LOD - 4.07	0.34	Angot et al. (2014)
Pacific	Ship	Osaka, Japan to Honolulu, Hawaii	1 May 2002 – 4 Jun 2002	2-h	0.15 – 9.24	9.5	Laurier et al. (2003)
	Ground	48.3 N, 124.6 W, 480 m asl	May 2001 – May 2002	4-h	Fall 2001 – winter 2002: <LOD (1.6) Spring 2002: 1.6	N/A	Weiss-Penzias et al. (2003)
	Ground	26.8°N, 128.2°E, 60 m amsl	23 Mar – 2 May 2004	3-h	<20	4.5 $\pm$ 5.4	Chand et al. (2008)
	Ship	44°S – 26°S E. Australia 27°S – 7°S Coral Sea 56°S – 26°S New Zealand 58°S – 33°S Chilean Coast	23 Nov – 15 Dec 2006 16 Dec 2006 – 3 Jan 2007 3 – 14 Jan 2007 31 Jan – 8 Feb 2007	40-min	N/A	1.9 $\pm$ 3 0.3 $\pm$ 1 0.1 $\pm$ 0 28.6 $\pm$ 30	Soerensen et al. (2010)
	Ground	Mt. Bachelor Obs.,	2005 – 2011	3-h	200 – 700	N/A	Timonen et al. (2013)

		Oregon, US 43.981°N 121.691°W 2.7 km a.s.l.					
	Ground	Puerto Villamil, Isabela Island Puerto Baquerizo Moreno, San Cristóbal Island	23 Feb– 29 Mar 2011 10–19 Jun 2011 10–26 Oct 2011	3-h	<0.42 (LOD) – 16	1.1±1.2 <0.42 (LOD) 3.8±3.4	Wang et al. (2014)
Mediterranean	Ground	Palma de Mallorca, Fusaldo, Porto Palo, Anatalya, Neve Yam	13 Nov – 6 Dec 1998 15 Feb – 1 Mar 1999 3 May – 17 May 1999 19 Jul – 2 Aug 1999	2-h	N/A	31.5±39.2 40.4±43 52.3±43.9 32.3±17.8	Pirrone et al. (2003)
	Ship	30 – 40°N Western and eastern sectors	14 Jul – 9 Aug 2000	2-h	0.2 – 30	8±8	Sprovieri et al. (2003)
Adriatic	Ship	Rome - Messina	26 Oct – 12 Nov 2004	2-h	0.1 – 62.8	6.7±11.7	Sprovieri & Pirrone (2008)
	Ship	Rome - Messina	26 Oct - 15 Nov 2004 15 Jun – 5 Jul 2005	2-h	N/A	1.6 2.0	Sprovieri et al. (2010)
Dead Sea	Ground	Ein Bokek, Israel 31.20° N, 35.37°E	29 Jun – 28 Jul 2009 29 Dec 2009 – 17 Jan 2010	1-h	22 – 450 ppqv (196 – 4018)	N/A	Obrist et al. (2011); Moore et al. (2013)
Arctic	Ground	Barrow, Alaska, US	1998 – 2001	2-h	0 – 950 (at surface)	5 m agl: 70 100 m agl: 20 100 m agl: 2	Lindberg et al. (2002)
	Ship	Göteborg – Barrow Barrow -Chukchi Peninsula - Wrangel Island – Barrow Barrow – Longyearbyen	Jul – Sep 2005	5-h	0.7 – 10	3.2±1.7	Sommar et al. (2010)
	Ground	Beaufort Sea	14 – 26 Mar 2009	1-h	3.5–105.4	30.1	Steffen et al. (2013)
Antarctic	Ground	Ross Island	1987 – 1989	N/A	0.00 – 0.63 DMM	0.04±0.08	De More et al. (1993)
	Ground	Terra Nova Bay	Nov 2000 – Jan 2001	2-h	10.5 – 334	116.2±77.8	Sprovieri et al. (2002)
	Ground	Neumayer	Dec 2000 – Feb 2001	1-h	5 – 300	N/A	Temme et al. (2003b)
	Ship	65°S – 55°S 65°S – 63°S	14 – 24 Jan 2007 25 – 28 Jan 2007	40-min	N/A	0±0 43±39	Soerensen et al. (2010)

Table S3. Marine boundary layer PBM ( $\text{pg m}^{-3}$ , noted otherwise). Units were converted in STP. The superscript T stands for total particulate mercury.

	Platform	Latitude	Sampling Period	Time Resolution	Range	Average ( $\pm\sigma$ )	Reference
Atlantic	Ship	54°N,10°E–20°N, 67°W	Oct – Nov 1973	2 – 4-h	< 300	N/A	Seiler et al. (1980)
	Ship	15°S 29° N	1980	N/A	N/A	0.013 $\pm$ 0.018	Slemr et al. (1985)
	Ship	1°N – 33°N	1977 - 1980	N/A	N/A	0.007 $\pm$ 0.004	Slemr et al. (1985)
	Ship	4.5°N – 4.5°S at 85°W; 155°W – 93°W in the equator	16 May – 20 June 1996	24-h	5-25 $\text{fmol m}^{-3}$ (1 – 5.5)	12 $\pm$ 1 $\text{fmol m}^{-3}$ (2.6 $\pm$ 0.2)	Lamborg et al. (1999)
	Ground	Chesapeake Biol. Lab	1997 – 1999	6 – 24-h	N/A	0.02 $\pm$ 0.05	Sheu&Mason (2001)
	Ship	Near Bermuda (31° 40' N, 64°10'W)	Sep & Dec 1999; Mar 2000	N/A	N/A	<0.01 $\text{pmol m}^{-3}$	Mason et al. (2001)
	Ground	Florida ~26°N		4-h	<11	6.3 $\pm$ 4.4	Malcom et al. (2003)
	Ship	78°N - 85°N	16 Jun – 29 Aug 2004	2 – 12-h	0.2 – 6.3	2.4 $\pm$ 1.1	Aspmo et al. (2006)
	Ground	Appledore Island 42.97° N, 70.62° W	Apr 2009 – Aug 2010	90-min	0 – 39 ppqv (0 – 348)	0.34 $\pm$ 0.78 ppqv (3 $\pm$ 7)	Mao & Talbot (2012)
Indian	Ground	Amsterdam Island 37°48'S, 77°34' E	Jan 2012 to Dec 2013	4-h	<DL - 12.67	0.67	Angot et al. (2014)
Pacific	Ground	48.3 N, 124.6 W, 480 m asl	May 2001 – May 2002	24-h	N/A	Spring 2002: 0.5	Weiss-Penzias et al. (2003)
	Ground	26.8°N, 128.2°E, 60 m amsl	23 Mar – 2 May 2004	3-h	<17	3.0 $\pm$ 2.5	Chand et al. (2008)
	Ground	Mt. Bachelor Obs., Oregon, US 43.981°N 121.691°W 2.7 km a.s.l.	2005 – 2011	3-h	MBL events <12		Timonen et al. (2013)
	Ground	Puerto Villamil, Isabela Island 0°57'S,90°58' W Puerto Baquerizo Moreno, San Cristóbal Island,0°54'S,89°36'W	23 Feb– 29 Mar 2011	3-h	N/A	0.6 $\pm$ 0.7	Wang et al. (2014)
10–19 Jun 2011			<LOD				
10–26 Oct 2011			1.1 $\pm$ 1.1				
Adriatic	Ship	Gulf of Venice, Gulf of Trieste	26 Oct – 12 Nov 2004	2-h	0.04 – 51	4.5 $\pm$ 8	Sprovieri & Pirrone (2008)
Baltic Sea		Gdynia 54°31'N, 18° 48'E	18 Dec 2007 – 15 Dec 2008	24-h	2 – 142	20 $\pm$ 18 24 $\pm$ 22(cold) 15 $\pm$ 7(warm)	Beldowska et al. (2012)
Arctic	Ship	Göteborg – Barrow Barrow -Chukchi Peninsula - Wrangel Island – Barrow Barrow – Longyearbyen	Jul – Sep 2005	5-h	0.2 – 3.2	1 $\pm$ 0.7	Sommar et al. (2010)
	Ground	Beaufort Sea	14 – 26 Mar 2009	1-h	47.1 – 900.1	393.5	Steffen et al. (2013)
Antarctic	Ground <sup>T</sup>	Neumayer 70°39'S, 8°15'W	Dec 2000 – Feb 2001	48-h	15 – 120	~38	Temme et al. (2003)

Table S4: Continental boundary layer TGM or GEM (ng m<sup>-3</sup>) at remote, rural, urban, and high elevation sites

Location (elevation in m.a.s.l.)	Sampling period	Time resolution	Range	Mean ( $\pm\sigma$ )	Reference
<b>Remote</b>					
Experimental Lakes Area, Ontario, Canada, 49°39'50"N, 93°43'16"W (369m)	May 2005 - Dec 2006	1-h	0.75-3.82	1.56±0.22	Cheng et al. (2012)
Salmon Falls Creek Reservoir, Idaho, USA (1510m)	2 weeks per season during summer 2005-fall 2006; all of summer 2006	5-min		1.3-1.9	Abbott et al. (2008)
Devil's Lake State Park, Wisconsin, USA, 43°25'48"N, 89°40'48"W (389m)	Apr 2003 - Apr 2004	5-min	0.1-9.1	1.6±0.3	Manolopoulos et al. (2007)
Devil's Lake State Park, Wisconsin, USA, 43°26'5"N, 89°40' 48"W	Apr 2003 - Mar 2004	5-min		1.61±0.01	Rutter et al. (2008)
Mt. Horeb, Wisconsin, USA, 42°57'36"N, 89°42'W (331m)	June & Sep 2004	5-min		1.34±0.26	Manolopoulos et al. (2007)
Contiguous USA	Jan 2009 - Dec 2011	1-h	0.2-11.3	1.31±0.29	Gay et al. (2013)
<b>Rural</b>					
Multiple sites across Canada, 42°10'-82°30'N, 135°37'-62°19'W	1994-2011	5-min		1.21-1.93	Cole et al. (2014)
Maryhill, Ontario, Canada, 43°31'N, 80°25'W	Oct 2004 - Nov 2004	5-min		1.8±0.2	Cobbett and Van Heyst (2007)
Central Alberta, Canada	2004-2007	5-min	0.7-9.5	1.5-1.6	Mazur et al. (2009)
Fort McMurray, Alberta, Canada, 56°45'N, 111°28'48"W (362m)	Oct 2010 - May 2013	5-min	0.64-4.43	1.45±0.18	Parsons et al. (2013)
Huntington Forest, New York, USA, 43°58'12"N, 74°13'12"W (500m)	Jun 2006 - May 2007	1-h	0.5-2.5	1.4±0.4	Choi et al. (2008)
Huntington Forest, New York, USA, 43°58'12"N, 74°13'12"W (510m)	Dec 2007 - Nov 2009	2-h	0.2-3.7	1.3±0.4	Choi et al. (2013)
Huntington Forest, New York, USA, 43°58'12"N, 74°13'12"W (500m)	Jan 2009 - Dec 2010	2 h	0.2-3.4		Cheng et al. (2013)
Atlanta, Georgia, USA	2005-2008	5-min		1.35	Nair et al. (2012)
Wisconsin, USA, 43°26'24"N, 89°40'48"W (389m)	2003-2009	5-min	0.45-6.48	1.49-1.82	Engle et al. (2010)
North Dakota, USA, 48°38'60"N, 102°24'W (692m)	2003-2009	5-min	0.99-5.13	1.61±0.22	Engle et al. (2010)
Virginia, USA, 38°31'12"N, 78°26'24"W (1074m)	2003-2009	5-min	<dl-3.63	1.27±0.31	Engle et al. (2010)
Dexter, Michigan, USA, 42°25'12"N, 83°54'W	2004	1-h	0.26-14.82	1.6±0.6	Liu et al. (2010)
Holland, Illinois, USA, 42°46'4.8"N, 86°8'56.4"W	1 Jul 2007 - 8 Nov 2007	1-h	≤6.0	1.3±0.3	Gratz et al. (2013)
San Francisco Bay Area, California, USA 37°11'24"N, 121°46'12"W (149m)	9 Sep 2005-27 Sep 2005, 21 Oct 2008-30 Oct 2008	5-min	0.84-11.7	1.85-2.37	Rothenberg et al. (2010)



Central Wisconsin, USA	Jun 2007 - Nov 2007	5-min		1.3-1.4	Kolker et al. (2010)
Athens, Ohio, USA 39°18'N, 82°7'W (330m)	Jul 2004 - Jul 2005	1-h	0.78-4.38	1.62±0.24	Yatavelli et al. (2006)
Mexico, 21°8'N, 98°25'W	3 days in Oct 2002	5-min	1.13-2.93	1.32±0.33	De la Rosa et al. 2004
13 UK sites	2004-2013	1-4 weeks		1.67±0.07	Brown et al. (2015)
Lichwin, Poland, 49°52.957'N, 20°57.024'E	19 Aug 2003 - 29 Aug 2003	23-h		1.63±0.35	Zielonka et al. (2005)
Lichwin, Poland, 49°52.957'N, 20°57.024'E	6 Jan 2004 - 3 Feb 2004	23-h	≤7.02	4.15±1.33	Zielonka et al. (2005)
Chongqing, China	Aug 2006 - Sep 2007	30-min		6.14±1.3	Yang et al. (2009)
Chongqing, China	Aug 2006 - Sep 2007	30-min		4.32±1.04	Yang et al. (2009)
Wanqingsha, Pearl River Delta, China, 22°42'N, 113°33'E	Nov 2008	5-min	0.5-18.67	2.94±2.02	Li et al. (2011)
Beijing, China, 40°28'51"N, 116°46'30"E (220m)	Dec 2008 - Nov 2009	5-min	0.39-14.83	3.22±1.74	Zhang et al. (2013)
Chuncheon, Korea, 37°59'24"N, 127°48'36"E	2006-2008	24-h	0.26-10.75	2.12±1.47	Han et al. (2014)
Dinghu, Pearl River Delta, China, 23°9'51"N, 112°32'57"E	Oct 2009 - Apr 2010	5-min	1.87-29.9	5.07±2.89	Chen et al. (2013)
<b>Urban</b>					
Halifax, Nova Scotia, Canada, 44°39'54.71"N, 63°34'9.69"W (125m)	Jan 2010 - Dec 2011	1-h		1.7±1.0	Cheng et al. (2014)
Windsor, Ontario, Canada, 42°18.27'N, 83°3.98'W	2007	1-h	0.83-40.9	2.0±1.6	Xu and Akhtar (2010)
Toronto, Ontario, Canada, 43°40'N, 79°24'W	Dec 2003 - Nov 2004	5-min	0.5-44.1	4.5±3.1	Song et al. (2009)
Windsor, Ontario, Canada, 42°18.27'N, 83°3.98'W	2007-2011	1-h	0.3-57	2.0±1.3	Xu et al. (2014)
Birmingham, Alabama, USA	2005-2008	5-min		2.12	Nair et al. (2012)
Houston, Texas, USA, 29°42'36"N, 95°20'24"W (75m)	Aug 2011 - Oct 2012	2.5-min	0.72-43.54	1.6±0.6	Lan et al. (2014)
Reno, Nevada, USA, 39°34'N, 119°48'W (1509m)	2002-2005	2.5-min	0.9-8.6	2.3	Stamenkovich et al. (2007)
Reno, Nevada, USA, 39°34'12"N, 119°47'W (1509m)	Nov 2004 - Nov 2007	2 h	0.5-6.4	1.6±0.5	Peterson et al. (2009)
Desert Research Institute, Nevada, USA, 39°34'12"N, 119°47'W (1497m)	Jun 2007 - Aug 2007	2-h	0.6-2.5	1.2±0.3	Weiss-Penzias et al. (2009)
NV98, Nevada, USA, 39°30'36"N, 119°43'12"W (1340m)	Jun 2007 - Aug 2007	2-h	0.9-4.6	1.7±0.6	Weiss-Penzias et al. (2009)
Illinois, USA, 38°36'36"N, 90°9'36"W (206m)	2003-2009	5-min	0.05-324	2.09-2.54	Engle et al. (2010)
Wisconsin, USA, 43°4'48"N, 87°52'48"W (184m)	2003-2009	5-min	1.29-24.7	2.26-2.94	Engle et al. (2010)
Detroit, Michigan, USA, 42°10'48"N, 83°3'36"W	2004	1-h	0.36-25.6	2.5±1.4	Liu et al. (2010)

San Francisco Bay Area, California, USA, 37°25'12"N, 122°2'24"W (11m)	30 Aug 2009 - 9 Sep 2005, 19 Sep 2008 - 30 Sep 2008	5-min	0.53-8.19	1.74-1.76	Rothenberg et al. (2010)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W (137m)	Dec 2007 – Apr 2008; Dec 2008 – Apr 2009	5-min		1.6	Wang et al. (2013)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W (137m)	Dec 2007 - Apr 2008 coal power plant in operation	5-min		1.8	Wang et al. (2013)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W (137m)	Dec 2007 - Apr 2009 coal power plant shutdown	5-min		1.5	Wang et al. (2013)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W	Sep 2008 - Jun 2013	2-h	1.17-1.61	1.40±0.26	Civerolo et al. (2014)
New York City, USA, 40°52'05"N, 73°52'42"W	Sep 2008 - Jun 2013	2-h	1.20-1.92	1.52±0.36	Civerolo et al. (2014)
Milwaukee, Wisconsin, USA, 43°6'29"N, 87°53'2"W	Jun 2004 - May 2005	5-min		2.48±0.02	Rutter et al. (2008)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W (136m)	Dec 2007 - Nov 2009	2-h	0.9-6.4	1.6±0.4	Choi et al. (2013)
Chicago, Illinois, USA, 41°47'27.6"N, 87°36'7.2"W	1 Jul 2007 - 8 Nov 2007	1-h	≤16.5	2.5±1.5	Gratz et al. (2013)
Oxford, Mississippi, USA, 34°21'50"N, 89°32'60"W (150m)	Jul 2011 - Jun 2012	5-min	0.8-7.9	1.54±0.32	Jiang et al. (2013)
Contiguous U.S.	Jan 2009 - Dec 2011	1-h	0.4-237.7	1.65±1.36	Gay et al. (2013)
Mexico City, 19°29'21.4"N, 99°8'51.8"W	Mar 2006	5-min		7.2±4.8	Rutter et al. (2009)
Mexico, 22°44'N, 102°28'W	3 days in Sep 2002	5-min	0.26-702.32	71.82±82.03	De la Rosa et al. (2004)
Mexico City, 19°21'N; 99°4'W	3 days in Oct 2002	5-min	2.8-34.21	9.81±3.97	De la Rosa et al. (2004)
São Paulo State, Brazil	2002-2003	2-h		7.0±5.8	Fostier and Michelazzo, (2006)
Oxford, UK	Feb 2008 - Apr 2008	5-min	1.32-23.2	3.17±1.59	Witt et al. (2010)
UK sites	2004-2013	1-4 weeks		2.07±0.03	Brown et al. (2015)
UK sites	2004-2013	1-4 weeks		2.01±0.12	Brown et al. (2015)
UK sites	2003-2013	1-4 weeks		3.74±0.7	Brown et al. (2015)
Göteborg, Sweden, 57°42'N, 11°54'E	Feb 2005	1-h	1.35-6.42	1.96±0.38	Li et al. (2008)
Seoul, Korea, 37°30'50.4"N, 127°0'3.6"E	Feb 2005 - Dec 2006	5-min		3.44±2.13	Choi et al. (2009)
Seoul, Korea, 37°30'50.4"N, 127°0'3.6"E	2006-2008	24-h	0.19-149.84	3.72±2.96	Han et al. (2014)
Guro-gu, Nowon-gu, Songpa-gu, and Yongsan-gu, Seoul, Korea, 27°31'-37°39'N, 127°7'-126°53'W	2010-2011	1-h		3.28-3.47	Kim et al. (2013)
Fukuoka, Japan, 33°32'55"N, 130°21'52"E	Jun 2012 - May 2013	5-min	1.47-6.39	2.33±0.49	Marumoto et al. (2015)
Chongqing, China	Aug 2006 - Sep 2007	30-min		7.07±1.04	Yang et al. (2009)

Guangzhou, Pearl River Delta, China, 23°7'26.8"N, 113°21'17.4"E	Nov 2010 - Oct 2011	5-min	2.66-11.1	4.60±1.36	Chen et al. (2013)
Nanjing, China, 32°2'N, 118°46'48"E	Jan 2011 - Dec 2011	5-min	0.8-180	7.9±7.0	Zhu et al. (2012)
<b>High elevation</b>					
Pad Monadnock, New Hampshire, USA, 42°51'36"N, 71°52'48"W (700m)	Jun 2005 - May 2007	5-min	1.08-1.69	1.26-1.54	Mao et al. (2008)
Pad Monadnock, New Hampshire, USA, 42°51'36"N, 71°52'48"W (700m)	2007	5-min	0.88-2.58	1.40±0.19	Sigler et al. (2009)
Look Rock, Tennessee, 35°36'36"N, 83°54'36"W (813m)	Spring-Summer 2004	1-h		1.65	Valente et al. (2007)
Storm Peak Laboratory, Colorado, USA (3200m)	Oct 2006 - May 2007	1-h	1.06-2.15	1.51±0.12	Obrist et al. (2008)
Storm Peak Laboratory, Colorado, USA (3220m)	Apr 2008 - Jul 2008	1-h	1.2-5.0	1.6±0.3	Fain et al. (2009)
Mt. Bachelor Observatory, Oregon, USA, 43°58'48"N, 121°41'24"W (2700m)	May 2005 - Aug 2005	3-h	1.36-1.83	1.54±0.18	Swartzendruber et al. (2006)
Northern Nevada, USA, 41°30' 3.6"N, 117°30'3.6"W and 41°33'3.6"N, 115°12'39.6"W (1388-1806m)	29 Mar 2005 - 4 Apr 2005, 26 Sep 2005 - 8 Aug 2005, 22 Oct 2005 - 28 Oct 2005, 24 Jan 2006 - 30 Jan 2006	5-min	≤106	2.5-3.0	Lyman and Gustin (2008)
Northern Nevada, USA, 41°30'N, 117°30'W (1388m)	Jun 2007 - Aug 2007	2-h	0.6-18.1	1.8±1.4	Weiss-Penzias et al. (2009)
Contiguous U.S. high elevation sites and Mt. Lulin, Taiwan	Jan 2009 - Dec 2011	1-h	0.4-8.3	1.45±0.51	Gay et al. (2013)
Mt. Gongga, China, 29°20'-30°20'N, 101°30'-102°15'E (1640m)	May 2005 - Jun 2006	5-15 min	0.52-21.03	3.98±1.62	Fu et al. (2008)
Mt. Gongga, China, 29°20'-30°20'N, 101°30'-102°15'E (1640m)	Jun 2006 - Jun 2007	5-min	0.9-43.7	3.91±1.2	Fu et al. (2009)
Mt. Leigong, China, 23°23'24"N, 108°12'E (2178m)	May 2008 - May 2009	10-min	0.41-23.9	2.80±1.51	Fu et al. (2010a)
Waliguan, China, 36°17'12"N, 100°53'52.7"E, (3816m)	Sep 2007 - Sep 2008	10-min		1.98±0.98	Fu et al. (2012a)
Mt. Changbai, China, 42°24'N, 128°28'E (738.1m)	Aug 2005 - Jul 2006	1-h	1.28-9.49	3.58±1.78	Wan et al. (2009a)
Mt. Changbai, China, 42°24'0.9"N, 128°6'45"E (741m)	Oct 2008 - Oct 2010	5-min	<dl-9.5	1.60±0.51	Fu et al. (2012b)
Mt. Lulin, Taiwan, 23°30'36"N, 120°55'12"E (2862m)	Apr 2006 - Sep 2007	5-min	0.24-5.75	1.73±0.61	Sheu et al. (2010)
Shangri-La, Tibetan Plateau, China, 28°1'N, 99°44'E (3580m)	Nov 2009 - Nov 2010	5-min	1.03-13.59	2.55±0.73	Zhang et al. (2015)
5 high-elevation sites	Apr 2006 - May 2009	5-min		1.65±0.32	Weiss-Penzias et al. (2015)
Mt. Front Lulin, Taiwan, 23°30'36"N, 120°55'12"E (2862m)	Mar, Jul, Sep, Dec 2008	5-min		1.4-2.2	Weiss-Penzias et al. (2015)
Mt. Bachelor Observatory, Oregon, USA, 43°58'48"N, 121°41'24"W (2763m)	2006-2009	5-min		1.44-1.6	Weiss-Penzias et al. (2015)
Reno, Nevada, USA, 39°34'12"N, 119°47'W (1497m)	Jan 2005 - Aug 2007	5-min		1.36-1.73	Weiss-Penzias et al. (2015)

Paradise Valley, Nevada, USA, 41°30'N, 117°30'W (1388m)	Jun 2007 - Aug 2007	5-min		1.79±1.12	Weiss-Penzias et al. (2015)
Storm Peak Laboratory, Colorado, USA, 40°27'36"N, 106°44'24"W (3200m)	May 2008 - Jun 2008	5-min		1.59±0.21	Weiss-Penzias et al. (2015)

Table S5: Continental boundary layer GOM ( $\mu\text{g m}^{-3}$ ) at remote, rural, urban, and high elevation sites. LOD indicates limit of detection.

Location (elevation in m.a.s.l.)	Sampling period	Time resolution	Range	Mean ( $\pm\sigma$ )	Reference
<b>Remote</b>					
Experimental Lakes Area, Ontario, Canada, 49°39'50"N, 93°43'16"W (369m)	May 2005 - Dec 2006	3-h	<LOD-24.89	0.99 $\pm$ 1.9	Cheng et al. (2012)
Salmon Falls Creek Reservoir, Idaho, USA (1510m)	2 weeks per season during summer 2005-fall 2006; all of summer 2006	1-h		3.2-8.1	Abbott et al. (2008)
Devil's Lake State Park, Wisconsin, USA, 43°25'48"N, 89°40'48"W (389m)	Apr 2003 - Apr 2004	1-h	<LOD-151	3.8 $\pm$ 8.9	Manolopoulos et al. (2007)
Devil's Lake State Park, Wisconsin, USA, 43°26'5"N, 89°40'48"W	Apr 2003 - Mar 2004	1-h		5.3 $\pm$ 0.2	Rutter et al. (2008)
Contiguous U.S.	Jan 2009 - Dec 2011	2-h	0-223.5	2.55 $\pm$ 5.60	Gay et al. (2013)
<b>Rural</b>					
Multiple sites in Canada, 43°32'-82°30'N, 114°12' - 62°19'W	2002-2011	1-3 h		0.5-100.9	Cole et al. (2014)
Huntington Forest, New York, USA, 43°58'12"N, 74°13'12"W (500m)	Jun 2006 - May 2007	1-h	<LOD-45.4	1.8 $\pm$ 2.2	Choi et al. (2008)
Huntington Forest, New York, USA, 43°58'12"N, 74°13'12"W (510m)	Dec 2007 - Nov 2009	2-h	<LOD-63.9	1.3 $\pm$ 3.3	Choi et al. (2013)
Huntington Forest, New York, USA, 43°58'12"N, 74°13'12"W (500m)	Jan 2009 - Dec 2010	2-h	0-44		Cheng et al. (2013)
Maryhill, Ontario, Canada, 43°31'N, 80°25'W	Oct 2004 - Nov 2004	2-h		2.3 $\pm$ 3.0	Cobbett and Van Heyst (2007)
Atlanta, Georgia, USA	2005-2008	1-h		8.6	Nair et al. (2012)
Wisconsin, USA, 43°26'24"N, 89°40'48"W (389m)	2003-2009	1-h	<LOD-151	2.8-11.2	Engle et al. (2010)
North Dakota, USA, 48°38'60"N, 102°24'W (692m)	2003-2009	1-h	<LOD-19.2	2.0 $\pm$ 2.6	Engle et al. (2010)
Virginia, USA 38°31'12"N, 78°26'24"W (1074m)	2003-2009	1-h	<LOD-37.7	1.8 $\pm$ 3.9	Engle et al. (2010)
Dexter, Michigan, USA, 42°25'12"N, 83°54'W	2004	1-h	1.0-121.7	3.8 $\pm$ 6.6	Liu et al. (2010)
San Francisco Bay Area, California, USA, 37°11'24"N, 121°46'12"W (149m)	9 Sep 2005-27 Sep 2005, 21 Oct 2008-30 Oct 2008	1-h	0.2-154	4.58-14.5	Rothenberg et al. (2010)
Central Wisconsin, USA	Jun 2007 - Nov 2007	1-h		0.6-0.8	Kolker et al. (2010)
Athens, Ohio, USA, 39°18'N, 82°7'W (330m)	Jul 2004 - Jul 2005	1-h	0-461.59	12.45 $\pm$ 24.53	Yatavelli et al. (2006)
Holland, Illinois, USA 42°46'4.8"N, 86°8'56.4"W	1 Jul 2007 - 8 Nov 2007	1-h	$\leq$ 137	8 $\pm$ 15	Gratz et al. (2013)
Beijing, China, 40°28'51"N, 116°46'30"E (220m)	Dec 2008 - Nov 2009	1-h	0.12-	10.1 $\pm$ 18.8	Zhang et al. (2013)

			301.20		
Chuncheon, Korea, 37°59'24"N, 127°48'36"E	2006-2008	24-h	0.1-16.9	2.7±2.7	Han et al. (2014)
<b>Urban</b>					
Halifax, Nova Scotia, Canada, 44°39'54.71"N, 63°34'9.69"W (125m)	Jan 2010 - Dec 2011	2-h		2.1±3.4	Cheng et al. (2014)
Toronto, Ontario, Canada, 43°40'N, 79°24'W	Dec 2003 - Nov 2004	1-h	<LOD-284	14.2±13.2	Song et al. (2009)
Birmingham, Alabama, USA	2005-2008	1-h		78.2	Nair et al. (2012)
Reno, Nevada, USA, 39°34'12"N, 119°47'W (1509m)	Nov 2004 - Nov 2007	2-h	0-401	26±35	Peterson et al. (2009)
Illinois, USA, 38°36'36"N, 90°9'36"W (206m)	2003-2009	1-h	<LOD-8160	11.6-63.3	Engle et al. (2010)
Wisconsin, USA, 43°4'48"N, 87°52'48"W (184m)	2003-2009	1-h	<LOD-124	4.1-13.8	Engle et al. (2010)
Detroit, Michigan, USA, 42°10'48"N, 83°3'36"W	2004	1-h	1.0-2472.9	15.5±54.9	Liu et al. (2010)
San Francisco Bay Area, California, USA, 37°25'12"N, 122°2'24"W (11m)	30 Aug 2009 - 9 Sep 2005, 19 Sep 2008 - 30 Sep 2008	1-h	0.2-19.5	1.81-2.58	Rothenberg et al. (2010)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W (137m)	Dec 2007 – Apr 2008; Dec 2008 – Apr 2009	2-h		5.1	Wang et al. (2013)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W (137m)	Dec 2007 - Apr 2008 coal power plant in operation	2-h		9.9	Wang et al. (2013)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W (137m)	Dec 2007 - Apr 2009 coal power plant shutdown	2-h		3.9	Wang et al. (2013)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W (136m)	Dec 2007 - Nov 2009	2-h	<LOD-177.6	5.6±10.3	Choi et al. (2013)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W	Sep 2008 - Jun 2013	2-h	0.29-23.25	8.24±12.94	Civerolo et al. (2014)
New York City, USA, 40°52'05"N, 73°52'42"W	Sep 2008 - Jun 2013	2-h	0.78-13.53	6.26±15.85	Civerolo et al. (2014)
Milwaukee, Wisconsin, USA, 43°6'29"N, 87°53'2"W	Jun 2004 - May 2005	1-h		10.3±0.2	Rutter et al. (2008)
NV98, Nevada, USA, 39°30'36"N, 119°43'12"W (1340m)	Jun 2007 - Aug 2007	2-h	<LOD-157	46±32	Weiss-Penzias et al. (2009)
DRI, Nevada, USA, 39°34'12"N, 119°47'W (1497m)	Jun 2007 - Aug 2007	2-h	5.7-401	87±57	Weiss-Penzias et al. (2009)
Chicago, Illinois, USA, 41°47'27.6"N, 87°36'7.2"W	1 Jul 2007 - 8 Nov 2007	1-h	≤2707	17±87	Gratz et al. (2013)
Oxford, Mississippi, USA, 34°21'50"N, 89°32'60"W (150m)	Jul 2011 - Jun 2012	1-h	0-84	3.87±6.17	Jiang et al. (2013)
Contiguous U.S.	Jan 2009 - Dec 2011	2-h	0-7694.8	9.69±59.64	Gay et al. (2013)
Mexico City, 19°29'21.4"N, 99°8'51.8"W	Mar 2006	1-h		62±64	Rutter et al. (2009)

Göteborg, Sweden, 57°42'N, 11°54'E	Feb 2005	1-h	0.34-32.84	2.53±4.09	Li et al. (2008)
Seoul, Korea, 37°30'50.4"N, 127°0'3.6"E	2006-2008	24 h	0.9-57.3	11.3±9.5	Han et al. (2014)
Fukuoka, Japan, 33°32'55"N, 130°21'52"E	Jun 2012 - May 2013	2-h	<LOD-123	5.7±9.4	Marumoto et al. (2015)
<b>High elevation</b>					
Pad Monadnock, New Hampshire, USA, 42°51'36"N, 71°52'48"W (700m)	2007	2-h	0-33	1.2±2.7	Sigler et al. (2009)
Pad Monadnock, New Hampshire, USA, 42°51'36"N, 71°52'48"W (700m)	Jun 2005 – Oct 2008	90-min	<LOD-5.1	0.18-1.4	Mao and Talbot (2012)
Look Rock, Tennessee, USA, 35°36'36"N, 83°54'36"W (813m)	Spring-Summer 2004	1-h		5	Valente et al. (2007)
Northern Nevada, USA, 41°30' 3.6"N, 117°30'3.6"W and 41°33'3.6"N, 115°12'39.6"W (1388-1806m)	29 Mar 2005 - 4 Apr 2005, 26 Sep 2005 - 8 Aug 2005, 22 Oct 2005 - 28 Oct 2005, 24 Jan 2006 - 30 Jan2006	2-h	≤150	7.0-13.0	Lyman and Gustin (2008)
NV02, Nevada, USA, 41°30'N, 117°30'W (1388m)	Jun 2007 - Aug 2007	2-h	<LOD-143	26±26	Weiss-Penzias et al. (2009)
Storm Peak Laboratory, Colorado, USA (3220m)	Apr 2008 - Jul 2008	1-h	0-137	20±21	Faïn et al. (2009)
Mt. Bachelor Observatory, Oregon, USA, 43°58'48"N, 121°41'24"W (2700m)	May 2005 - Aug 2005	3-h	<LOD-141	39±75	Swartzendruber et al. (2006)
Contiguous U.S. high elevation sites and Mt. Lulin, Taiwan	Jan 2009 - Dec 2011	2-h	0.2-555	143.25±105.91	Gay et al. (2013)
Waliguan, China, 36°17'12"N, 100°53'52.7"E, (3816m)	Sep 2007 - Sep 2008	3.5-7.5 h		7.4±4.8	Fu et al. (2012a)
Mt. Changbai, China, 42°24'N, 128°28'E (738.1m)	Aug 2005 - Jul 2006	1-h	0-880	29-84	Wan et al. (2009b)
Mt. Lulin, Taiwan, 23°30'36"N, 120°55'12"E (2862m)	Apr 2006 - Sep 2007	1-2 h	≤194.7	12.1±20	Sheu et al. (2010)
Shangri-La, Tibetan Plateau, China, 28°1'N, 99°44'E (3580m)	Nov 2009 - Nov 2010	1-h	0.71-39.85	8.22±7.90	Zhang et al. (2015)
5 high-elevation sites	Apr 2006 - May 2009	2-h		30±24 (GOM+PBM)	Weiss-Penzias et al. (2015)
Mt. Front Lulin, Taiwan, 23°30'36"N, 120°55'12"E (2862m)	Mar, Jul, Sep, Dec 2008	2-h		11-27 (GOM+PBM)	Weiss-Penzias et al. (2015)
Mt. Bachelor Observatory, Oregon, USA, 43°58'48"N,121°41'24"W (2763m)	2006-2009	2-h		32-44 (GOM+PBM)	Weiss-Penzias et al. (2015)
Reno, Nevada, USA, 39°34'12"N, 119°47'W (1497m)	Jan 2005 - Aug 2007	2-h		14-68 (GOM+PBM)	Weiss-Penzias et al. (2015)
Paradise Valley, Nevada, USA 41°30'N, 117°30'W (1388m)	Jun 2007 - Aug 2007	2-h		32±16 (GOM+PBM)	Weiss-Penzias et al. (2015)
Storm Peak Laboratory, Colorado, USA, 40°27'36"N, 106°44'24"W (3200m)	May 2008 - Jun 2008	2-h		27-34 (GOM+PBM)	Weiss-Penzias et al. (2015)

Table S6: Continental boundary layer PBM ( $\text{pg m}^{-3}$ ) at remote, rural, urban, and high elevation sites. LOD indicates limit of detection; TPM indicates total particulate mercury; FPBM indicates fine particulate mercury; CPBM indicates coarse particulate mercury.

Location (elevation in m.a.s.l.)	Sampling period	Time resolution	Range	Mean ( $\pm\sigma$ )	Reference
<b>Remote</b>					
Experimental Lakes Area, Ontario, Canada, 49°39'50"N, 93°43'16"W (369m)	May 2005 - Dec 2006	3-h	<LOD-42.33	4.4 $\pm$ 3.7	Cheng et al. (2012)
Devil's Lake State Park, Wisconsin, USA, 43°25'48"N, 89°40'48"W (389m)	Apr 2003 - Apr 2004	1-h	<LOD-205	8.6 $\pm$ 8.3	Manolopoulos et al. (2007)
Devil's Lake State Park, Wisconsin, USA, 43°26'5"N, 89°40'48"W	Apr 2003 - Mar 2004	1-h		9.1 $\pm$ 0.1	Rutter et al. (2008)
Contiguous U.S.	Jan 2009 - Dec 2011	2-h	0-403.6	5.12 $\pm$ 7.43	Gay et al. (2013)
<b>Rural</b>					
Multiple sites in Canada, 43°32'-82°30'N, 114°12' - 62°19'W	2002-2011	1-3 h		2.3-168.5	Cole et al. (2014)
Maryhill, Ontario, Canada 43°31'N, 80°25'W	Oct 2004 - Nov 2004	2-h		3.0 $\pm$ 6.2	Cobbett and Van Heyst (2007)
Atlanta, Georgia, USA	2005-2008	1-h		4.4	Nair et al. (2012)
Wisconsin, USA, 43°26'24"N, 89°40'48"W (389m)	2003-2009	1-h	<LOD-205	6.4-12.2	Engle et al. (2010)
North Dakota, USA, 48°38'60"N, 102°24'W (692m)	2003-2009	1-h	<LOD-11.9	2.2 $\pm$ 2.4	Engle et al. (2010)
Virginia, USA 38°31'12"N, 78°26'24"W (1074m)	2003-2009	1-h	<LOD-19.4	4.6 $\pm$ 3.9	Engle et al. (2010)
Dexter, Michigan, USA, 42°25'12"N, 83°54'W	2004	1-h	1.0-90.6	6.1 $\pm$ 5.5	Liu et al. (2010)
Huntington Forest, New York, USA, 43°58'12"N, 74°13'12"W (500m)	Jun 2006 - May 2007	1-h	<LOD-54.0	3.2 $\pm$ 3.7	Choi et al. (2008)
Huntington Forest, New York, USA, 43°58'12"N, 74°13'12"W (510m)	Dec 2007 - Nov 2009	2-h	<LOD-97.6	4.1 $\pm$ 7.8	Choi et al. (2013)
Huntington Forest, New York, USA, 43°58'12"N, 74°13'12"W (500m)	Jan 2009 - Dec 2010	2-h	0-39		Cheng et al. (2013)
San Francisco Bay Area, California, USA, 37°11'24"N, 121°46'12"W (149m)	9 Sep 2005-27 Sep 2005, 21 Oct 2008-30 Oct 2008	1-h	0.2-37.5	3.68-7.99	Rothenberg et al. (2010)
Central Wisconsin, USA	Jun 2007 - Nov 2007	1-h		2.6-5.0	Kolker et al. (2010)
Athens, Ohio, USA, 39°18'N, 82°7'W (330m)	Jul 2004 - Jul 2005	1-h	0-76.82	5.29 $\pm$ 6.04	Yatavelli et al. (2006)
Holland, Illinois, USA 42°46'4.8"N, 86°8'56.4"W	1 Jul 2007 - 8 Nov 2007	1-h	$\leq$ 31	6 $\pm$ 4	Gratz et al. (2013)
Lichwin, Poland, 49°52.957'N, 20°57.024'E	19 Aug 2003 - 29 Aug 2003	23-h	0.02-0.2 ng m <sup>-3</sup> TPM	0.11 $\pm$ 0.05 ng m <sup>-3</sup> TPM	Zielonka et al. (2005)



Lichwin, Poland, 49°52.957'N, 20°57.024'E	6 Jan 2004 - 3 Feb 2004	23-h		1.05±0.18	Zielonka et al. (2005)
Jeziory, Poland	Apr 2013 - Oct 2014	24-72 h	FPBM: <LOD- 16.1, CPBM: <LOD- 142.5	FPBM: 2.4±2.8, CPBM: 20.8±21.6	Siudek et al. (2016)
Beijing, China, 40°28'51"N, 116°46'30"E (220m)	Dec 2008 - Nov 2009	1-h	0.49- 1090.24	98.2±112.7	Zhang et al. (2013)
Chuncheon, Korea, 37°59'24"N, 127°48'36"E	2006-2008; Fall 2009, winter 2009, summer 2010 for TPM	24-48 h	0.1-30	3.7±5.7, TPM: 0.9- 8.5	Kim et al. (2012); Han et al. (2014)
<b>Urban</b>					
Halifax, Nova Scotia, Canada, 44°39'54.71"N, 63°34'9.69"W (125m)	Jan 2010 - Dec 2011	2-h		2.3±3.1	Cheng et al. (2014)
Toronto, Ontario, Canada, 43°40'N, 79°24'W	Dec 2003 - Nov 2004	1-h	<LOD- 252	21.5±16.4	Song et al. (2009)
Birmingham, Alabama, USA	2005-2008	1-h		39.5	Nair et al. (2012)
Reno, Nevada, USA, 39°34'12"N, 119°47'W (1509m)	Nov 2004 - Nov 2007	2-h	0-180	9±10	Peterson et al. (2009)
Illinois, USA, 38°36'36"N, 90°9'36"W (206m)	2003-2009	1-h	<LOD- 11600	12.9-37.9	Engle et al. (2010)
Wisconsin, USA, 43°4'48"N, 87°52'48"W (184m)	2003-2009	1-h	<LOD- 507	10.2-15.7	Engle et al. (2010)
Detroit, Michigan, USA, 42°10'48"N, 83°3'36"W	2004	1-h	1.0-1345.2	18.1±61	Liu et al. (2010)
San Francisco Bay Area, California, USA, 37°25'12"N, 122°2'24"W (11m)	30 Aug 2009 - 9 Sep 2005, 19 Sep 2008 - 30 Sep 2008	1-h	0.2-29.4	3.06-3.17	Rothenberg et al. (2010)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W (137m)	Dec 2007 - Apr 2008; Dec 2008 - Apr 2009	2-h		8.9	Wang et al. (2013)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W (137m)	Dec 2007 - Apr 2008 coal power plant in operation	2-h		21.8	Wang et al. (2013)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W (137m)	Dec 2007 - Apr 2009 coal power plant shutdown	2-h		7.3	Wang et al. (2013)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W (136m)	Dec 2007 - Nov 2009	2-h	<LOD- 271.2	8.7±12.8	Choi et al. (2013)
Rochester, New York, USA, 43°8'46"N, 77°32'54"W	Sep 2008 - Jun 2013	2-h	2.36-27.32	13.48±21.01	Civerolo et al. (2014)
New York City, USA, 40°52'05"N, 73°52'42"W	Sep 2008 - Jun 2013	2-h	1.42-15.82	7.96±12.13	Civerolo et al. (2014)
Milwaukee, Wisconsin, USA, 43°6'29"N, 87°53'2"W	Jun 2004 - May 2005	1-h		11.8±0.3	Rutter et al. (2008)
NV98, Nevada, USA, 39°30'36"N, 119°43'12"W (1340m)	Jun 2007 - Aug 2007	2-h	<LOD-55	5±4	Weiss-Penzias et al. (2009)
DRI, Nevada, USA, 39°34'12"N, 119°47'W (1497m)	Jun 2007 - Aug 2007	2-h	<LOD-	10±10	Weiss-Penzias et al. (2009)

			180		
Chicago, Illinois, USA, 41°47'27.6"N, 87°36'7.2"W	1 Jul 2007 - 8 Nov 2007	1-h	≤401	9±20	Gratz et al. (2013)
Oxford, Mississippi, USA, 34°21'50"N, 89°32'60"W (150m)	Jul 2011 - Jun 2012	1-h	0-33	4.58±3.40	Jiang et al. (2013)
Contiguous U.S.	Jan 2009 - Dec 2011	2-h	0-3687.2	8.46±29.05	Gay et al. (2013)
Mexico City, 19°29'21.4"N, 99°8'51.8"W	Mar 2006	1-h		187±300	Rutter et al. (2009)
São Paulo State, Brazil	2002-2003	24-h		TPM: 400±300	Fostier and Michelazzo, (2006)
Göteborg, Sweden, 57°42'N, 11°54'E	Feb 2005	1-h	3.89-20.26	12.5±5.88	Li et al. (2008)
Poznań, Poland, 52°42'N, 16°88'E	Apr 2013 - Oct 2014	24-72 h	FPBM: <LOD- 77.1, CPBM: <LOD- 604.9	FPBM: 7.3±9.1, CPBM: 22.6±45.3	Siudek et al. (2016)
Shanghai, China	Jul 2004 - Apr 2006	24-h	0.07-1.45 ng m <sup>-3</sup> TPM	0.56±0.22 ng m <sup>-3</sup> TPM	Xiu et al. (2009)
Shanghai, China	Jul 2004 - Apr 2006	24-h	0.2-0.47 ng m <sup>-3</sup> TPM	0.33±0.09 ng m <sup>-3</sup> TPM	Xiu et al. (2009)
Beijing, China	Jan 2006 - Dec 2006	12-h		263±246 (day), 280±383 (night)	Schleicher et al. (2015)
Seoul, Korea, 37°30'50.4"N, 127°0'3.6"E	2006-2008; winter, summer 2010 for TPM	24-48 h	2.1-64.3	13.4±12.0, TPM: 1.1- 18.9	Kim et al. (2012); Han et al. (2014)
Fukuoka, Japan, 33°32'55"N, 130°21'52"E	Jun 2012 - May 2013	2-h	<LOD- 139	10±11	Marumoto et al. (2015)
<b>High elevation</b>					
Look Rock, Tennessee, USA, 35°36'36"N, 83°54'36"W (813m)	Spring-Summer 2004	1-h		7	Valente et al. (2007)
Northern Nevada, USA, 41°30'3.6"N, 117°30'3.6"W and 41°33'3.6"N, 115°12'39.6"W (1388-1806m)	29 Mar 2005 - 4 Apr 2005, 26 Sep 2005 - 8 Aug 2005, 22 Oct 2005 - 28 Oct 2005, 24 Jan 2006 - 30 Jan 2006	2-h	≤65	9.0-13.0	Lyman and Gustin (2008)
NV02, Nevada, USA, 41°30'N, 117°30'W (1388m)	Jun 2007 - Aug 2007	2-h	<LOD- 102	6±9	Weiss-Penzias et al. (2009)
Storm Peak Laboratory, Colorado, USA (3220m)	Apr 2008 - Jul 2008	1-h	0-33	9±6	Faïn et al. (2009)

Mt. Bachelor Observatory, Oregon, USA, 43°58'48"N, 121°41'24"W (2700m)	May 2005 - Aug 2005	3-h	<LOD-11	4.4±4.4	Swartzendruber et al. (2006)
Contiguous U.S. high elevation sites and Mt. Lulin, Taiwan	Jan 2009 - Dec 2011	2-h	0-555.6	90.69±92.07	Gay et al. (2013)
Waliguan, China, 36°17'12"N, 100°53'52.7"E, (3816m)	Sep 2007 - Sep 2008	3.5-7.5 h		19.4±18.1	Fu et al. (2012a)
Mt. Changbai, China, 42°24'N, 128°28'E (738.1m)	Aug 2005 - Jul 2006	1-h	0-1001	15-167	Wan et al. (2009b)
Mt. Lulin, Taiwan, 23°30'36"N, 120°55'12"E (2862m)	Apr 2006 - Sep 2007	1-2 h	38.4	2.3±3.9	Sheu et al. (2010)
Shangri-La, Tibetan Plateau, China, 28°1'N, 99°44'E (3580m)	Nov 2009 - Nov 2010	1-h	3.02-185.59	38.32±31.26	Zhang et al. (2015)

Table S7. Airborne measurements of TGM/GEM ( $\text{ng m}^{-3}$ ) and GOM/PBM ( $\text{pg m}^{-3}$ ), units stated otherwise.

Region	Flight domain	Time Period	Altitude	Hg species	Range	Reference
Atlantic	Cologne, via Greenland, Canada, San Francisco, to Putna Arena, Chile	1977	8 km	TGM	SH: $1.08 \pm 0.36$ NH: $1.45 \pm 0.22$ ITCZ: 2.4 – 2.7	Seiler et al. (1980)
Central Europe	--	Jun & Aug 1977 – 1979; Feb 1980	6 – 8 km	TGM	$2.24 \pm 0.51$	Slemr et al. (1985)
Germany	400 km btw Munich and Halle	13 Jun 1996	0.9 km 2.5 km	TGM	$1.774 \pm 0.101$ $1.635 \pm 0.094$	Ebinghaus & Slemr (2000)
Barrow, AK	Northeast of Point Barrow	Mar – Apr 2000	100 m 1000 m	GOM	20 2	Lindberg et al. (2002)
Canada	Southeastern Southern & central ON Arctic	Summer 1995, 97 Winter 1997-98 Spring 1998	0.1 – 7 km	GEM	1.5 1.7 1.7 (>1 km); 1.2 (<1 km)	Banic et al. (2003)
East Asia	24°N-44°N, 124°E-146°E	April 2001	SL – 7 km	TGM PBM	Mean: 1.68 (2.5 – 2 km) – 2.13 (0 – 0.5 km) 14 – 917	Friedli et al. (2004)
South America	Btw São Paulo and Santiago de Chile, 25°S – 50°N Btw Frankfurt and São Paulo	31 Aug, 5 Oct 2005 4 Oct 2005	SL – 11 km	GEM	NH: 1.70 – 2.21 SH: 1.35 – ~1.7	Ebinghaus et al. (2007)
North Pacific Coastal CA&WA	35°N – 55°N	Spring 2002	0 – 8 km	GEM	Median: 1.2 – 0.5	Radke et al. (2007)
Pacific Northwest	46°N – 49°N, 125°W – 121°W	12 Apr – 15 May 2006	Surface – 6 km	GEM	$1.43 \pm 0.166$ ; 0.994–2.02	Swatzendruber et al. (2008)
West Pacific	Honolulu, HW; Anchorage, AL; Mexico City	4 Mar – 12 May 2006	Surface – 12 km	GEM	0 – ~250 ppqv	Talbot et al. (2007, 2008)
North America & Europe	40°N – 44°N, 75°W – 100°W	5 Nov 2011	~ 6 km	GEM GOM	>0.3 - ~1.3 0 – 600	Lyman & Jaffe (2012)
Tullahoma, Tennessee, USA	--	Aug 2012 – Jun 2013	0 – 6 km	GEM GOM PBM	$1.38 \pm 0.17$ ; 0.75 – 2.05 $34.8 \pm 28.9$ ; 0 – 20 (win); <125 (sum) $29.6 \pm 29.5$ ; 4.4 – 194.9	Brooks et al. (2014)
Southeastern US	Flights over 32°N – 36°N, 122°W – 77°W	1 Jun - 15 Jul 2013	--	THg GOM	$1.49 \pm 0.16$ $212 \pm 112$ ; up to 300 – 680	Gratz et al. (2015) Shah et al. (2016)

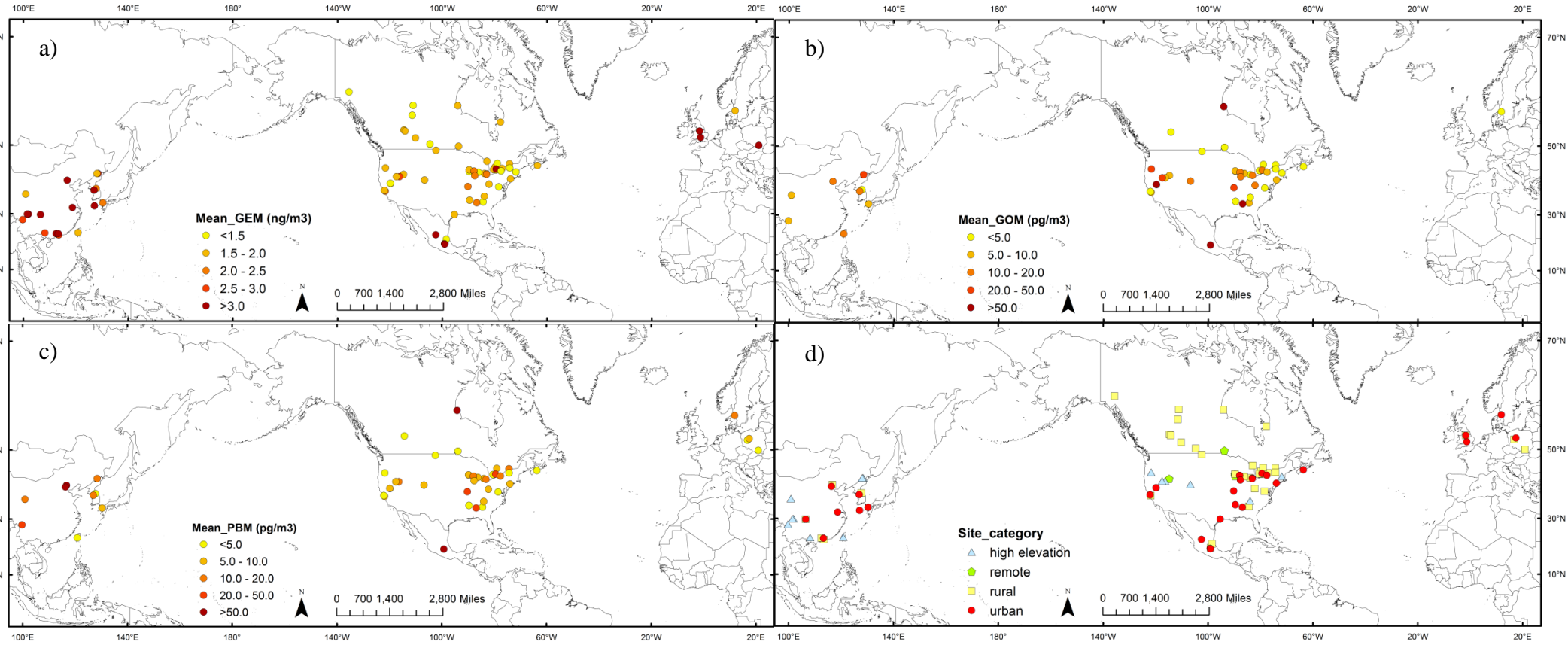


Figure S1. Maps of mean TGM/GEM (a), GOM (b) and PBM (c) concentrations at the continental sites (d) as listed in Tables S4-S6.

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