Table S1: TGM and ancillary data

Measurement	Unit	Data frequency	Network
TGM	ng m^{-3}	hourly	ECCC CAPMoN[1]
SO$_2$, inorganic ions (Na$^+$, K$^+$, Cl$^-$, Ca^{2+}, Mg^{2+}, SO$_4^{2-}$, NH$_4^+$)	µg m^{-3}	24-h	ECCC CAPMoN[2]
CO	ppm	hourly	ECCC National Air Pollution Surveillance (NAPS) program[3], ECCC Canadian Greenhouse Gas Measurement program[4], USEPA Air Quality System (AQS)[5]
Total carbon	µg m^{-3}	1-in-3 day	Interagency Monitoring of Protected Visual Environments (IMPROVE)[6]
Air temperature	°C	hourly	ECCC Historical Climate Data[7]

[1] https://doi.org/10.18164/e1df5764-1eec-4a9f-9c03-f515b396b717

[2] https://doi.org/10.18164/e73c7f47-df9c-4877-923c-20e09db28176

[3] https://data-donnees.az.ec.gc.ca/data/air/monitor/national-air-pollution-surveillance-naps-program/

[4] https://gaw.kishou.go.jp/

[5] https://www.epa.gov/aqs

[6] https://vista.cira.colostate.edu/Improve/improve-data/

[7] https://climate.weather.gc.ca/

Table S2: Descriptive statistics of 24-h average TGM concentrations (ng m^{-3}) at Egbert (EGB), Kejimkujik (KEJ) and Saturna (SAT). P denotes percentile.

Site	Year	N	Mean	Median	StdDev	P5	P25	P75	P95	Min	Max
EGB	2005	344	1.60	1.57	0.21	1.33	1.45	1.66	2.05	1.19	2.53
EGB	2006	335	1.64	1.63	0.18	1.36	1.52	1.75	1.95	1.25	2.30
EGB	2007	301	1.54	1.55	0.20	1.19	1.41	1.66	1.85	1.08	2.36
EGB	2008	345	1.46	1.45	0.14	1.23	1.36	1.56	1.68	1.10	1.85
EGB	2009	358	1.41	1.43	0.15	1.14	1.30	1.51	1.62	0.92	1.76
EGB	2010	340	1.39	1.39	0.19	1.09	1.25	1.53	1.69	0.97	1.88
EGB	2011	104	1.27	1.25	0.16	1.06	1.17	1.37	1.57	0.97	1.84
EGB	2012	272	1.19	1.16	0.15	1.00	1.11	1.27	1.47	0.92	1.81
EGB	2013	305	1.09	1.06	0.16	0.87	0.97	1.17	1.35	0.80	1.68
EGB	2014	341	1.28	1.31	0.23	0.90	1.06	1.46	1.60	0.84	1.80
EGB	2015	344	1.30	1.32	0.14	1.06	1.20	1.41	1.52	0.96	1.70
EGB	2016	356	1.28	1.28	0.16	1.03	1.17	1.41	1.52	0.82	1.69
EGB	2017	386	1.36	1.37	0.16	1.09	1.23	1.49	1.58	1.00	1.77
EGB	2018	485	1.25	1.25	0.13	1.05	1.17	1.34	1.44	0.85	1.75
KEJ	2005	332	1.73	1.53	0.75	1.12	1.32	1.82	3.19	0.99	6.87
KEJ	2006	342	1.10	1.09	0.21	0.79	0.93	1.28	1.43	0.65	1.62
KEJ	2007	312	1.16	1.15	0.15	0.94	1.05	1.27	1.40	0.71	1.52
KEJ	2008	307	1.32	1.32	0.16	1.06	1.20	1.44	1.58	0.95	1.72
KEJ	2009	341	1.27	1.27	0.17	1.01	1.15	1.40	1.48	0.83	2.38
KEJ	2010	354	1.34	1.36	0.15	1.09	1.23	1.44	1.57	0.84	1.79
KEJ	2011	346	1.37	1.41	0.17	1.05	1.24	1.50	1.57	0.97	1.62
KEJ	2012	349	1.36	1.37	0.16	1.10	1.23	1.50	1.61	0.98	1.67
KEJ	2013	345	1.30	1.33	0.16	1.01	1.19	1.44	1.51	0.79	1.67
KEJ	2014	336	1.30	1.30	0.16	1.02	1.18	1.44	1.53	0.91	1.61
KEJ	2015	350	1.19	1.21	0.16	0.91	1.06	1.33	1.39	0.74	1.53
KEJ	2016	312	1.14	1.12	0.16	0.91	1.01	1.28	1.38	0.82	1.50
KEJ	2017	140	1.17	1.21	0.11	0.92	1.12	1.25	1.28	0.83	1.34
SAT	2009	274	1.36	1.35	0.21	1.06	1.20	1.52	1.73	0.94	2.01
SAT	2010	266	1.50	1.51	0.17	1.24	1.39	1.61	1.75	1.14	2.53
SAT	2011	302	1.35	1.36	0.13	1.15	1.25	1.43	1.55	0.83	1.75
SAT	2012	334	1.22	1.24	0.12	1.01	1.14	1.31	1.40	0.91	1.54
SAT	2013	287	1.33	1.33	0.12	1.13	1.25	1.43	1.51	0.90	1.61
SAT	2014	333	1.26	1.27	0.10	1.10	1.20	1.33	1.41	1.00	1.53
SAT	2015	263	1.16	1.16	0.11	0.99	1.09	1.22	1.31	0.76	1.61
SAT	2016	153	1.37	1.38	0.07	1.23	1.33	1.42	1.49	1.18	1.57
SAT	2018	314	1.38	1.38	0.11	1.19	1.32	1.44	1.54	1.08	1.97

Table S3: Long-term trends in cold and warm season TGM concentrations and source contributions from the PMF model. SAT: 2009-2015, EGB: 2006-2017, KEJ: 2005-2015. Cold season: Nov to Apr; warm season: May to Oct. Statistically significant (p<0.1) trends are shown in bold.

Site	Parameter/Source	Cold Season Trend		Significance	Warm Season Trend		Significance
		Slope (ng m^{-3} yr^{-1})	Slope (% yr^{-1})	p-value	Slope (ng m^{-3} yr^{-1})	Slope (% yr^{-1})	p-value
SAT	Obs TGM	-0.0402	-2.79	0.114	**-0.0587**	**-4.19**	0.053
	Modeled TGM	-0.0303	-2.15	0.114	**-0.0590**	**-4.23**	0.053
	Background Hg	-0.0030	-0.34	0.821	**0.0430**	**9.43**	0.030
	Shipping and SSA processing	-0.0025	-6.49	0.788	**-0.0551**	**-20.25**	0.070
	Terrestrial GEM re-emissions	-0.0067	-5.36	0.718	-0.0386	-11.40	0.287
	Local combustion	-0.0253	-24.69	0.311	-0.0244	-19.57	0.497
	Secondary sulfate	0.0191	-51.30	0.297	0.0130	-52.52	0.297
	Oceanic evasion	-0.0107	-6.91	0.902	0.0013	1.91	0.371
	Wildfires	-0.0033	-23.27	0.117	0.0042	4.99	0.942
EGB	Obs TGM	**-0.0209**	**-1.27**	0.017	**-0.0304**	**-2.07**	0.017
	Modeled TGM	**-0.0244**	**-1.46**	0.063	**-0.0281**	**-1.94**	0.017
	Background Hg	-0.0199	-1.64	0.264	**-0.0310**	**-3.64**	0.067
	Road salt	1.79E-10	0.00	0.982	**-0.0009**	**-11.48**	0.073
	Terrestrial GEM re-emissions	-0.0010	-1.78	0.701	-0.0059	-1.45	0.765
	Local combustion	0.0021	3.08	0.694	0.0027	7.56	0.267
	Secondary sulfate	**-3.99E-05**	**-12.45**	0.053	-3.97E-07	-12.45	0.285
	Wildfires	0	NA	0.798	-0.0035	-6.72	0.195
	Crustal/soil	0.0033	4.67	0.651	0.0103	14.38	0.447
KEJ	Obs TGM	-0.0232	-1.50	0.427	-0.0199	-1.49	0.294
	Modeled TGM	-0.0224	-1.45	0.434	-0.0172	-1.32	0.294
	Background Hg	-0.0134	-1.20	0.284	**-0.0553**	**-4.61**	0.047
	Regional emission and SSA processing	**-0.0183**	**-7.32**	0.083	-0.0160	-7.27	0.157
	Terrestrial GEM re-emissions	0.0008	6.45	0.573	**0.0306**	**-24.82**	0.050
	Local combustion	0.0002	3.74	0.469	0	0	0.791
	Secondary sulfate	0	0	0.696	0	NA	0.765
	Oceanic evasion	0.0036	3.44	0.331	0.0068	105.14	0.220
	Wildfires	0.0002	-692.67	0.239	**0.0029**	**-35.76**	0.057

Table S4: Long-term trends in cold and warm season relative source contributions. SAT: 2009-2015, EGB: 2006-2017, KEJ: 2005-2015. Cold season: Nov to Apr; warm season: May to Oct. Statistically significant (p<0.1) trends are shown in bold.

Site	Parameter/Source	Cold Season Trend Slope (% yr^{-1})	Significance p-value	Warm Season Trend Slope (% yr^{-1})	Significance p-value
SAT	pct Natural surface	0.54%	0.985	2.00%	0.447
	pct Anthropogenic	-0.54%	0.985	-2.00%	0.447
	pct Background Hg	1.43%	0.354	**5.52%**	0.000
	pct Shipping and SSA processing	-0.13%	0.788	**-3.75%**	0.070
	pct Terrestrial GEM re-emissions	-0.56%	0.604	-1.75%	0.494
	pct Local combustion	-1.75%	0.311	-1.80%	0.740
	pct Secondary sulfate	1.42%	0.297	1.04%	0.297
	pct Oceanic evasion	-0.66%	0.902	0.39%	0.217
	pct Wildfires	-0.20%	0.127	-0.26%	0.651
EGB	pct Natural surface	0.07%	0.711	-0.03%	0.821
	pct Anthropogenic	-0.07%	0.711	0.03%	0.821
	pct Background Hg	-0.13%	0.885	-0.68%	0.487
	pct Road salt	0.22%	0.598	**-0.06%**	0.083
	pct Terrestrial GEM re-emissions	-0.08%	0.725	0.12%	0.841
	pct Local combustion	0.15%	0.631	0.33%	0.114
	pct Secondary sulfate	**-2.00E-3%**	0.070	1.67E-5%	0.349
	pct Wildfires	0%	0.654	-0.16%	0.501
	pct Crustal/soil	0.29%	0.497	0.93%	0.304
KEJ	pct Natural surface	0.59%	0.524	1.60%	0.237
	pct Anthropogenic	-0.59%	0.524	-1.60%	0.237
	pct Background Hg	0.39%	0.304	**-3.01%**	0.010
	pct Regional emission and SSA processing	**-1.24%**	0.100	**-1.22%**	0.104
	pct Terrestrial GEM re-emissions	0.06%	0.746	**3.40%**	0.048
	pct Local combustion	0.01%	0.469	0%	0.791
	pct Secondary sulfate	0%	0.696	0%	0.765
	pct Oceanic evasion	0.24%	0.317	0.58%	0.140
	pct Wildfires	0.01%	0.361	**0.32%**	0.052
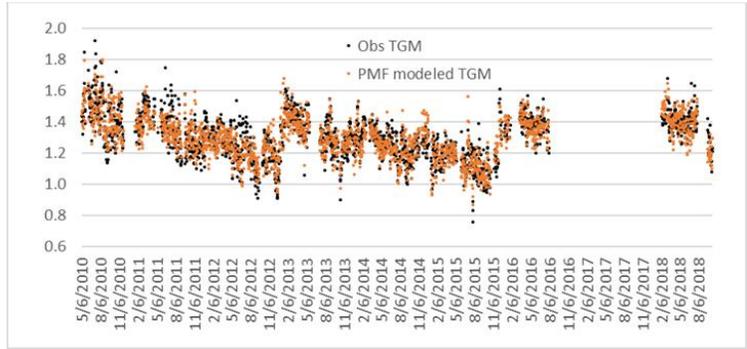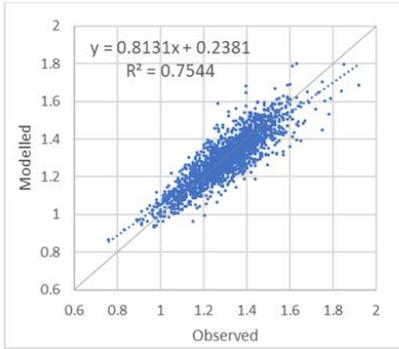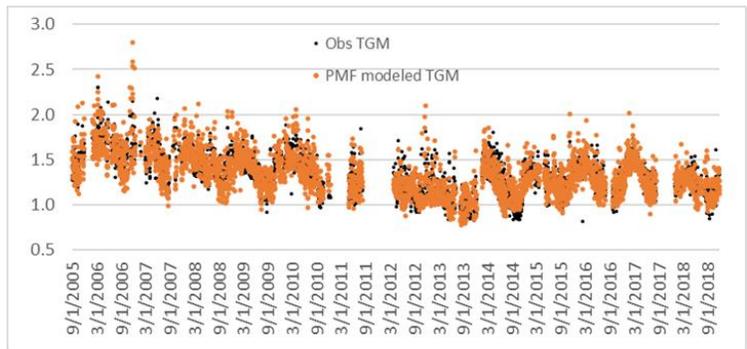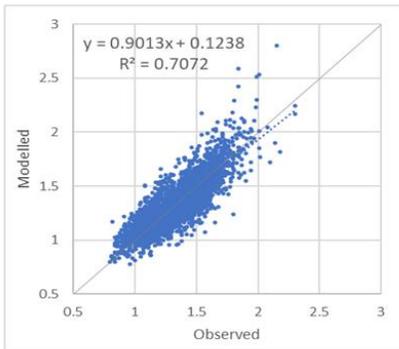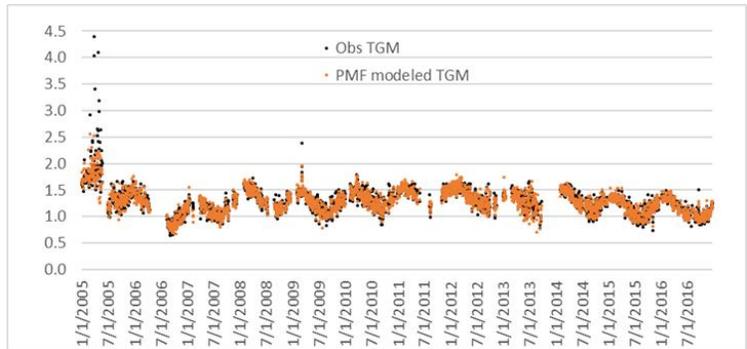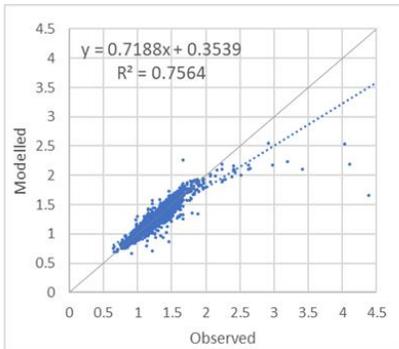
Figure S1: Comparison of PMF modelled and observed 24-h TGM using regression analysis (left) and time-series analysis (right)
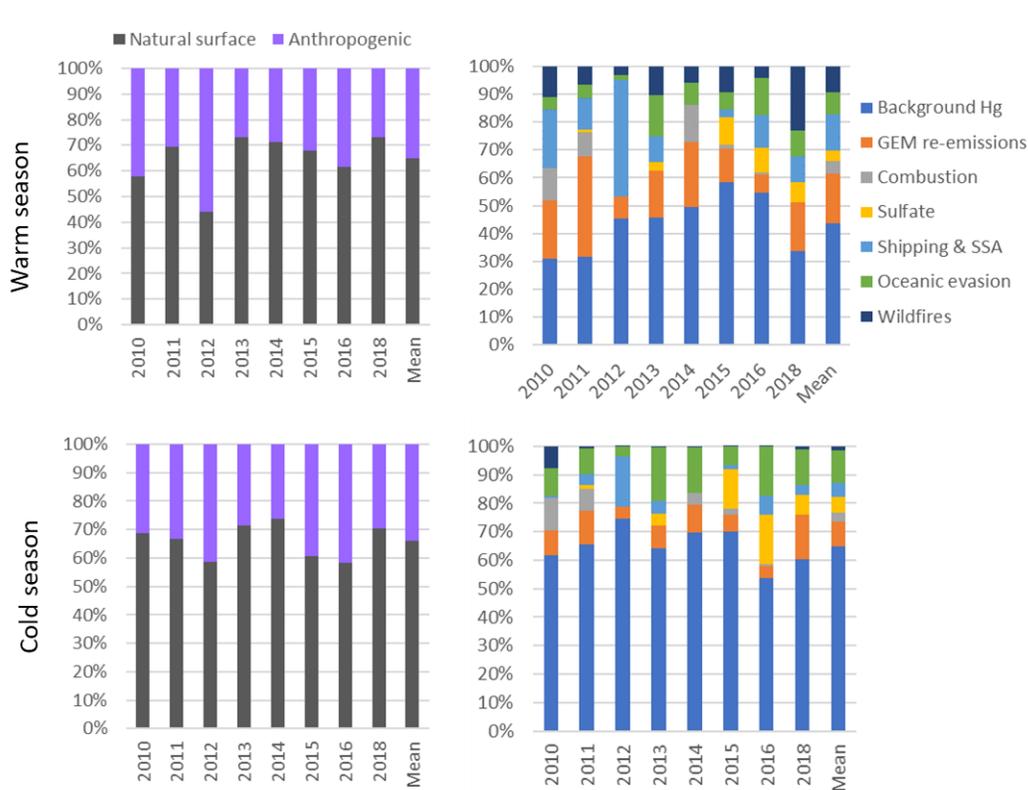
Figure S2: Relative source contributions to warm and cold season mean TGM at SAT. Mean applies to the 2010-2018 period except 2017. Warm season: May to Oct; cold season: Nov to Apr. Natural surface emissions comprise wildfires, GEM re-emissions, oceanic evasion, and natural surface emissions contributing to background Hg. Anthropogenic emissions comprise local combustion, sulfate, and anthropogenic emissions contributing to background Hg.
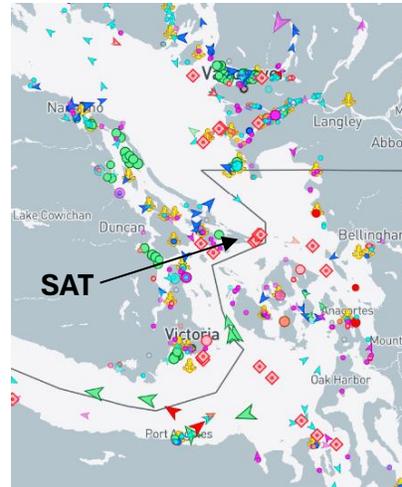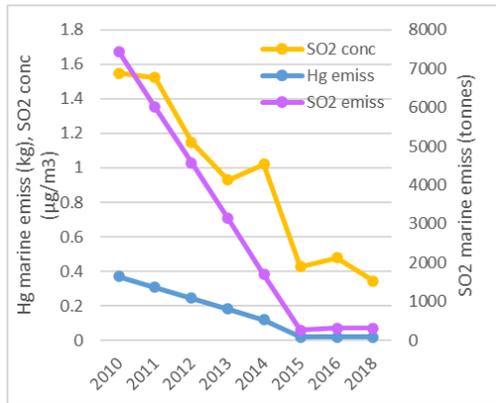
Figure S3: Left - Annual emissions of SO_2 and Hg from marine transportation reported for British Columbia (ECCC APEI, 2023). SO_2 ambient concentrations measured at SAT are also plotted. Right – Symbols represent ship traffic along the Strait of Georgia (http://www.shiptraffic.net/marine-traffic/straits/Strait_of_Georgia).
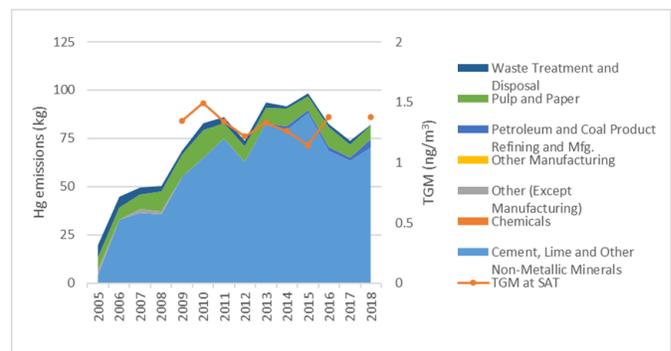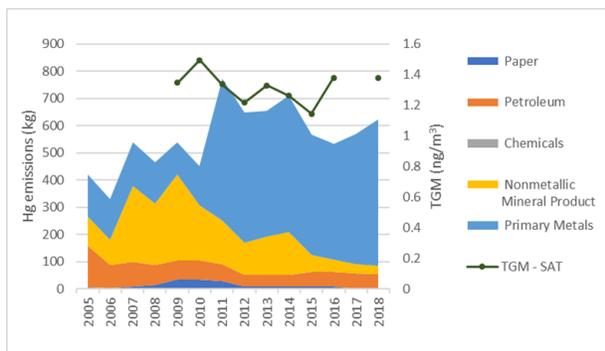
Figure S4: Hg emission sources within 150 km of SAT. Left: Washington State emissions (USEPA TRI, 2023); right: British Columbia emissions (ECCC NPRI, 2023)
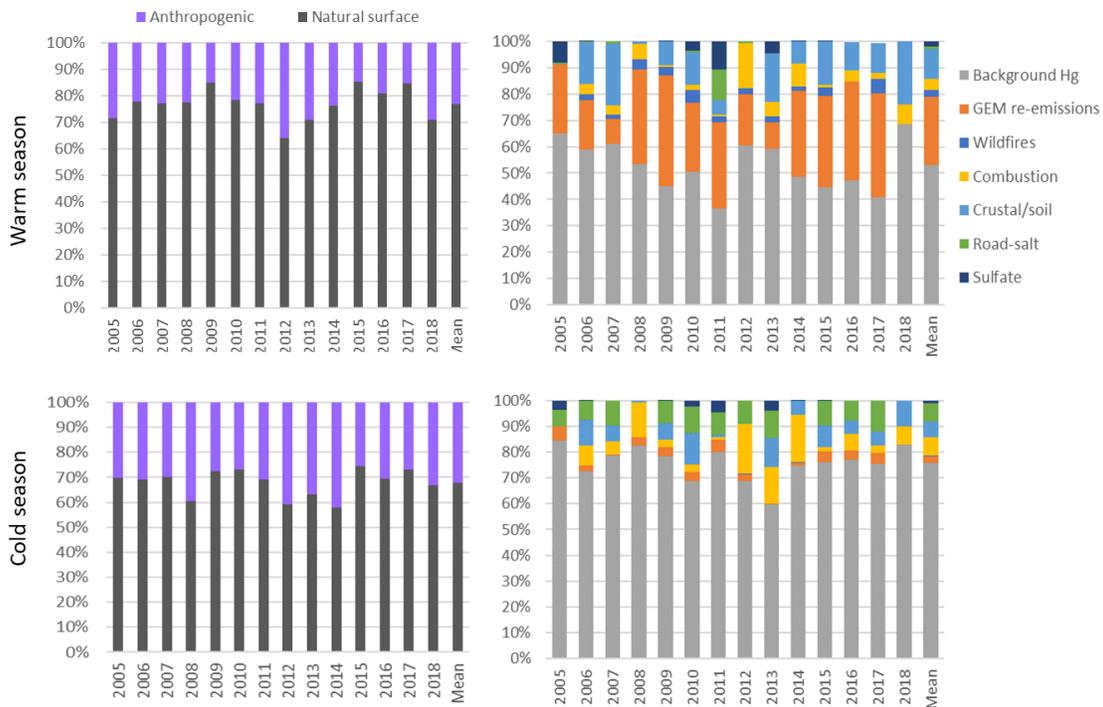
Figure S5: Relative source contributions to warm and cold season mean TGM at EGB. Mean applies to the 2005-2018 period. Warm season: May to Oct; cold season: Nov to Apr. Natural surface emissions comprise wildfires, GEM re-emissions, crustal/soil dust, road salt, and natural surface emissions contributing to background Hg. Anthropogenic emissions comprise local combustion, sulfate, and anthropogenic emissions contributing to background Hg.
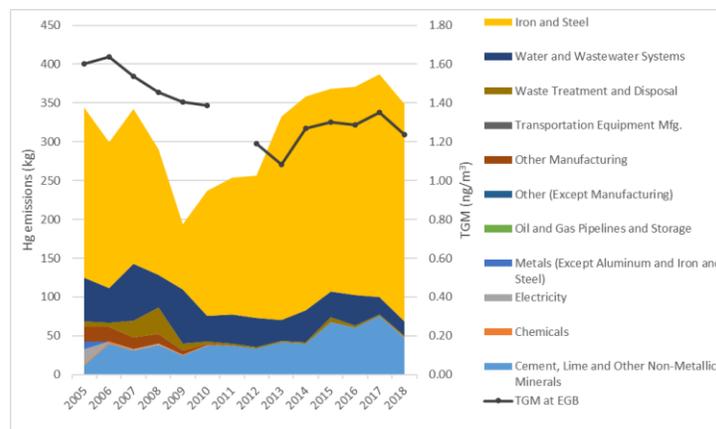
Figure S6: Hg emission sources within 150 km of EGB in Province of Ontario (ECCC NPRI, 2023)
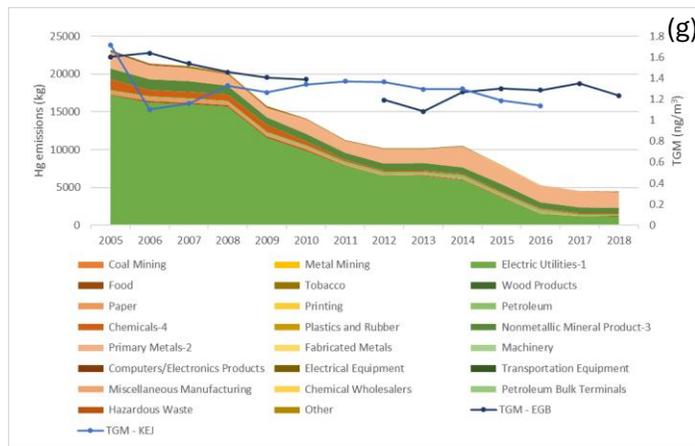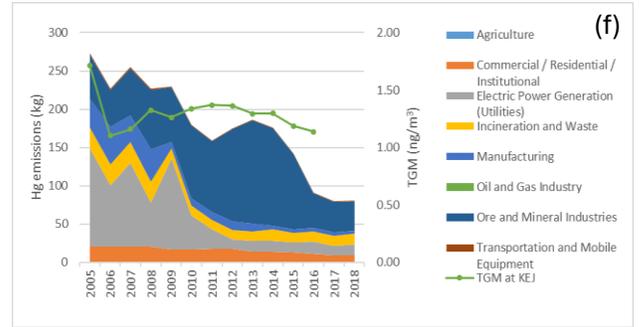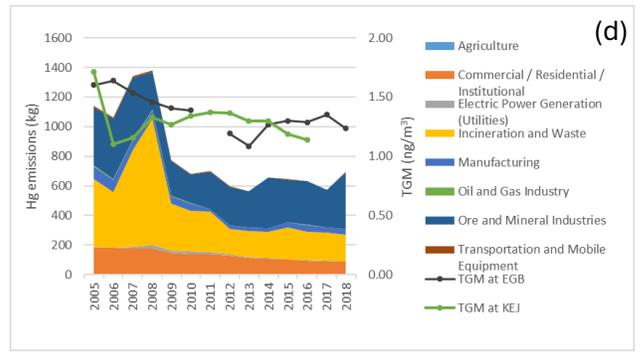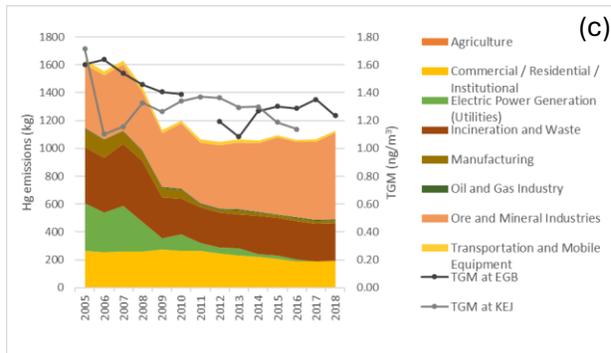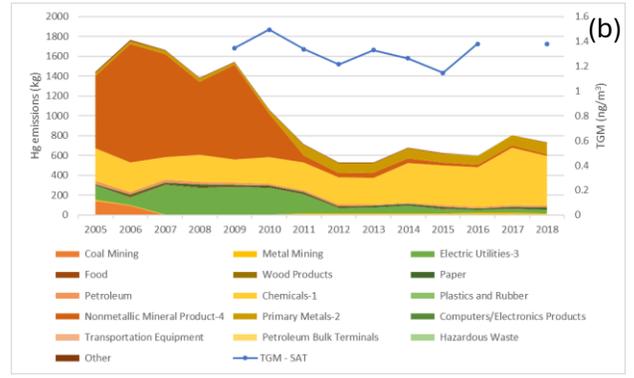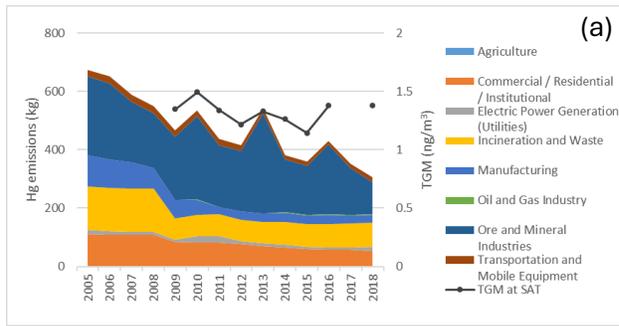
Figure S7: Regional Hg emission sources around SAT, EGB and KEJ. Emissions data for (a) British Columbia, (b) Northwestern U.S., (c) Ontario, (d) Quebec, (e) Nova Scotia, (f) New Brunswick, (g) Northeastern and midwestern U.S. Data sources: ECCC APEI (2023) for Canadian emissions, USEPA TRI (2023) for U.S. emissions.
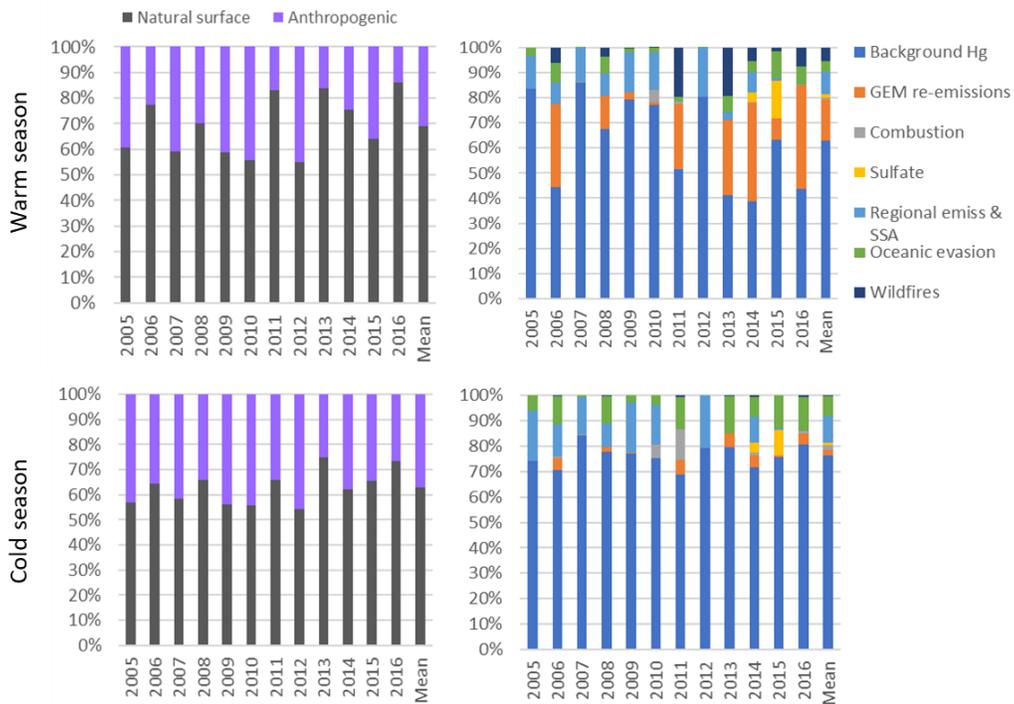
Figure S8: Relative source contributions to warm and cold season mean TGM at KEJ. Mean applies to the 2005-2016 period. Warm season: May to Oct; cold season: Nov to Apr. Natural surface emissions comprise wildfires, GEM re-emissions, oceanic evasion, and natural surface emissions contributing to background Hg. Anthropogenic emissions comprise local combustion, sulfate, regional emissions and sea-salt processing, and anthropogenic emissions contributing to background Hg.
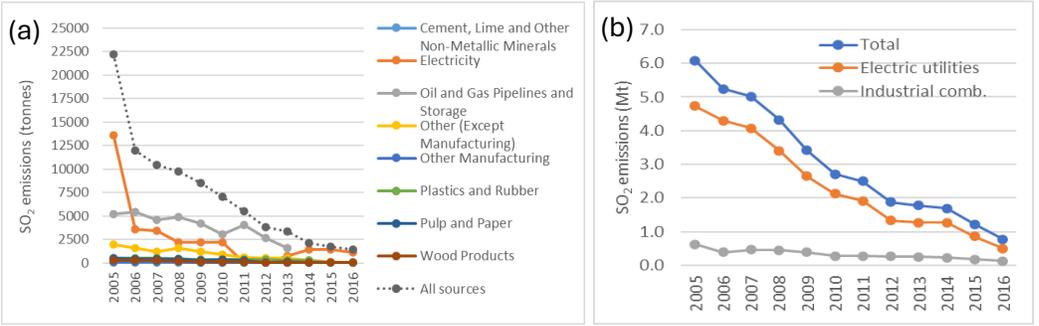
Figure S9: SO$_2$ emissions (a) local, within 150 km of KEJ (ECCC NPRI, 2023) and (b) regional, U.S. northeast and Midwest regions (USEPA NEI, 2023)
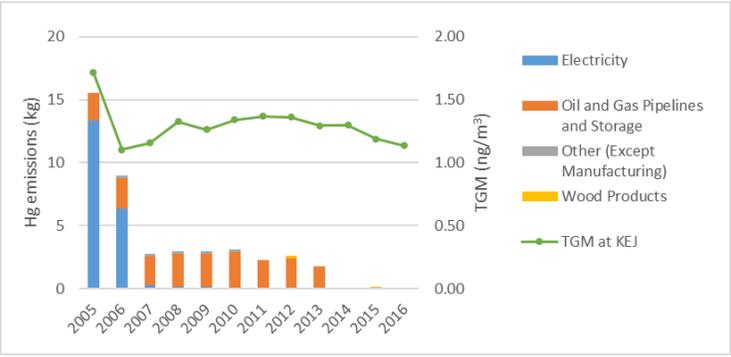
Figure S10: Hg emission sources within 150 km of KEJ in Provinces of Nova Scotia and New Brunswick (ECCC NPRI, 2023)

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