Supplementary Information

Identifying decadal trends in deweathered concentrations of criteria air pollutants in Canadian urban atmospheres with machine learning approaches

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Table S1a. Sampling site and period information for NO_2 at ten Canadian cities.

City	Period	Site ID	Latitude	Longitude	Note
Halifax	1996-2017	30118	44.646	-63.573	
Montreal	1995-2019	50103	45.641	-73.500	
Quebec	1996-2019	50308	46.821	-71.220	Data collected at 50307 (Lat. 46.824, Long. -71.235) in 1996-1997 were used for data loss.
Toronto	2003-2019	60430	43.709	-79.544	
Hamilton	1996-2019	60512	43.258	-79.862	
Winnipeg	1984-2018	70119	49.898	-97.147	
Edmonton	1994-2019	90130	53.545	-113.499	
Calgary	1986-2007	90227	51.048	-114.076	
Vancouver	1986-2019	100111	49.281	-122.849	
Victoria	1993-2019	100304	48.442	-123.363	

Table S1b. Sampling site and period information for CO at ten Canadian cities.

City	Period	Site ID	Latitude	Longitude	Note
Halifax	1983-2019	30118	44.646	-63.573	Date collected at other three sites within 300 m were used in 1983-1985, 1986-1990 and 2017-2019 for data loss
Montreal	1995-2010	50103	45.641	-73.500	
Quebec	1996-2019	50308	46.821	-71.220	Data collected at 50307 (Lat. 46.824, Long. -71.235) in 1996-1997 were used for data loss.
Toronto	2003-2019	60430	43.709	-79.544	
Hamilton	2000-2019	60512	43.258	-79.862	
Winnipeg	1982-2018	70119	49.898	-97.147	
Edmonton	1981-2019	90130	53.545	-113.499	
Calgary	1981-2015	90227	51.048	-114.076	Data collected at 90228 (Lat. 51.047, Long. -114.076) in 2009-2015 were used for data loss.
Vancouver	1981-2019	100111	49.281	-122.849	
Victoria	1998-2019	100304	48.442	-123.363	

Table S1c. Sampling site and period information for SO_2 at ten Canadian cities.

City	Period	Site ID	Latitude	Longitude	Note
Halifax	1982-2019	30118	44.646	-63.573	Data collected other two sites within 100 m were used in 1982-1999 and 2019 for the data loss.
Montreal	1995-2010	50103	45.641	-73.500	
Quebec	1996-2019	50308	46.821	-71.220	Data collected at 50307 (Lat. 46.824, Long. -71.235) in 1996-1997 were used for data loss.
Toronto	2003-2019	60430	43.709	-79.544	
Hamilton	1996-2019	60512	43.258	-79.862	
Winnipeg	1987-2018	70119	49.898	-97.147	
Edmonton	1987-2019	90121	53.545	-113.499	
Calgary	1983-2010	90218	51.009	-114.025	
Vancouver	1981-2019	100111	49.281	-122.849	
Victoria	1998-2019	100304	48.442	-123.363	

Table S1d. Sampling site and period information for O_3 at ten Canadian cities.

City	Period	Site ID	Latitude	Longitude	Note
Halifax	2000-2017	30118	44.646	-63.573	
Montreal	1995-2019	50103	45.641	-73.500	
Quebec	1995-2019	50308	46.821	-71.220	Data collected at 50307 (Lat. 46.824, Long. -71.235) in 1995-1997 were used for data loss.
Toronto	2003-2019	60430	43.709	-79.544	
Hamilton	1987-2019	60512	43.258	-79.862	
Winnipeg	1982-2018	70119	49.898	-97.147	
Edmonton	1981-2019	90130	53.545	-113.499	
Calgary	1982-2015	90227	51.048	-114.076	Data collected at 90228 （Lat. 51.047, Long. -114.076） were used in 2009-2015 for data loss
Vancouver	1981-2019	100111	49.281	-122.849	
Victoria	1998-2019	100304	48.442	-123.363	

Table S1f. Sampling site and period information for $PM_{2.5}$ at ten Canadian cities.

City	Period	Site ID	Latitude	Longitude	Note
Halifax	2006-2019	30113	44.647	-63.574	
Montreal	2004-2019	50103	45.641	-73.500	
Quebec	1998-2019	50308	46.821	-71.221	
Toronto	2000-2019	60430	43.709	-79.544	
Hamilton	1998-2019	60512	43.258	-79.862	
Winnipeg	1995-2018	70119	49.898	-97.147	
Edmonton	1998-2019	90130	53.545	-113.499	
Calgary	1998-2014	90227	51.048	-114.076	Data collected at 90228 (Lat. 51.047, Long. -114.076) in 2009-2014 were used after 2009
Vancouver	2003-2019	100111	49.281	-122.849	
Victoria	1998-2019	100304	48.444	-123.363	

Table S2. Correlations between deweathered CO mixing ratios and its original annual averages together with their correlations with provincial grand total and transportation CO emissions, and the decreasing extents of the variables in ten Canadian cities during the last decades ([&] decreasing trends were always obtained with $P<0.05$ except in Montreal; [&&]Provincial grand total and transportation CO emission decreasing percentage; [#] $P>0.05$; [##] since 1990; $R^2>0.8$ was highlighted in purple).

City	Correlation with original annual average (P<0.01)		Annual decreasing rate and overall decreasing percentage (unit: ppm $year^{-1}$, %) [&]			R^2 values between different types of mixing ratios with provincial total and transportation CO emissions (P<0.05)			Emission decreasing percentage (total, transportation; unit: %)[&&]
	BRTs	RF	BRTs	RF	original	BRTs	RF	original	
Halifax (1984-2019)	y=1.01*x	y=1.07*x	0.029, 90	0.030, 90	0.029, 92	0.83, 0.81	0.86, 0.83	0.77, 0.75	58, 63[##]
Montreal (1995-2010)	y=1.03*x	y=1.03*x	/[#]	/[#]	/[#]	/[#]	/[#]	/[#]	37, 53
Quebec (1996-2019)	y=1.07*x	y=1.12*x	0.013, 56	0.014, 56	0.010, 58	0.94, 0.93	0.93, 0.93	0.83, 83	42, 60
Toronto (2000-2019)	y=0.92*x	y=0.99*x	0.041, 84	0.046, 83	0.048, 86	0.71, 69	0.71, 0.69	0.70, 0.68	59, 62
Hamilton (2000-2019)	y=1.03*x	y=1.03*x	0.021, 70	0.021, 66	0.019, 68	0.80, 0.79	0.80-0.79	0.72, 0.71	59, 62
Winnipeg (1982-2018)	y=0.97*x	y=1.01*x	0.019, 84	0.020, 84	0.020, 88	0.91, 0.91	0.90, 0.90	0.84, 0.85	55, 55[##]
Edmonton (1981-2019)	y=0.98*x	y=1.02*x	0.048, 86	0.046, 86	0.048, 86	0.74, 0.82	0.73, 0.82	0.76, 0.83	45, 62[##]
Calgary (1981-2013)	y=1.01*x	y=1.07*x	0.064, 90	0.067, 90	0.063, 0.91	0.76, 0.88	0.75, 0.88	0.78, 0.89	42, 59[##]
Vancouver (1981-2019)	y=1.00*x	y=1.03*x	0.026, 82	0.027, 82	0.026, 83	0.96, 0.85	0.96, 0.87	0.96, 0.86	71, 53[##]
Victoria (1999-2019)	y=1.01*x	y=1.05*x	0.011, 57	0.012, 59	0.010, 58	0.91, 0.89	0.90, 0.89	0.84, 0.82	62, 56

Table S3. Correlations between deweathered SO_2 mixing ratios and its original annual averages together with their correlations with provincial grand total SO_2 emissions, and the decreasing extents of the variables in ten Canadian cities during the last decades ([&] decreasing trends were always obtained with P<0.05; [&&]Provincial grand total SO_2 emission decreasing percentage; [^] since 1990; [#] P>0.05; bond number represents lower decreasing percentage of SO_2 mixing ratios than that of provincial total grand SO_2 emissions; R^2>0.8 was highlighted in purple).

City	Correlation with original annual average (P<0.01)		Annual decreasing rate and overall decreasing percentage (unit: ppb year^{-1}, %) [&]			R^2 values between different types of mixing ratios with provincial grand total SO_2 emissions (P<0.05)			Emission decreasing percentage (total grand, unit: %)[&&]
	BRTs	RF	BRTs	RF	original	BRTs	RF	original	
Halifax (2000-2019)	y=1.00*x	y=1.08*x	0.63, 93	0.61, 90	0.59, 93	0.89	0.92	0.80	72
Montreal (1995-2010)	y=1.14*x	y=1.02*x	0.37, 86	0.35, 86	0.34, 79	0.86	0.82	0.82	76
Quebec (1996-2019)	y=0.96*x	y=1.05*x	0.10, 85	0.11, 78	0.11, 79	0.68	0.70	0.65	75
Toronto (2003-2019)	y=1.03*x	y=1.13*x	0.16, 0.95	0.17, 0.89	0.15, 90	0.84	0.89	0.85	79
Hamilton (1996-2019)	y=1.01*x	y=0.99*x	0.09, **23**	0.08, **27**	0.10, **28**	0.57	0.51	0.42	**81**
Winnipeg (1987-2018)	y=1.04*x	y=1.16*x	0.06, 96	0.07, 97	0.06, 95	0.91	0.85	0.90	92[^]
Edmonton (1987-2019)	y=1.00*x	y=1.05*x	0.07, 55	0.07, 53	0.07, 52	0.80	0.81	0.73	57[^]
Calgary (1983-2010)	y=1.00*x	y=1.04*x	0.09, 62	0.10, 62	0.09, 64	0.83	0.84	0.80	30[^]
Vancouver (1982-2019)	y=0.98*x	y=1.02*x	0.11, 90	0.11, 91	0.12, 95	0.41	0.40	/[#]	38[^]
Victoria (1999-2019)	y=1.00*x	y=1.08*x	0.05, 80	0.05, 73	0.04, 82	/[#]	/[#]	/[#]	/[#]

Table S4. Correlations between deweathered O_3 (NO_2+O_3) and its original annual averages, and their increasing or decreasing extents in ten Canadian cities during the last decades ([#] P<0.05; [##]P>0.05; [&] 1996-2019 in Quebec and Hamilton, 1994-2019 in Edmonton, 1986-2010 in Calgary).

O_3 mixing ratio						NO_2+O_3 mixing ratio				
City	Correlation with original annual average (P<0.01)		Annual **increasing** rate and overall increase (unit: ppb year^{-1}, ppb)[#]			Correlation with original annual average (P<0.01)		Annual **decreasing** rate and overall decrease (unit: ppb year^{-1}, ppb)[#]		
	BRTs	RF	BRTs	RF	Original	BRTs	RF	BRTs	RF	Original
Halifax (2000-2017)	y=0.98*x	y=0.98*x	/[##]	/[##]	/[##]	y=1.00*x	y=1.03*x	0.51, 10	0.52, 10	0.62, 10
Montreal (1997-2010)	y=0.97*x	y=0.98*x	0.13, 3	0.12, 3	0.16, 4	y=1.00*x	y=1.00*x	0.23, 4	0.22, 4	0.22, 4
Quebec (1995-2019)	y=1.01*x	y=1.01*x	0.33, 7	0.34, 7	0.27, 6	y=1.00*x	y=1.00*x	0.15[&], 1	0.14[&], 1	0.13[&], 1
Toronto (2003-2019)	y=0.96*x	y=0.95*x	0.10, 2	0.14, 2	0.22, 3	y=1.00*x	y=1.00*x	0.62, 10	0.62, 10	0.59, 10
Hamilton (1996-2019)	y=1.00*x	y=1.01*x	0.32, 8	0.31, 8	0.35, 8	y=1.00*x	y=1.00*x	0.21[&], 3	0.19[&], 3	0.21[&], 3
Winnipeg (1985-2018)	y=0.98*x	y=0.98*x	0.24, 5	0.25, 5	0.25, 7	y=1.00*x	y=1.00*x	0.14, 6	0.14, 6	0.17, 6
Edmonton (1981-2019)	y=1.00*x	y=1.01*x	0.16, 10	0.17, 10	0.17, 10	y=1.00*x	y=1.01*x	0.28[&], 8	0.30[&], 9	0.33[&], 10
Calgary (1986-2014)	y=1.00*x	y=1.01*x	0.26, 8	0.27, 8	0.24, 8	y=1.00*x	y=1.01*x	0.37[&], 6	0.41[&], 6	0.42[&], 6
Vancouver (1986-2019)	y=0.99*x	y=1.02*x	0.11, 2	0.10, 2	0.11, 2	y=1.00*x	y=1.01*x	0.30, 12	0.29, 11	0.28, 10
Victoria (1999-2019)	y=1.00*x	y=0.99*x	0.20, 2	0.19, 2	0.16, 1	y=0.99*x	y=0.99*x	/[##]	/[##]	/[##]

Fig. S1. Time series of $PM_{2.5}$ concentrations during two large wildfire periods in Edmonton.

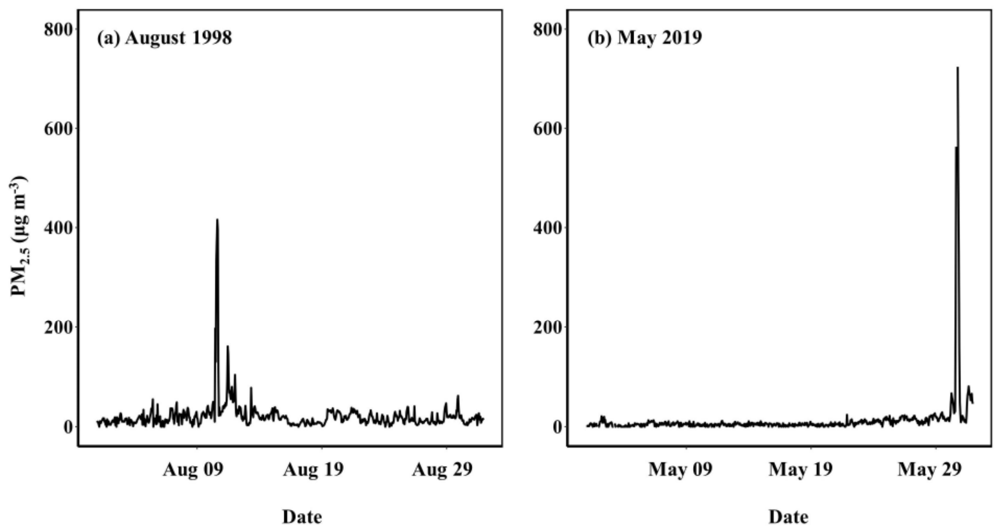
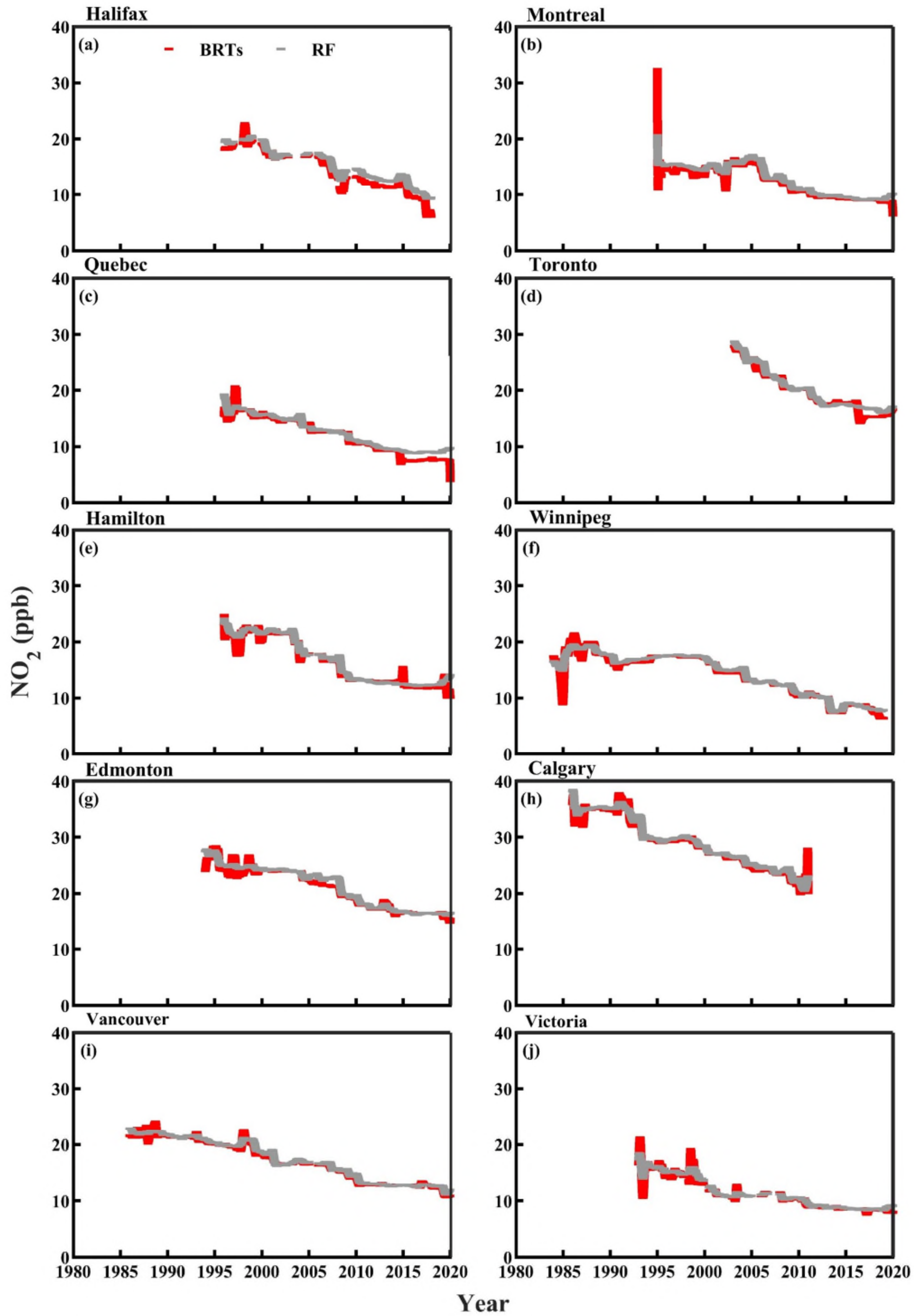


Fig. S2. BRTs-deweathered and RF-deweathered hourly mixing ratios of NO_2 in ten Canadian cities.



Note: In theory, the deweathered values should be invariant in absence of mitigation of NO_x in one or several consecutive years. In reality, the deweathered values showed a step-wise decrease due to the implemented air pollution control measures, as can be seen in Figures a—j. The spikes were probably associated with unpredictably increased emissions of NO_x, e.g., in January of 1995 in Montreal (Figure b); the trough might be associated with unpredictably decreased emissions of NO_x, e.g., in January of 1985 in Winnipeg (Figure f). BRTs-deweathered values apparently captured, but RF-deweathered values did not capture the unpredictably increased or decreased emissions.

Fig. S3. Correlations of BRTs-deweathered and RF-deweathered annual averages of NO_2 and $PM_{2.5}$ with their respective original annual averages in Halifax ((a): NO_2; (b): $PM_{2.5}$).

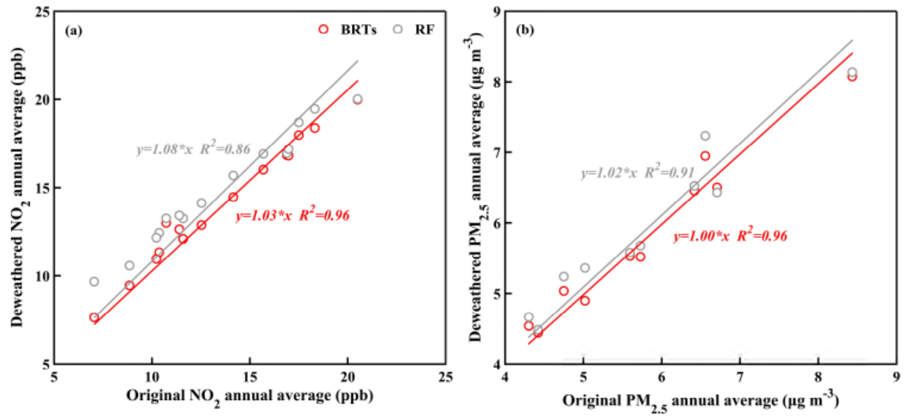


Fig. S4. Trends in original annul averages of CO and SO_2 in five eastern (top row) and five western (bottom row) Canadian cities.

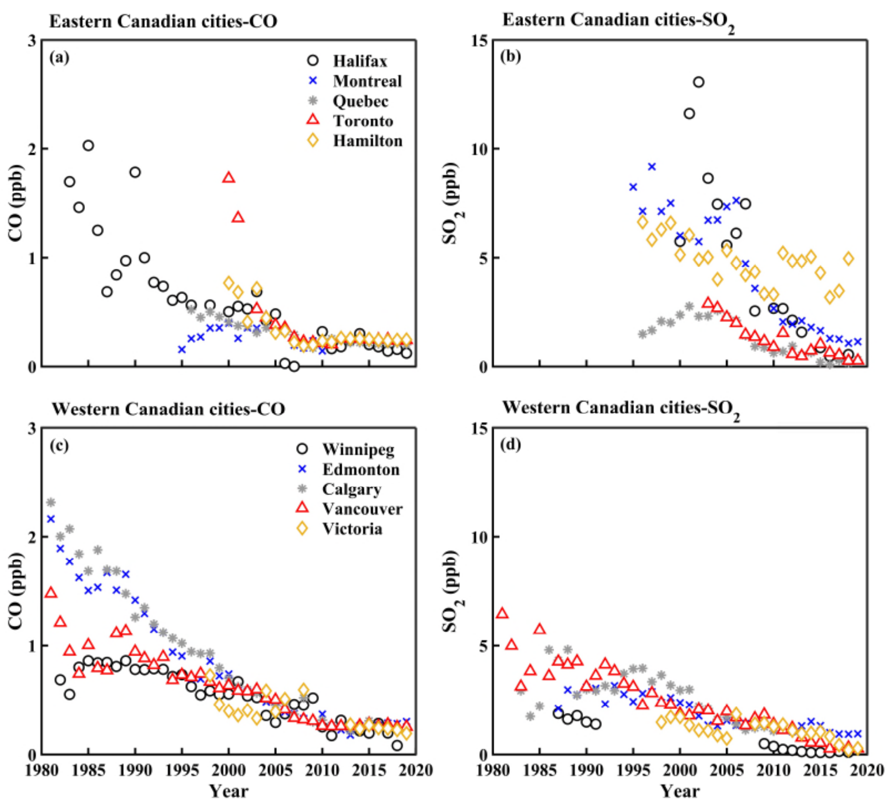


Fig. S5. Trends in original annul averages of O_3 and NO_2+O_3 in five eastern (top row) and five western (bottom row) Canadian cities.

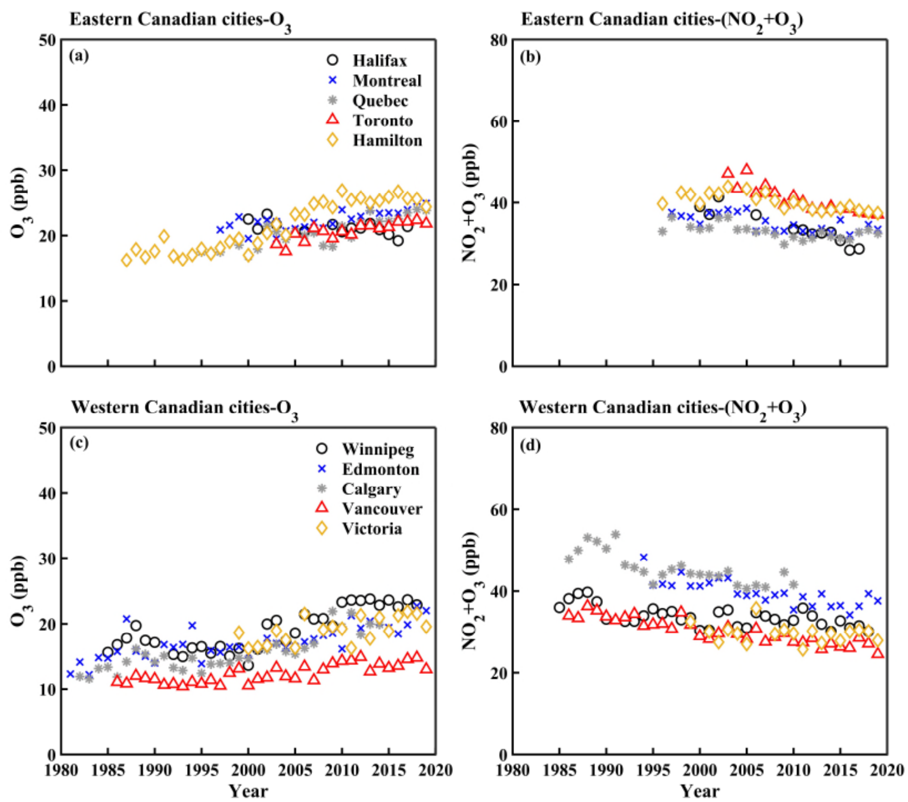


Fig. S6. BRTs-deweathered and RF-deweathered hourly mass concentrations of $PM_{2.5}$ in ten Canadian cities.

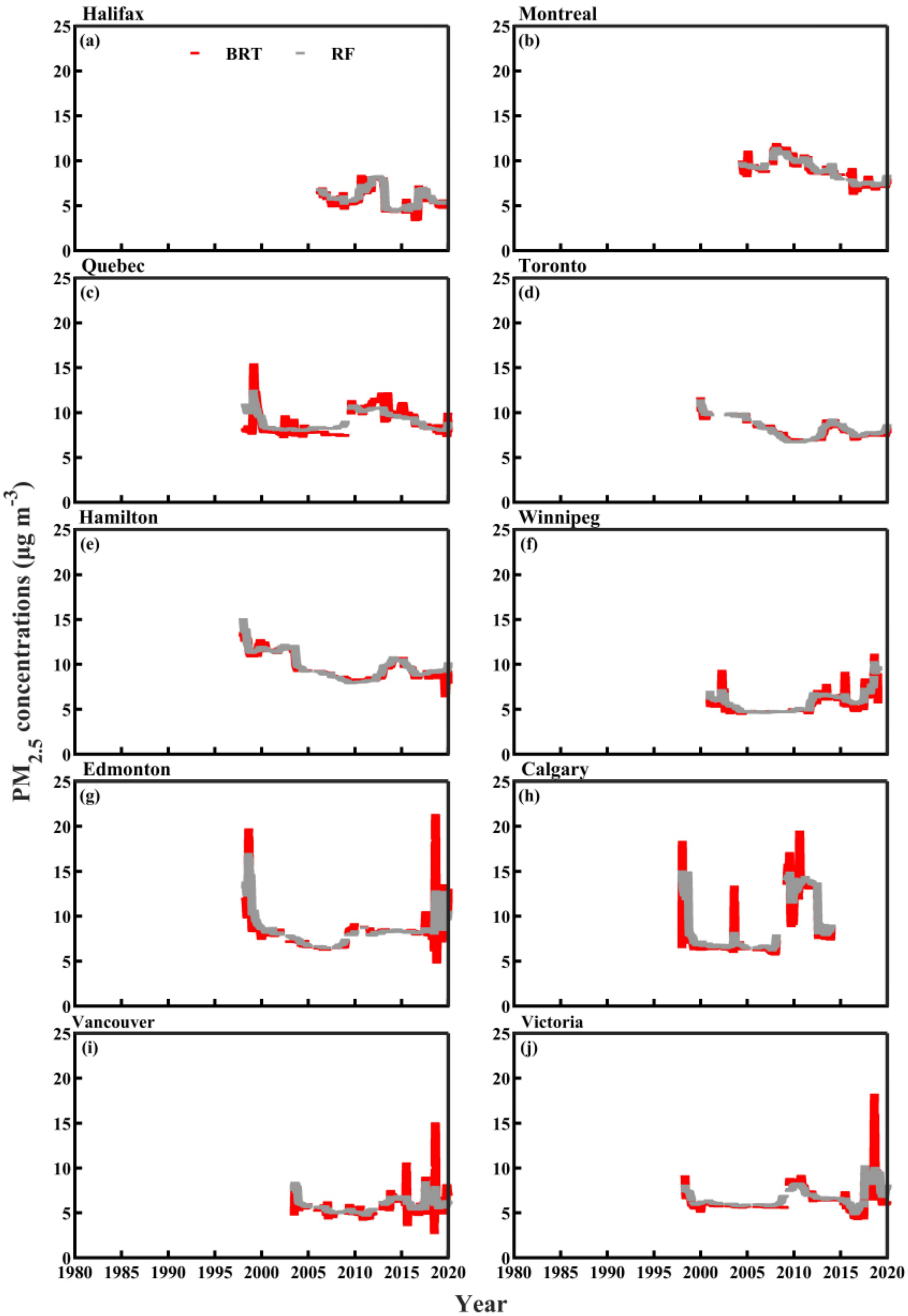


Fig. S7. Average and standard deviation of 95th-100th percentile concentrations of $PM_{2.5}$ in ten Canadian cities.

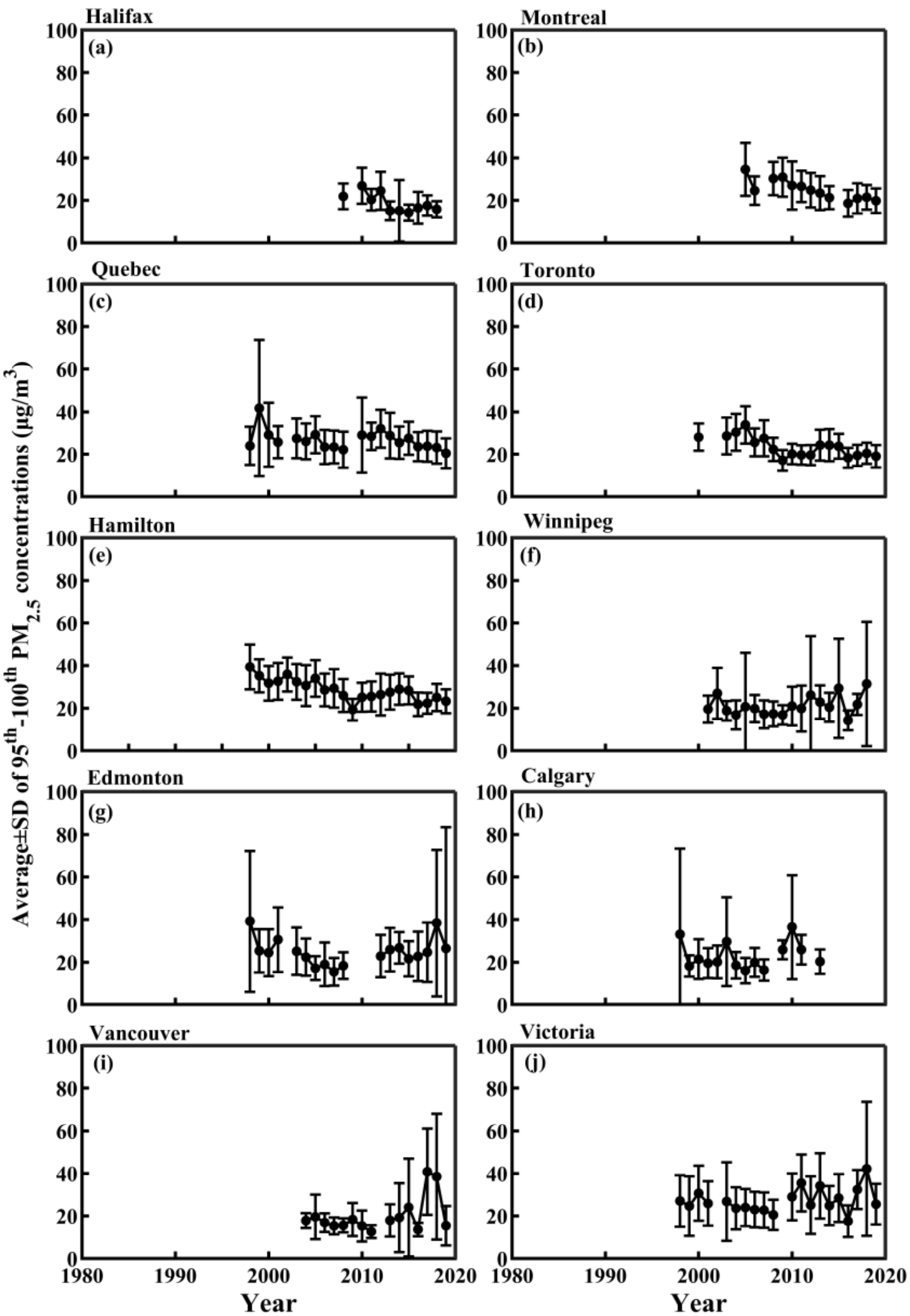


Fig. S8. BRTs-deweathered and RF-deweathered hourly mixing ratios of O_3 (left column) and NO_2+O_3 (right column) at the high level ≥40 ppb in five western Canadian cities.

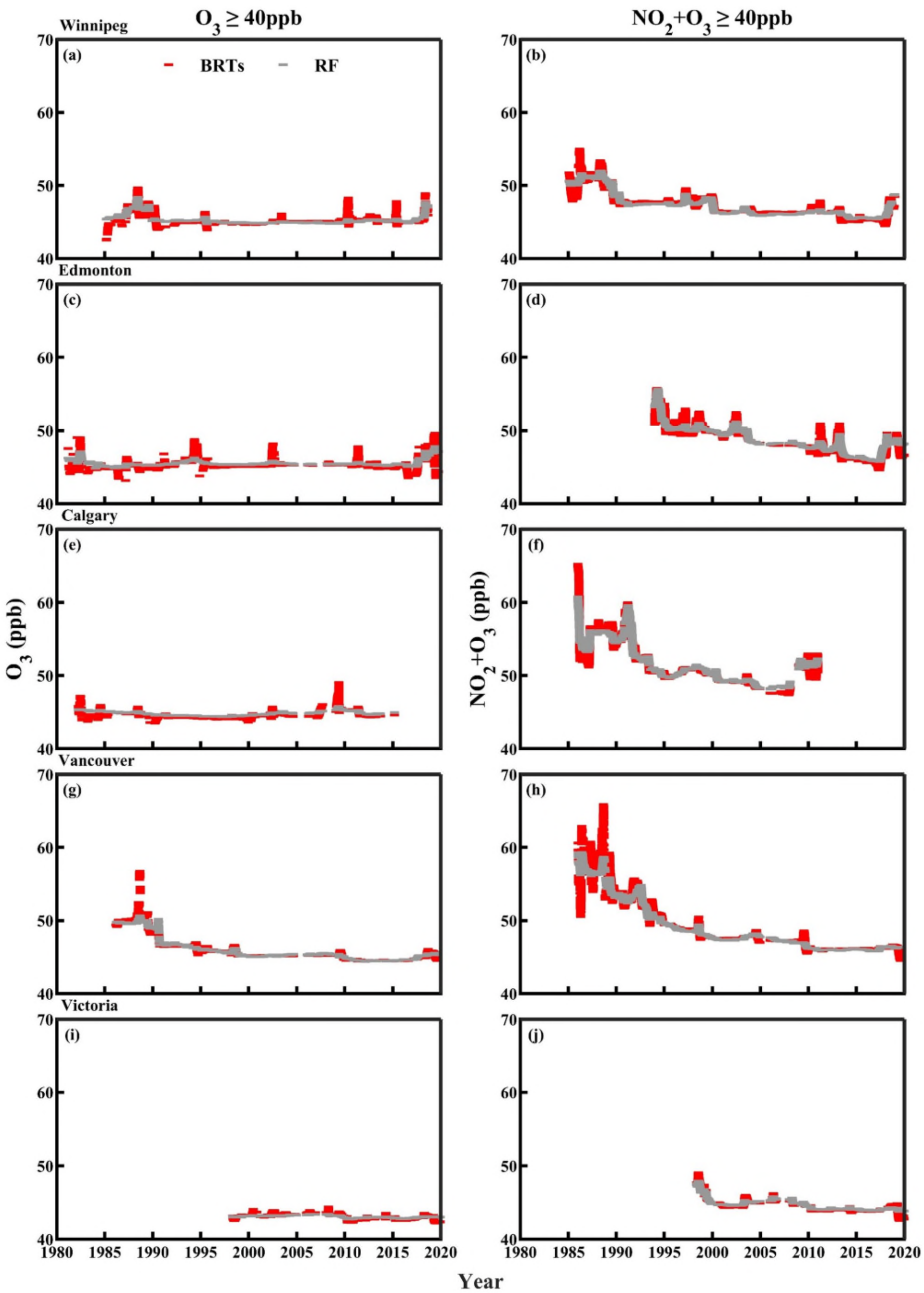


Fig. S9. The calculated AQHI in five eastern Canadian cities (left column shows original AQHI; right column shows annual average of AQHI and percentages of AQHI ≥7 and ≥10).

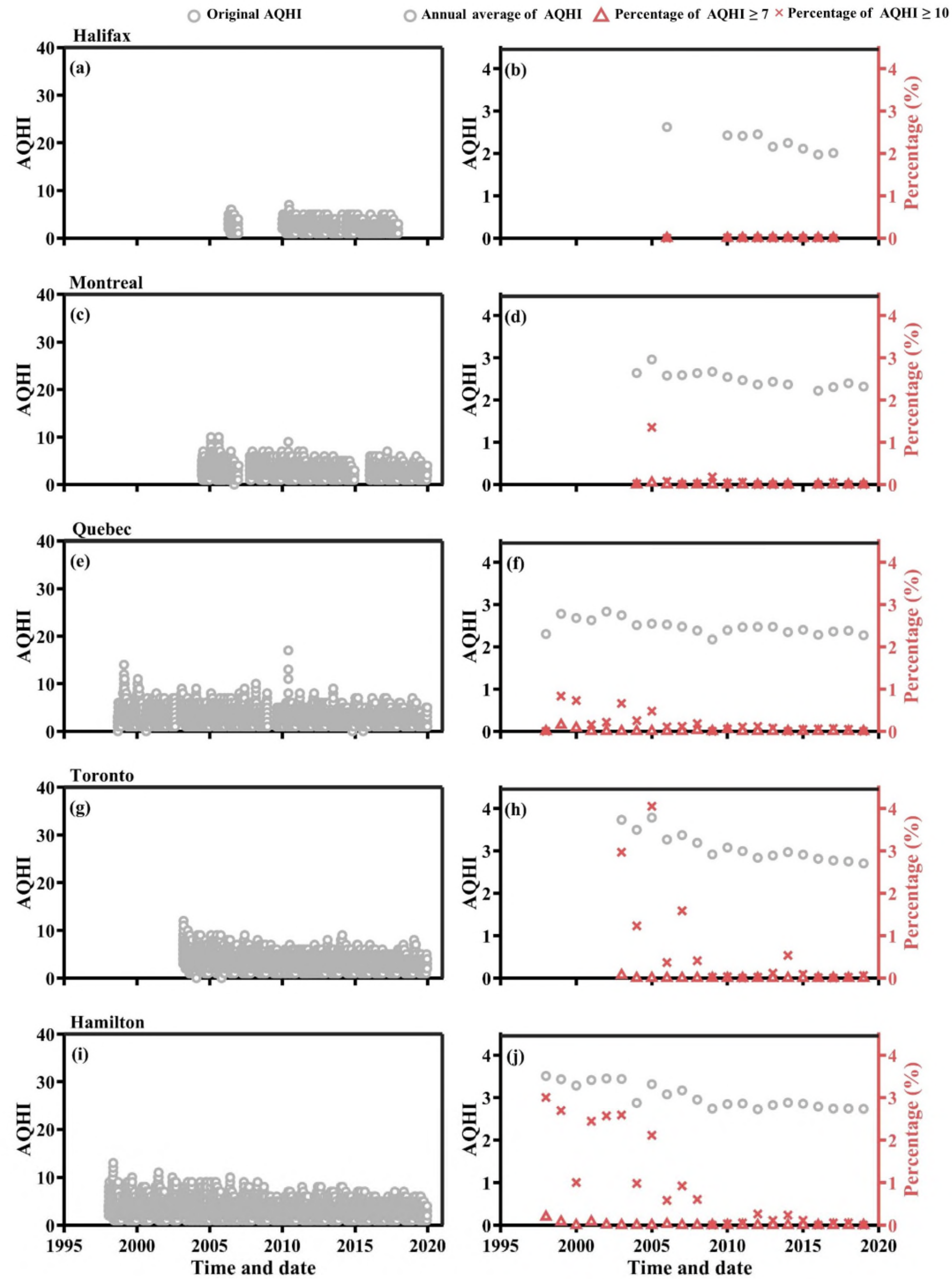


Fig. S10. The calculated AQHI in five western Canadian cities (left column shows original AQHI; right column shows annual average of AQHI and percentages of AQHI ≥7 and ≥10).