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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 sites and periods for NO_2 at ten Canadian cities (hourly averages in ppb were available).

City	Period	Site ID	Latitude	Longitude	Note
Halifax	1996-2017	30118	44.646	-63.573	
Montreal	1995-2019	50103	45.641	-73.500	
Quebec city	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 sites and periods for CO at ten Canadian cities (hourly averages in ppb were available).

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 city	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 sites and periods for SO_2 at ten Canadian cities (hourly averages in ppb were available).

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 city	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 sites and periods for O_3 at ten Canadian cities (hourly averages in ppb were available).

City	Period	Site ID	Latitude	Longitude	Note
Halifax	2000-2017	30118	44.646	-63.573	
Montreal	1995-2019	50103	45.641	-73.500	
Quebec city	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 S1e. Sampling sites and periods for $PM_{2.5}$ at ten Canadian cities (hourly averages in μg m^{-3} were available).

City	Period	Site ID	Latitude	Longitude	Note
Halifax	2006-2019	30113	44.647	-63.574	
Montreal	2004-2019	50103	45.641	-73.500	
Quebec city	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 S2a. SO_2 emissions from registered facilities in ten Canadian cities (tons $year^{-1}$). Blank cells represent no data available.

Year	Halifax	Montreal	Quebec City	Toronto	Hamilton	Winnipeg	Edmonton	Calgary	Vancouver	Victoria
2002	638	3628		273	8073	0	1908	0	0	217
2003	741	4197		269	8134	0	1788	0	0	212
2004	705	3313		272	8435	0	2195	0	0	273
2005	41	3649		283	12864	0	1445	6	0	269
2006	35	2786		171	8306	0	1531	6	2	261
2007	555	3008		152	7649	0	1373	6	0	
2008	568	2660		105	8586		3995	6	2	265
2009	328	3113		25	7284	2	5176	14	0	215
2010	271	2540	163	29	11803	3	3733	6	86	8
2011	302	2490	661	1	12776		4276	1	119	11
2012	277	1188	631	0	13310		3463	0	164	2
2013	260	1175	361	1	11558		4558	0	158	3
2014	185	1148	643	0	10977		3728	0	104	7
2015	63	1148	959	0	11436		3538	1	119	11
2016	10	2675	982	0	10072		3814	1	118	
2017	4	3404	977	1	8934		4014	1	124	
2018	0	3183	1024	0	12154		3101	0	96	
2019	0	3338	847	0	2507		3534	0	119	

Table S2b. NO_x emissions from registered facilities in ten Canadian cities (tons $year^{-1}$). Blank cells represent no data available.

Year	Halifax	Montreal	Quebec City	Toronto	Hamilton	Winnipeg	Edmonton	Calgary	Vancouver	Victoria
2002	102	1791	0	1142	6306	3	3329	68	178	
2003	85	1876	1	1051	6279	1	3285	166	208	113
2004	77	1751	25	1253	6086	23	3111	229	297	117
2005	14	1666	21	1119	7059	22	3258	171	318	84
2006	12	1388	21	960	5961	21	4732	271	267	81
2007	192	1669	22	791	5890	60	4444	262	278	29
2008	201	1386		644	5732	70	4274	192	253	94
2009	98	1464		322	4248	34	3832	131	188	60
2010	81	1369	44	267	6394	42	3849	108	593	20
2011	113	1408	202	186	5843	26	3205	184	602	22
2012	109	1402	202	179	5971	24	3497	152	665	17
2013	80	1430	100	162	5792	22	2822	197	650	19
2014	45	1252	167	232	5448	18	2448	150	587	20
2015	21	1315	856	227	4942	10	2631	218	268	21
2016	18	1327	866	191	4800	14	2467	202	447	19
2017	7	1397	860	184	4867	13	2624	295	573	19
2018	6	1406	902	177	5254	13	3109	352	533	15
2019	6	1598	753	178	1410	33	2719	391	278	

Table S3. Regression of (BRTs and RF) deweathered against original annul average CO mixing ratios, annual decreasing rate (ppm year^{-1}) and overall decreasing percentage (%) of deweathered and original CO mixing ratios (decreasing trends were always obtained with $P<0.05$ except in Montreal), correlation (R^2) of deweathered and original CO mixing ratios against provincial total CO emissions and transportation CO emissions ($P<0.05$), and percentage decreases (%) of the provincial total CO emissions and transportation CO emissions ($^{\#}P>0.05$; $^{\#\#}$since 1990; and $R^2>0.8$ was highlighted in purple).

City	Regression of deweathered against original mixing ratio (P<0.01)		Annual decreasing rate (ppm year^{-1}) and overall decreasing percentage (%) (P<0.05)			Correlation (R^2) of mixing ratios against provincial total and transportation CO emissions (P<0.05)			Percentage decreases (%) of provincial total and transportation emissions
	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 city (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 S4. Regression of (BRTs and RF) deweathered against original annul average SO_2 mixing ratios, annual decreasing rate (ppb year^{-1}) and overall decreasing percentage (%) of deweathered and original SO_2 mixing ratios (decreasing trends were always obtained with $P<0.05$), correlation (R^2) of deweathered and original SO_2 mixing ratios against provincial total SO_2 emissions and transportation SO_2 emissions ($P<0.05$), and percentage decreases (%) of the provincial total SO_2 emissions and transportation SO_2 emissions ($^{\#}P>0.05$; $^{\#\#}$since 1990; Bold font numbers represent cases with smaller deceasing percentages in SO_2 mixing ratios than in corresponding provincial emissions; and $R^2>0.8$ was highlighted in purple).

City	Regression of deweathered against original mixing ratio (P<0.01)		Annual decreasing rate (ppb year^{-1}) and overall decreasing percentage (%) (P<0.05)			Correlation (R^2) of mixing ratios against provincial total and transportation SO_2 emissions (P<0.05)			Percentage decreases (%) of provincial total and transportation emissions
	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 City (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 S5. Regression of (BRTs and RF) deweathered against original annul average O_3 and O_x missing ratios, and their increasing or decreasing rates (ppb year^{-1}) and overall increases or decreases (ppb) in ten Canadian cities during the last decades ($^{\#}$P>0.05; $^{\&}$1996-2019 in Quebec and Hamilton, 1994-2019 in Edmonton, and 1986-2010 in Calgary).

	O_3 mixing ratio					O_x mixing ratio				
City	Regression of deweathered against original mixing ratio (P<0.01)		Annual decreasing rate (ppb year^{-1}) and overall decrease (ppb) (%) (P<0.05)			Regression of deweathered against original mixing ratio (P<0.01)		Annual decreasing rate (ppb year^{-1}) and overall decrease (ppb) (%) (P<0.05)		
	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 City (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.

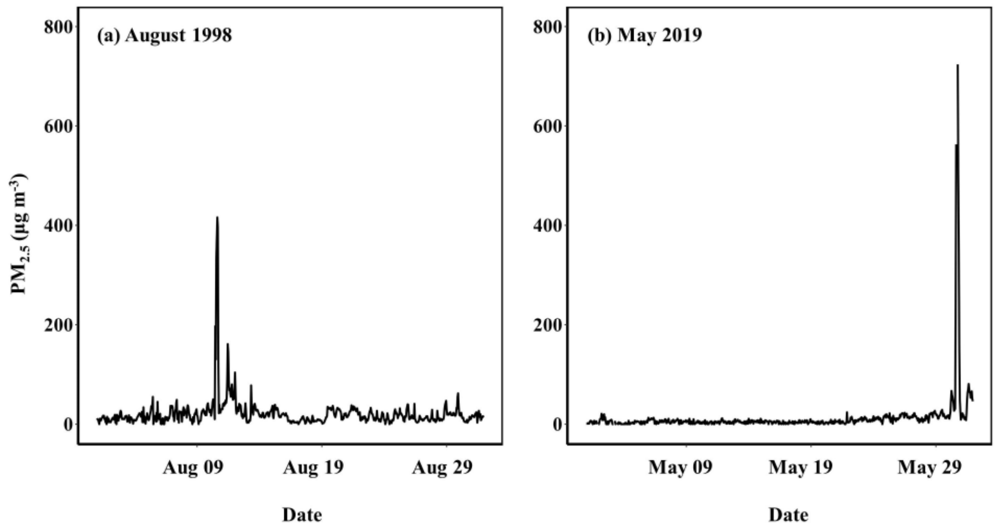
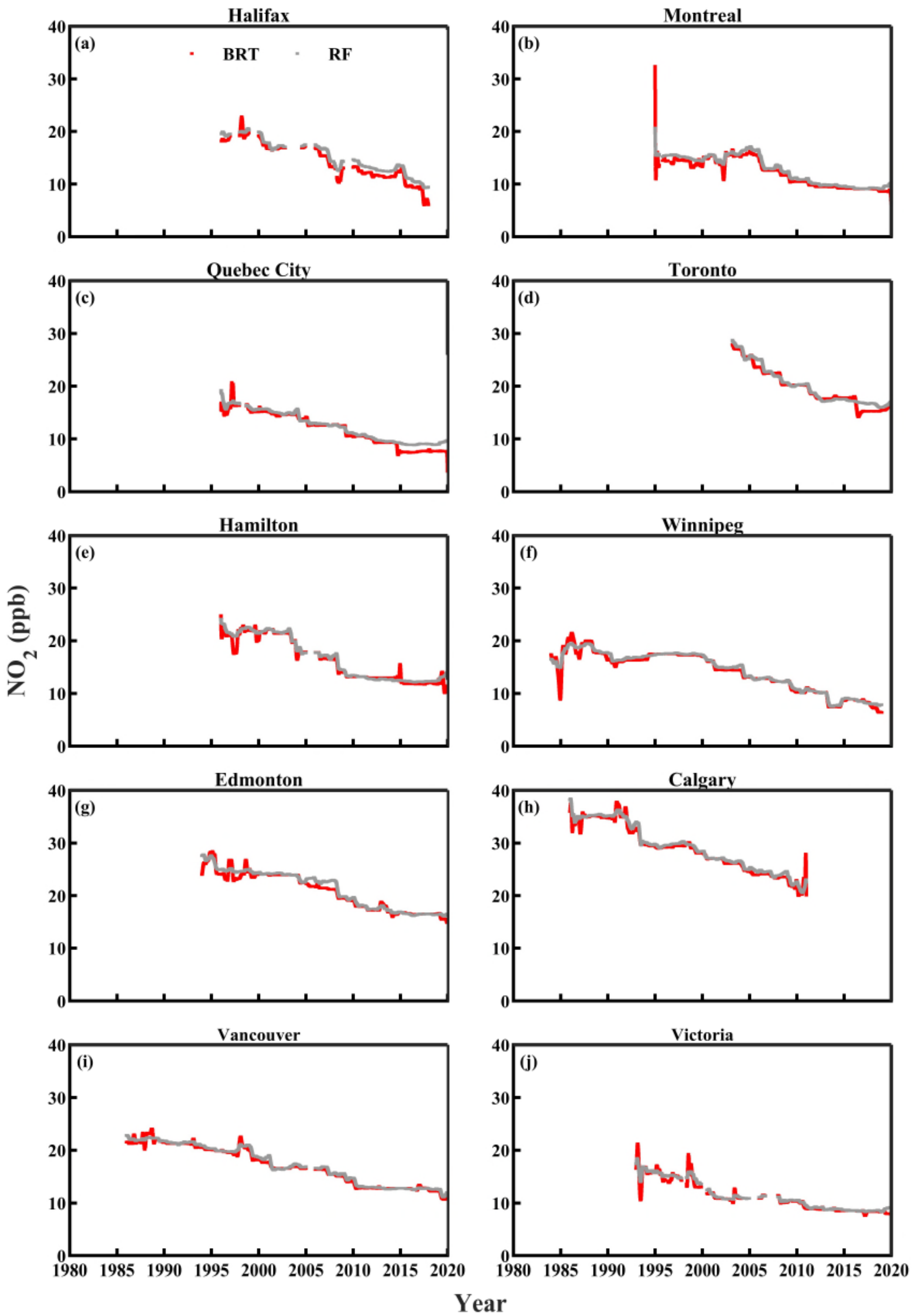


Fig. S2. BRTs-deweathered and RF-deweathered hourly mixing ratios of NO_2 in ten Canadian cities.



Note: The deweathered values are theocratically invariant in absence of mitigation of NO_x in one year or consecutive several years. However, the deweathered values showed a step-wise decrease with the implemented emission control measures, which were generally observed in a—j. The spikes were probably associated with unpredictably increased emissions of NO_x, e.g., in January of 1995 in Montreal, and the trough might be associated with unpredictably decreased emissions of NO_x, e.g., in January of 1985 in Winnipeg. BRTs-deweathered values apparently better captured the unpredictably increased or decreased emissions, but this is not the case for the RF-deweathered values.

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

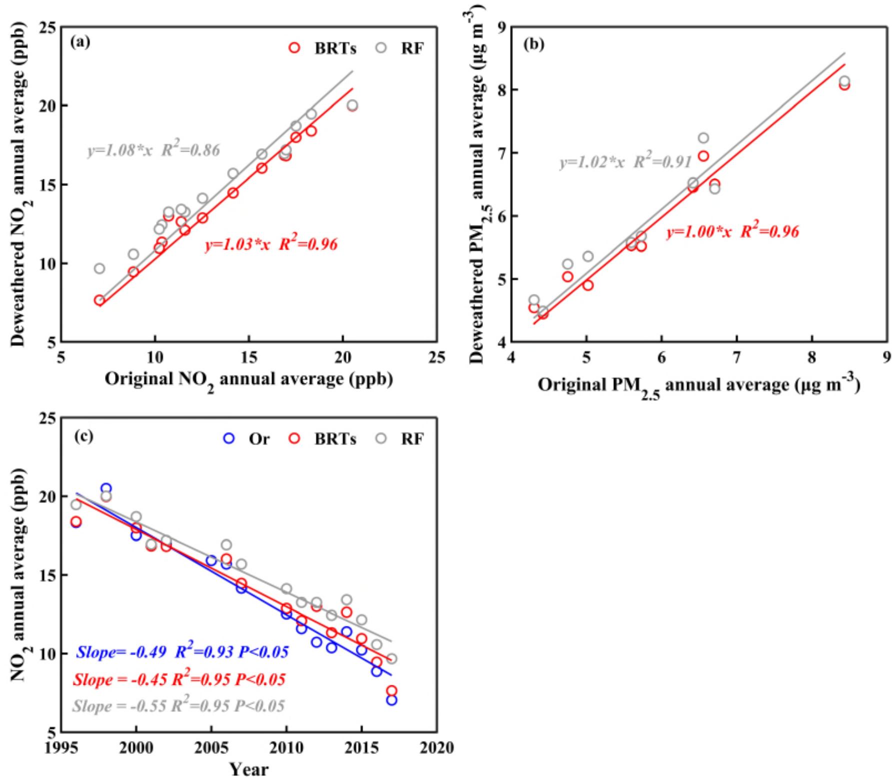


Fig. S4. Trends in original annul averages of CO and SO_2 in five eastern (top row) and five western (bottom row) Canadian cities.

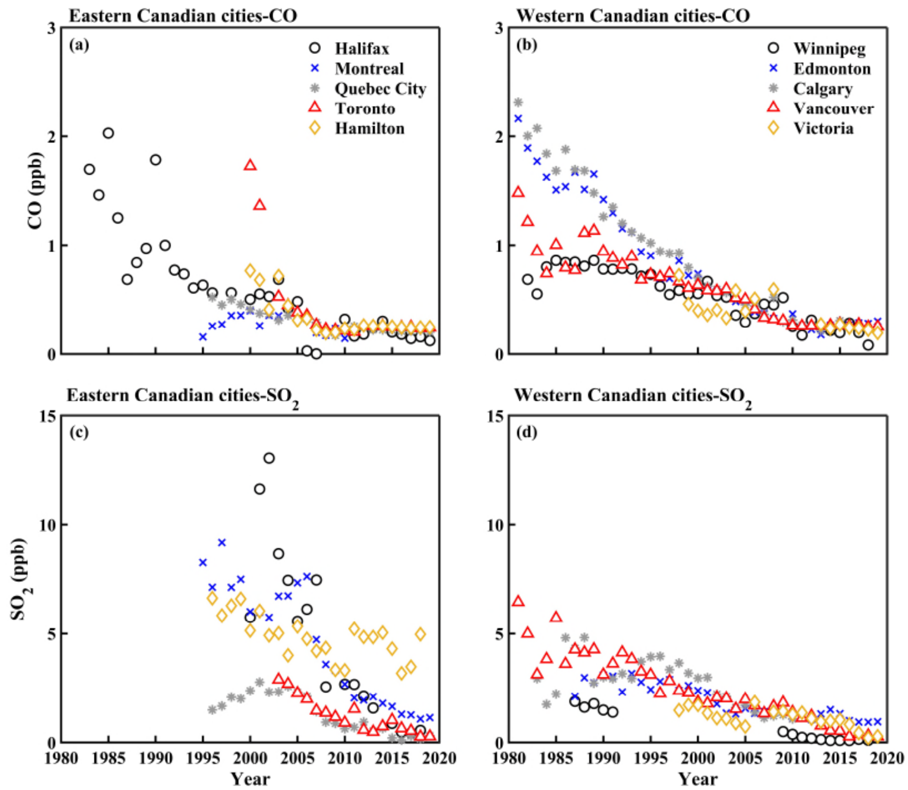


Fig. S5. Trends in original annul averages of O_3 and O_x in five eastern (top row) and five western (bottom row) Canadian cities.

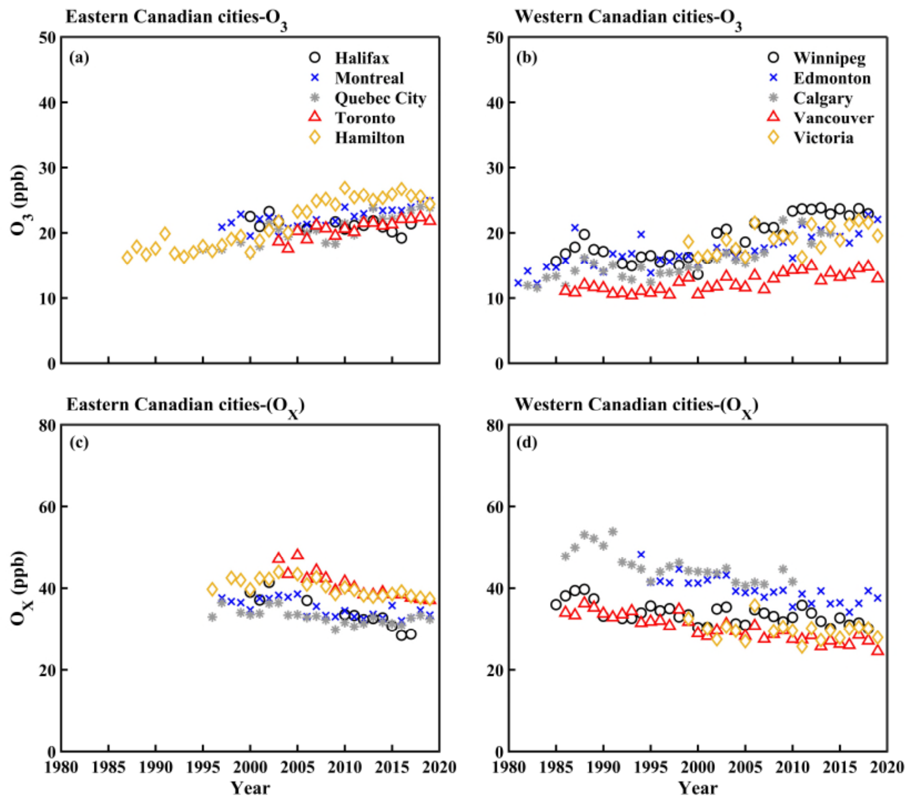


Fig. S6. BRTs- and RF-deweathered hourly mass concentrations of $PM_{2.5}$ in ten Canadian cities.

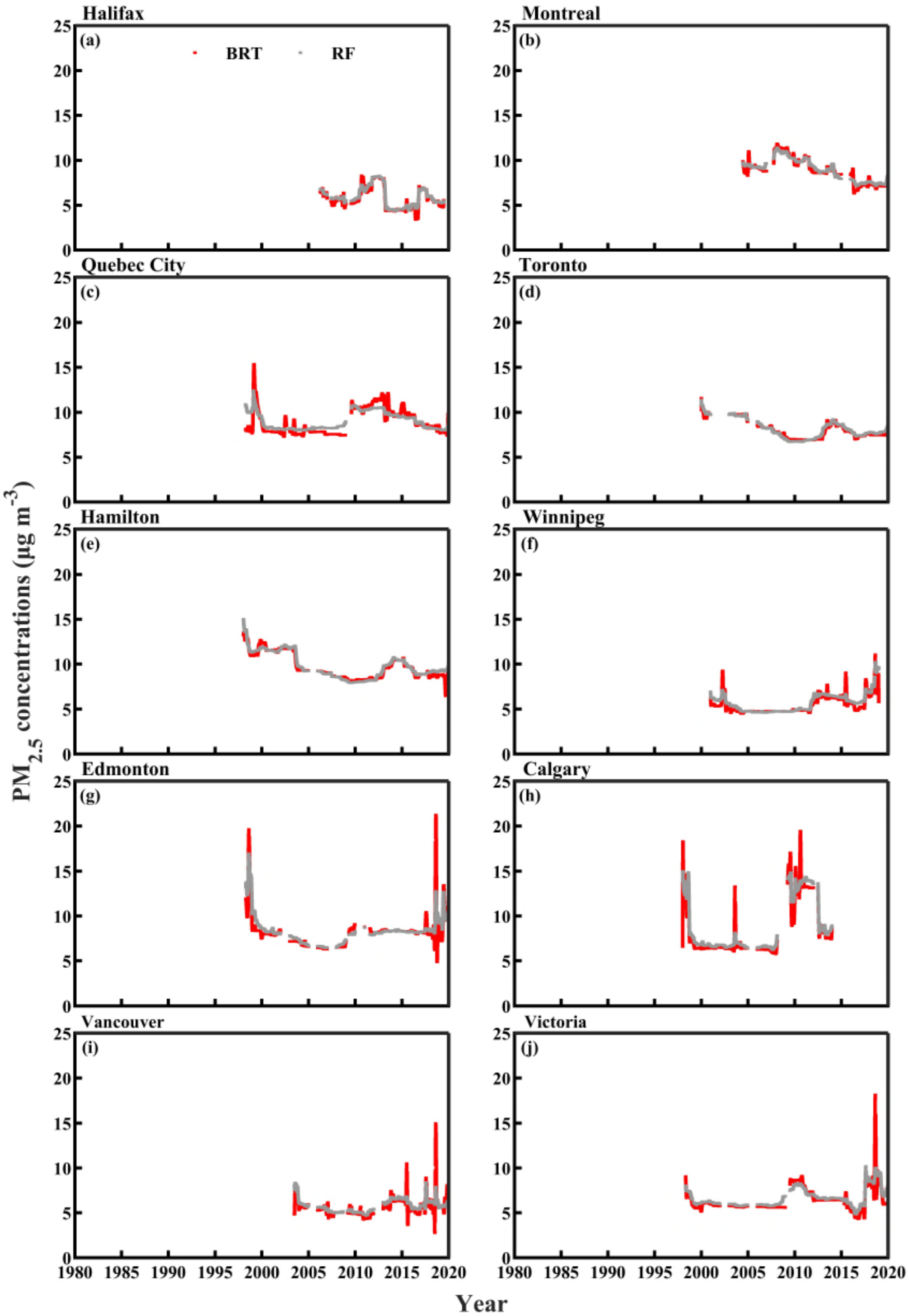


Fig. S7. Average and standard deviation of 95^{th}-100^{th} percentile concentrations of $PM_{2.5}$ in ten Canadian cities.

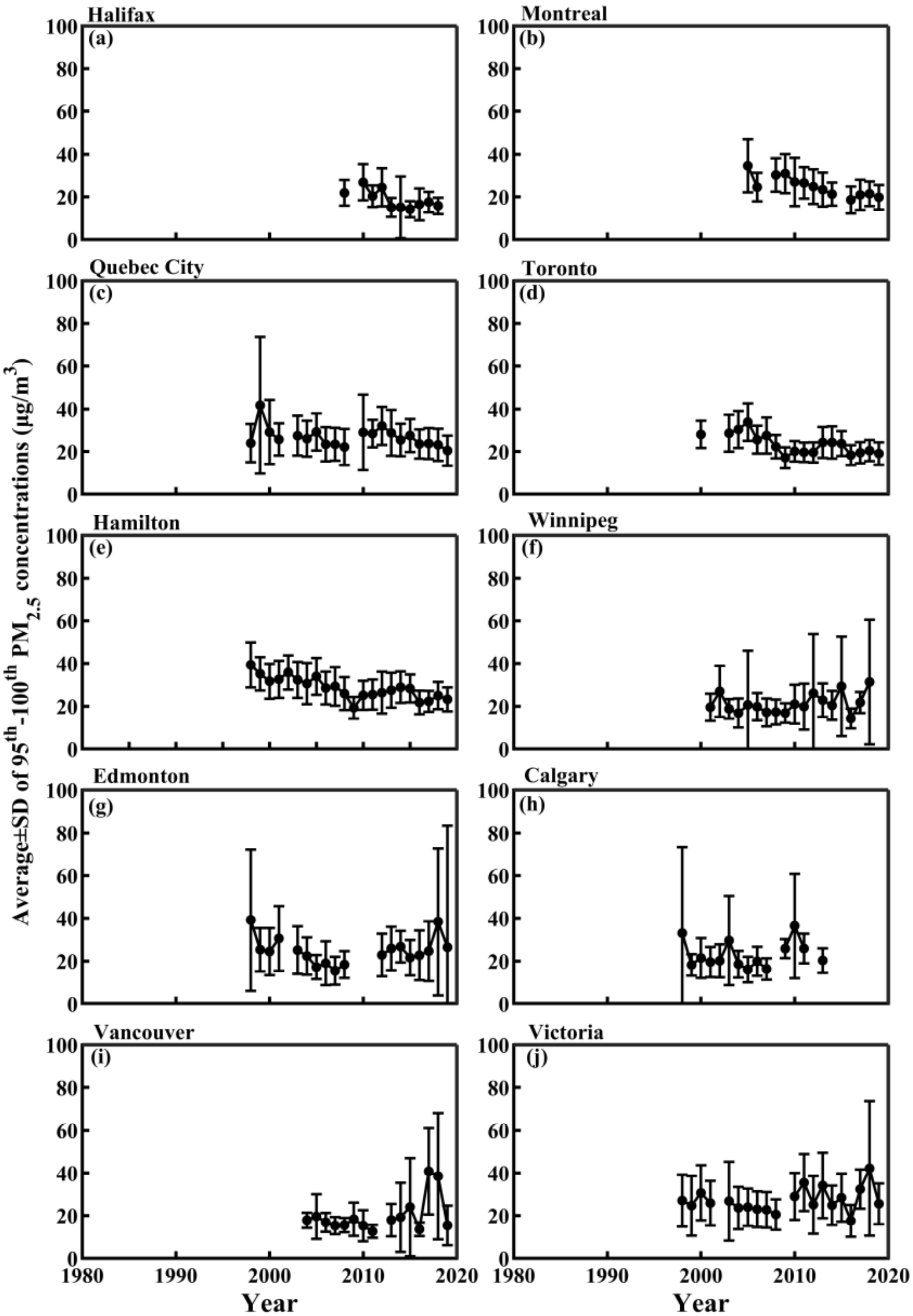


Fig. S8. BRTs- and RF-deweathered hourly mixing ratios of O_3 (left column) and O_x (right column) at the level ≥40 ppb in five western Canadian cities.

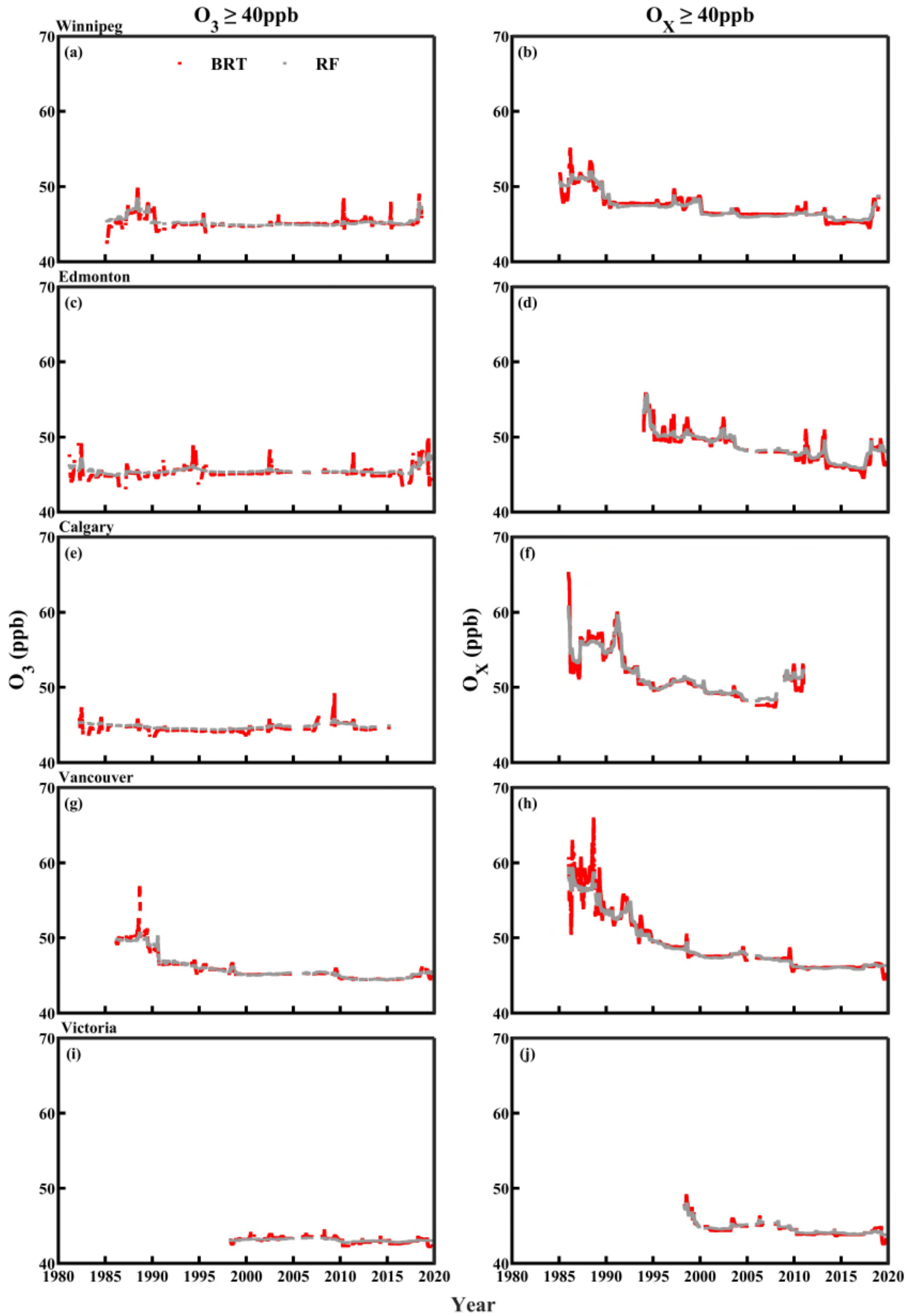


Fig. S9. The calculated AQHI in five eastern Canadian cities (left column shows original AQHI and right column shows annual average of AQHI and percentages of AQHI ≥7 and ≥10).

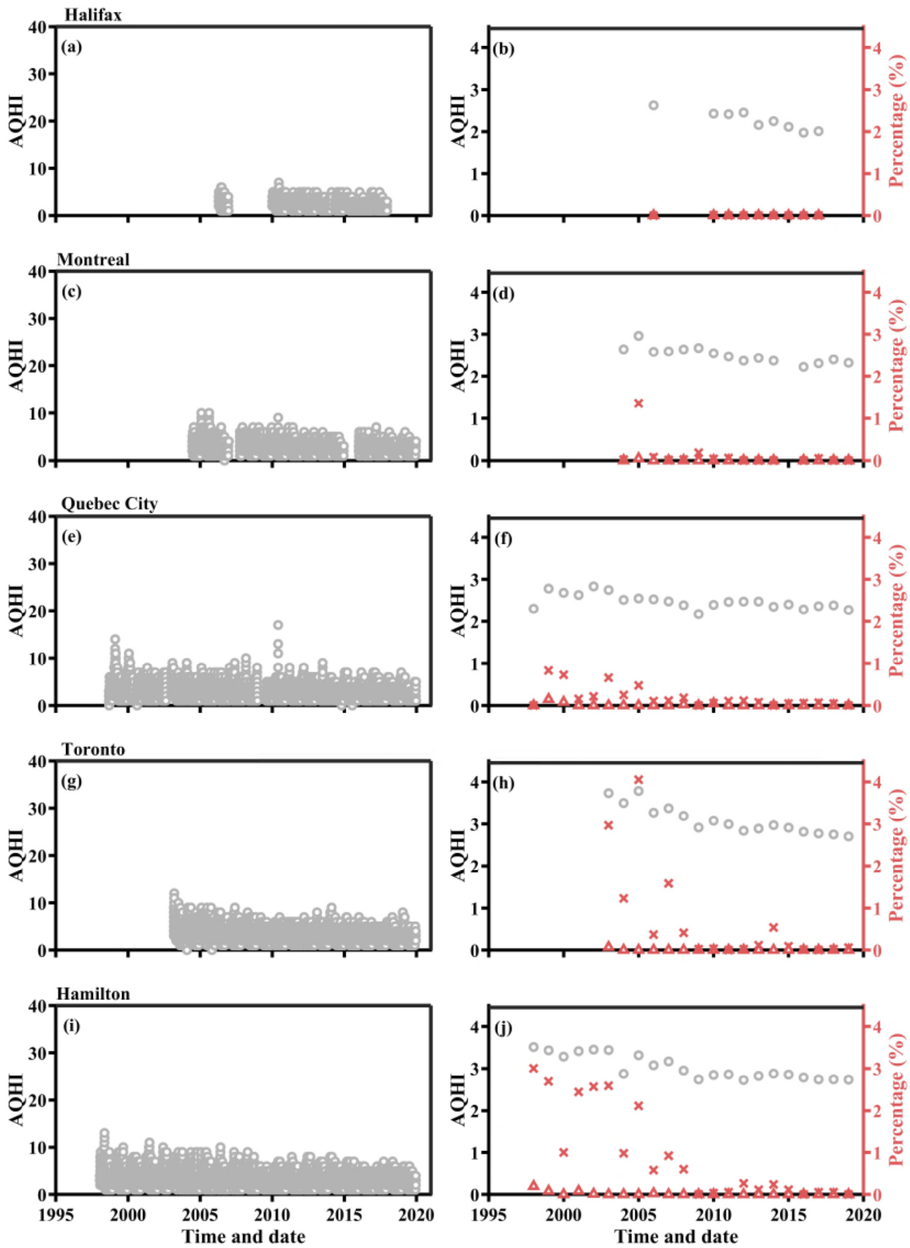


Fig. S10. The calculated AQHI in five western Canadian cities (left column shows original AQHI and right column shows annual average of AQHI and percentages of AQHI ≥7 and ≥10).