Evaluation of Chemical Transport Model Predictions of Primary Organic Aerosol (POA) for Air Masses Classified using Meteorological Back Trajectories and Particle-Component-Based Factor Analysis

Environment Canada Environnement Canada

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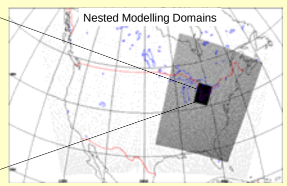
Introduction

The carbonaceous component of PM is the most complex component to characterize due to the wide range of constituent organic compounds with different sources and varying chemical reactivity and physical properties such as volatility, light scattering, light absorption, and hygroscopicity. Primary organic aerosol (POA) and black carbon (BC) are two key PM chemical components that are co-emitted from combustion sources. POA is important because it is emitted in large quantities, it can act as a seed for vapour condensation, it can coat and alter the optical properties of BC particles , and it is believed to contain some toxic materials. Traditionally, POA has been treated as an inert tracer transported in models; however, recent laboratory studies have raised some doubts about this assumption (Robinson et al., 2010). For the above reasons, it is important to try to isolate POA from SOA in model evaluation studies. This study presents a detailed evaluation on the current state of chemical transport model (CTM) predictions of primary organic aerosol (POA) using Environment Canada's unified regional AQ modelling system (AURAMS) (Makar *et al*., 2010a; Stroud *et al*., 2011), recent Canadian and U.S. national emissions inventories, and detailed field measurements from the BAQS-Met field study.

The objectives of this study are to quantify biases that can be expected in EC's unified AQ modelling system and to identify where the greatest uncertainties reside so as to guide future model improvements. The specific goals are to (1) generate POA model performance statistics for the entire BAQS-Met period, (2) evaluate model POA bias as a function of POA mass concentration and other indicators of air-mass sources and history (e.g., carbon monoxide, BC, sulfate, back trajectories), (3) identify case study periods to diagnose systematic biases further, and (4) evaluate case-study periods to determine which source regions and PMF factors have large model errors and identify possible causes.

BAQS-Met Study

The 2007 Border Air Quality and Meteorology Study (BAQS-Met) collected state-of-the-art, high-time-resolution OA measurements at two rural sites and one urban site in southwestern Ontario (Brook *et al*., 2011). Southwestern Ontario is home to some of the highest PM concentration levels in Canada. Southwestern Ontario is also one of the most challenging regions for AQ predictions due to the presence of both a wide range of sources and complex mesoscale meteorology associated with the southern Great Lakes of North America.



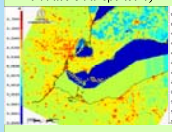
Four AQ super-sites were operated during the study; their locations are shown above. The Windsor site was located in the eastern half of that city just south of a major highway carrying cross-border traffic to the Ambassador Bridge joining Canada and the U.S. The Bear Creek site was located ~60 km east of Detroit-Windsor across Lake St. Clair in a wetland area surrounded by farmland. The Harrow site was ~40 km southeast of Detroit-Windsor and ~5 km north of Lake Erie. Harrow is also located on farmland. The Ridgetown site was the eastern-most site and was also located on farmland just north of the Lake Erie shoreline. This site geometry allowed both the Detroit-Windsor urban plume and regional air masses to be sampled simultaneously at the rural sites on many days. The high-temporal-resolution PM1 OA data considered in this study were collected by AMS instruments at three of these sites: Windsor, Harrow, and Bear Creek. BC and CO measurements were also made at Harrow and Bear Creek.

AQ Modelling System Description and Setup

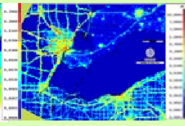
AURAMS consists of three main components: an emissions processing system; a numerical weather prediction model; and an off-line regional chemical transport model (CTM). Version 1.4.0 of the AURAMS CTM, which was used in this study, was driven by version 3.2.2 of the Canadian operational weather forecast model, GEM (Global Environmental Multiscale model) (Côté *et al*., 1998) and emissions files prepared using version 2.3 of the SMOKE emissions processing system (CEP, 2011).

Treatment of emissions

Files of hourly, speciated, gridded anthropogenic emissions were prepared for the three AURAMS CTM grids based on the 2006 Canadian, 2005 U.S., and 1999 Mexican national emissions inventories (NEI) using version 2.3 of the SMOKE emissions processing system. Emissions were separated into four major emissions streams: on-road mobile sources; off-road mobile and area sources; minor-point sources; and major-point sources. To obtain emissions of POA for the four source streams from the bulk PM10 emissions in the inventories (unspeciated emissions of particles with diameter less than 10 µm), four source-stream-specific PM10 chemical speciation profiles were employed. The POA percentages for bulk PM10 were 34% for on-road mobile emissions, 26% for off-road and area emissions, 16% for minor-point emissions and 16% for major-point emissions. These splitting percentages were derived using the 2001 U.S. NEI and PM chemical speciation profiles from the EPA SPECIATE4.2 profile library. The profiles were grouped into the four source streams and then each profile was mass-weighted (based on the summed emissions in the NEI that use each profile divided by the total PM in a particular source stream) to arrive at the representative averaged splitting factors for each source stream. The speciated PM emissions obtained must also be size-disaggregated into the 12 size bins used by the CTM; a weighted average of measured PM size distributions for the different source types was applied for each of the four source streams based on measured size distributions presented in Eldering and Cass (1996). For example, the PM1 size fraction of PM2.5 bulk emissions is 0.96, 0.73, and 0.99 for on-road mobile, off-road and area, and point sources, respectively. In the base case simulation, the POA was treated as chemically inert tracers transported by winds and deposited by wet and dry deposition.



Off-road+Area POA Emissions
July, Monday, 17 UTC



On-road POA Emissions
July, Monday, 17 UTC

Minor Point Source POA
July, Monday, 17 UTC

Major Point Source POA
July, Monday, 17 UTC

Analysis of Ontario and Indiana Emissions Inventory for Summer POA

Table 1. POA emissions inventory for the province of Ontario calculated with the 2006 Canadian $PM_{2.5}$ emissions inventory with the application of SCC-specific PM chemical speciation profiles and sorted by importance into four aggregate source streams (where 1 ton = 0.9072 tonnes).

Class	Ontario 2006 PM Inventory	$PM_{2.5}$ POC (tons C/yr)	Summer Total[1] (tons C/yr)	PM_1 Summer Source Stream Contribution[2] (in brackets)
On-road Mobile	Heavy Duty Diesel Vehicles	**299**	74.8 A	**183*0.96** (5%)
	Light Duty Gasoline Cars	212	53 A	
	Light Duty Gasoline Trucks	180	45 A	
	Heavy Duty Gasoline Vehicles	40.4	10.1 A	
Off-road Mobile	Diesel Agricultural Tractors	**958**	479 B	**602*0.96** (17%)
	Diesel Agricultural Combines	95.6	47.8 B	
	Gasoline Recreational - Snowmobiles	**641**	0 C	
	Diesel Trains	221	55.3 A	
	Diesel Trucks for Construction and Mining	78.8	19.7 A	
Area	Residential Wood Combustion	**12,748**	0 C	**3,450*0.73** (75%)
	Residential Natural Gas	**948**	0 C	
	Residential Petroleum, Kerosene and Oil	21.9	0 C	
	Commercial/Utility Petroleum Oil, Kerosene	25.5	6.38 A	
	Commercial/Utility Natural Gas	227.3	56.83 A	
	Agricultural Production-Crop Drying, Loading	10.6	5.3 B	
	Charcoal Grilling	**2,690**	1345 B	
	Unavailable	**1,822.2**	455.5 A	
	Dust (roads, construction, agricultural)	**2,370**	1,185 B	
	Waste Disposal Burning	269.8	67.4 A	
	Industrial Metal Production	**733**	183 A	
	Industrial Mineral Processes	129.5	32.37 A	
	Structure Fires	7.2	1.8 A	
	Industrial Petroleum Refining	**445**	111 A	
Point	Sum of all major and minor stacks	313	78.3 A	**78.3*0.99** (2%)

Table 2. POA inventory for the state of Indiana calculated with the 2005 U.S. $PM_{2.5}$ emissions inventory with the application of SCC-specific PM chemical speciation profiles and sorted by importance into four aggregate source streams.

Class	Indiana 2006 PM Inventory	$PM_{2.5}$ (Tons POC/yr)	Summer Total[1] (Tons POC/yr)	PM_1 Summer Source Stream Contribution[2] (in brackets)
On-road Mobile	Heavy Duty Diesel Vehicles	N/A	92 D	**196*0.96** (7%)
	Light Duty Gasoline Cars	N/A	51 D	
	Light Duty Gasoline Trucks	N/A	35 D	
	Heavy Duty Gasoline Vehicles	N/A	19 D	
Off-road Mobile	Diesel Agricultural Tractors	N/A	75 D	**355*0.96** (13%)
	Diesel Agricultural Combines	N/A	8.2 D	
	Gasoline Pleasure Craft	N/A	81 D	
	Gasoline Recreational	N/A	75 D	
	Gasoline Lawn and Garden	N/A	76 D	
	Diesel Construction and Mining	N/A	28 D	
	Forklifts	N/A	12 D	
Area	Residential Wood Combustion	**2,249**	0 C	**1,963*0.73** (51%)
	Residential Coal, Oil, Natural gas	26	0 C	
	Residential Petroleum, Kerosene and Oil	96	0 C	
	Combustion Commercial/Institutional (oil, natural gas)	22	5.5 A	
	Combustion Industrial (coal, oil)	**620**	155 A	
	Charcoal Grilling	**1,185**	593B	
	Dust (roads, construction, agricultural)	**1,653**	827 B	
	Waste Disposal Burning	**1,127**	282 A	
Point	Sum of all major and minor stacks (coal-based electric generation units (EGUs), natural gas EGUs, industry natural gas combustion, oil-based commercial boilers, metal production, fiberglass production, coatings, others)	**1,990 non-EGUs + 1,070 EGUs**	765 A (~1/3 by EGUs)	**765*0.99** (29%)

Table 3. Percentage emission source contributions for different model domains (PM emissions are for PM_1 size range) for a July Monday at 17 UTC (12 EST).

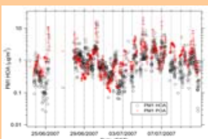
Species	Source Classification	Detroit 42km Grid Cell	15km Domain	2.5 km Domain
PM_1 POA	On-road	15%	5.8%	6.4%
	Off-road+Area	**47%**	**74%**	**74%**
	Minor+Major Point	10%+28%=**38%**	5.5%+14.5%=20%	5.2%+12%=17%
CO	On-road	**47%**	**46%**	**44%**
	Off-road+Area	**49%**	**51%**	**53%**
	Minor+Major Point	1.8%+2.0%=3.8%	1.2%+1.9%=3.1%	0.7%+2.2%=2.9%
PM_1 BC	On-road	**54%**	**27%**	**38%**
	Off-road+Area	**31%**	**63%**	**57%**
	Minor+Major Point	3.9+10.7%=14.6%	2.7+7.3%=10%	2.2%+5.3%=7.5%

Percentages greater than 20% are highlighted for emphasis.

- Calculations with the Ontario PM inventory resulted in surprisingly large POA contributions from area and mobile off-road sources such as food cooking, tractor fuel combustion, and road and soil dust, in addition to the well recognized on-road fuel combustion. In the model, the Detroit/Windsor air shed had POA emission contributions from off-road+area sources at 47%, point sources contributing 38% followed by mobile on-road at 15%.
- An analysis of the PM emissions inventory was performed for upwind Indiana which had large POA sources from area (51%) and point sources (29%) and almost negligible contributions from on-road mobile sources (7%).

Windsor Urban Site

At the urban site, good agreement was observed for the comparison of daytime campaign PM1 POA and HOA mean values: 1.1 µg/m3 vs. 1.2 µg/m3, respectively. However, a POA overprediction was evident on calm nights due to an overly-stable model surface layer. Biases in model POA predictions trended from negative to positive with decreasing HOA values. This trend has several possible explanations, including (1) underweighting of urban locations in particulate matter (PM) spatial surrogate fields, (2) overly-coarse model grid spacing for resolving urban-scale sources, and (3) lack of a dynamic evaporation process during dilution of vehicular POA tail-pipe emissions to urban scales, as has been observed in recent gasoline exhaust studies at high dilution ratios.



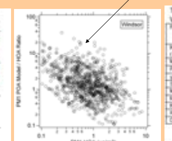


Table 4. AURAMS PM_1 POA model performance statistics with 2.5km grid spacing for the urban Windsor Site (concentrations in µg/m³)

PM1 POA	Windsor Daytime 09:00-14:00 EST	Windsor Evening and Night-Time 18:00-06:00 EST	Windsor Rush-Hour 06:00-09:00 EST 14:00-18:00 EST
Number of points (15 min averages)	194 13 days	532 13 days	264 13 days
Model POA Average	1.1 ± 0.9 µg/m³	2.1 ± 2.8 µg/m³	1.6 ± 1.5 µg/m³
PMF HOA Average	1.2 ± 1.0 µg/m³	1.0 ± 1.0 µg/m³	1.4 ± 2.1 µg/m³
y-intercept (model value)	0.65 µg/m³	0.85 µg/m³	1.4 µg/m³
Slope	0.39	1.2	0.16
Mean Bias (model-meas)	[illegible]	1.0 µg/m³	0.16 µg/m³
Normalized Mean Bias	[illegible]	+100 %	+11 %
Normalized Mean Error	17 %	140 %	91%
RMSE	0.31 µg/m³	2.7 µg/m³	2.3 µg/m³
Correlation, R	0.43	0.41	0.28

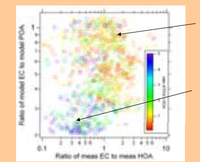
Table 5. AURAMS CO model performance statistics with 2.5km grid spacing for the urban Windsor Site (mixing ratios in ppbv)

CO	Windsor Daytime 09:00-14:00 EST	Windsor Evening and Night-Time 18:00-06:00 EST	Windsor Rush-Hour 06:00-09:00 EST 14:00-18:00 EST
Number of points (15 min averages)	189 points 13 days	535 13 days	262 13 days
Model CO Average	592 ± 405 ppbv	566 ± 343 ppbv	665 ± 538
Meas CO Average	253 ± 145 ppbv	265 ± 219 ppbv	301 ± 319
y-intercept (model value)	420 ± 58 ppbv	346 ± 21 ppbv	474 ± 43 ppbv
Slope	0.68 ± 0.20	0.77 ± 0.07	0.64 ± 0.10
Mean Bias (model-meas)	+339 ppbv	+261 ppbv	+364 ppbv
Normalized Mean Bias	+134 %	+99 %	+121 %
Normalized Mean Error	142 %	107 %	137 %
RMSE	520 ppbv	455 ppbv	628 ppbv
Correlation, R	0.24	0.43	0.38

Table 6. AURAMS PM2.5 BC model performance statistics with 2.5km grid spacing for the urban Windsor Site (concentrations in µg/m³)

PM2.5 BC	Windsor Daytime 09:00-14:00 EST	Windsor Evening and Night-Time 18:00-06:00 EST	Windsor Rush-Hour 06:00-09:00 EST 14:00-18:00 EST
Number of points (15 min averages)	190 13 days	535 13 days	243 13 days
Model BC Average	0.70 ± 0.60 µg/m³	0.63 ± 0.62 µg/m³	0.88 ± 0.88
Meas BC Average	1.3 ± 1.3 µg/m³	1.4 ± 1.5 µg/m³	1.3 ± 1.4
y-intercept (model value)	0.45 ± 0.06 µg/m³	0.50 ± 0.04 µg/m³	0.36 ± 0.06
Slope	0.19 ± 0.03	0.24 ± 0.02	0.41 ± 0.03
Mean Bias (model-meas)	-0.55 µg/m³	-0.77 µg/m³	-0.38
Normalized Mean Bias	-46 %	-47 %	-30 %
Normalized Mean Error	59 %	61 %	55 %
RMSE	1.38 µg/m³	1.47 µg/m³	1.12
Correlation, R	0.42	0.43	0.64

Furthermore, a trend in POA bias was observed at the urban site as a function of the BC/HOA ratio, suggesting a possible association of POA underprediction for diesel combustion sources. For several time periods, POA overprediction was also observed for sulphate-rich plumes, suggesting that our model POA fractions for the PM2.5 chemical speciation profiles may be too high for these point sources.



Model POA underprediction at high EC/POA ratios (indicative of diesel emissions)

Model POA overprediction at low EC/POA ratios (indicative of area sources)

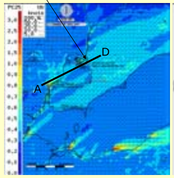
Bear Creek and Harrow Rural Sites

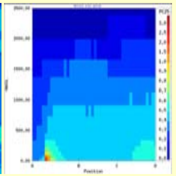
At the rural Harrow site, significant underpredictions in PM1 POA were observed compared to observed HOA and were associated, based on back-trajectory analysis, with (1) transport from the Detroit/Windsor urban complex, (2) longer-range transport from the U.S. Midwest, and (3) biomass burning. Daytime CO concentrations were significantly overpredicted at Windsor but were unbiased at Harrow.

At Bear Creek, POA underprediction was observed for the case of outflow from Detroit-Windsor. In general, there was good agreement at Bear Creek for periods when clean background air flowed into the study region from the northwest. Measurement-derived HOA was considerably lower at Bear Creek than Windsor or Harrow.

Table 8. Detroit-Windsor Urban-Influenced Air Masses

Date (EST) PM1	Model POA HOA % NMB RMSE	HOA/OA	Winds	Meteorology	Model BC Meas BC % NMB RMSE	Model CO Meas CO % NMB RMSE	Model SO4 Meas SO4 % NMB RMSE	Source Description
Harrow June 21 10-15	0.26 ± 0.06 1.4 ± 0.4 -81 % 1.24	17%	NNW moderate	Winds and PBL heights predicted well	0.15 ± 0.03 0.94 ± 0.17 -84 % 0.80	165 ± 15 199 ± 31 -17 % 41	1.86 1.31 42 % 0.68	"Organic" factor, south Detroit origin
Bear Creek July 8 10-14:20	0.32±0.10 0.96±0.23 -67 % 0.66	6.0%	SW moderate	Winds and PBL heights predicted well	0.15 0.53 -71 % 0.38	194 245 -21 % 53	2.3 13.0 83% 10.6	South Detroit and Windsor origin

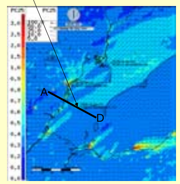


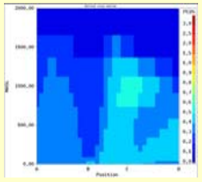


POA underprediction for flow from Detroit/Windsor

Table 9. Transport from Southwest

Date (EST) PM1	Model POA HOA % NMB RMSE	HOA/OA	Winds	Meteorology	Model BC Meas BC % NMB RMSE	Model CO Meas CO % NMB RMSE	Model SO4 Meas SO4 % NMB RMSE	PMF Description
Harrow July 8 10-14:50	0.33 ± 0.12 1.2 ± 0.2 -73 % 0.95	16%	SW Moderate	Well predicted, evidence for polluted layer aloft	0.16 0.67 -76 % 0.52	186 248 -25 % 64	3.2 5.2 -39 % 2.5	"Transport" US Midwest
Harrow June 27 10:30-14	0.47 ± 0.10 0.40 ± 0.14 17 % 0.099	21%	SW Moderate	Well predicted, evidence for polluted layer aloft	0.16 0.70 -77 % 0.54	207 231 -11 % 35	7.4 4.5 64 % 3.0	"Transport" US Midwest





- POA underprediction for periods of longer range transport from southwest.
- Evidence for POA overprediction in modeled sulfate rich plumes (June 27 case) although exact location of plume is difficult to model for such a narrow feature.

Table 10. Biomass Influence

Date (EST)	Model POA HOA % NMB RMSE	HOA/OA	Winds	Model BC Meas BC % NMB RMSE	Model CO Meas CO % NMB RMSE	Model SO4 Meas SO4 % NMB RMSE	PMF Description
Harrow July 6 9-11:30	0.20 ± 0.04 3.6 ± 0.2 -94 % 3.4	20%	SE Light	0.15 0.83 -82 % 0.68	201 296 -32 % 104	0.59 1.2 -50 % 0.60	"Biomass Burning"
Harrow July 7 9-14:50	0.48 ± 0.26 1.7 ± 0.3 -71 % 1.22	16%	WSW Moderate	0.12 0.82 -86 % 0.70	205 182 13 % 69	5.4 1.1 403 % 5.8	"Biomass Burning"

POA underprediction for periods of biomass burning influence.

Table 5. AURAMS PM1 POA model performance statistics with 2.5km grid spacing for the sub-urban Harrow and Bear Creek sites (concentrations in µg/m³)

PM1 POA	Harrow Daytime 09:00-18:00 EST	Harrow Night-Time 18:00-09:00 EST	Bear Creek Daytime 09:00-18:00 EST	Bear Creek Night-Time 18:00-09:00 EST
Number of points	1826 (15 min avges) 23 days	3480 23 days	297 (3 min avges) 12 days	538 12 days
Model POA Average	0.3±0.2 µg/m³	0.4±0.3 µg/m³	0.24±0.21 µg/m³	0.36±0.23 µg/m³
PMF HOA Average	0.8±0.7 µg/m³	0.9±0.9 µg/m³	0.31±0.39 µg/m³	0.32±0.31 µg/m³
y-intercept (model value)	0.246 µg/m³	0.336 µg/m³	0.14 µg/m³	0.29 µg/m³
Slope	0.058	0.116	0.32	0.21
Mean Bias	-0.48 µg/m³	-0.45 µg/m³	-0.07 µg/m³	0.039 µg/m³
Normalized Mean Bias	-62 %	-51 %	-23 %	+12 %
Normalized Mean Error	69 %	69 %	63 %	73 %
RMSE	0.87 µg/m³	0.93 µg/m³	0.32 µg/m³	0.34 µg/m³
Correlation, R	0.23	0.32	0.58	0.28

Table 6. AURAMS CO model performance statistics with 2.5km grid spacing for the sub-urban Harrow and Bear Creek sites (concentrations in ppbv)

CO	Harrow Daytime 09:00-18:00 EST	Harrow Night-Time 18:00-09:00 EST	Bear Creek Daytime 09:00-18:00 EST	Bear Creek Night-Time 18:00-09:00 EST
Number of points	1514 (15 min avges) 23 days	3139 23 days	227 (3 min avges) 12 days	477 12 days
Model CO Average	180 ± 49 ppbv	192 ± 62 ppbv	172 ± 44 ppbv	172 ± 44 ppbv
Meas CO Average	197 ± 87 ppbv	204 ± 82 ppbv	199 ± 97ppbv	210 ± 93 ppbv
y-intercept (model value)	129 ± 3 ppbv	111 ± 3 ppbv	129 ± 6 ppbv	103 ± 4
Slope	0.26 ± 0.01	0.40 ± 0.01	0.22 ± 0.03	0.33 ± 0.02
Mean Bias (model-meas)	-17 ppbv	-12 ppbv	-26 ppbv	-38 ppbv
Normalized Mean Bias	+2.3 %	-5.8 %	-13 %	-11 %
Normalized Mean Error	28 %	25 %	26 %	24 %
RMSE	79 ppbv	73 ppbv	89 ppbv	80 ppbv
Correlation, R	0.46	0.53	0.49	0.69

Table 7. AURAMS PM2.5 BC model performance statistics with 2.5km grid spacing for the sub-urban Harrow and Bear Creek sites (concentrations µg/m³)

PM2.5 BC	Harrow Daytime 09:00-18:00 EST	Harrow Night-Time 18:00-09:00 EST	Bear Creek Daytime 09:00-18:00 EST	Bear Creek Night-Time 18:00-09:00 EST
Number of points	239 (105 min avges) 23 days	139 23 days	295 (3 min avges) 12 days	532 12 days
Model BC Average	0.16 ± 0.11 µg/m³	0.19 ± 0.12 µg/m³	0.11 ± 0.09 µg/m³	0.15 ± 0.09 µg/m³
Meas BC Average	0.60 ± 0.26 µg/m³	0.61 ± 0.27 µg/m³	0.38 ± 0.26 µg/m³	0.46 ± 0.32 µg/m³
y-intercept (model value)	0.070 ± 0.016 µg/m³	0.099 ± 0.02 µg/m³	0.018 ± 0.006 µg/m³	0.090 ± 0.006 µg/m³
Slope	0.15 ± 0.02	0.15 ± 0.04	0.24 ± 0.01	0.14 ± 0.01
Mean Bias (model-meas)	-0.44 µg/m³	-0.42 µg/m³	-0.27 µg/m³	-0.31 µg/m³
Normalized Mean Bias	-73 %	-67 %	-71 %	-67 %
Normalized Mean Error	73 %	69 %	71 %	68 %
RMSE	0.50 µg/m³	0.49 µg/m³	0.34 µg/m³	0.42 µg/m³
Correlation, R	0.39	0.33	0.72	0.46

Conclusions

The POA agreement in Windsor, POA underprediction at Harrow, CO overprediction at Windsor and CO agreement at Harrow are contrasting and revealing biases. In addition, the more consistent underprediction of PM2.5 BC for all sites is another observation to consider in evaluating model performance. Collectively, these biases provide support for a hypothesis that combines a current underweighting of PM spatial surrogate fields for urban locations (especially food cooking and dust spatial surrogates) with insufficient model vertical mixing for sources close to the urban measurement sites (especially at night). Diesel emission underestimates may be the cause of the BC underpredictions and POA underpredictions at high BC/POA ratios. The magnitude of the area POA emissions sources in the U.S. and Canadian inventories (e.g., food cooking, road and soil dust, waste disposal burning) suggests that more effort should be placed at reducing uncertainties in these sectors, especially applied spatial and temporal surrogate fields (shape files). POA in episodes of biomass burning were also significantly underestimated sometimes with a NMB as high as -94%. Future efforts should also be placed on incorporating biomass burning emissions and chemistry into forecast models.

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