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5755 NORTH POINT PARKWAY'SUITE 265'ALPHARETTA, GA 30022

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Prepared by: ENVIRON International Corporation 773 San Marin Drive, Suite 2115 Novato, California, 94945 P-415-899-0700 F-415-899-0707

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Till E. Stoeckenius¹*, Christian Hogrefe², Justin Zagunis¹

¹ENVIRON International Corp., Novato, CA, USA ²U.S. Environmental Protection Agency, Research Triangle Park, NC, USA

*Corresponding Author: Till Stoeckenius 773 San Marin Dr., Suite 2115 Novato CA, 94998 USA T: 415.899.0709 F: 415.899.0707 till@environcorp.com

Abstract

Several participants in Phase 2 of the Air Quality Model Evaluation International Initiative (AQMEII-2) who are applying coupled models to the North American domain are comparing model results for two years: 2006 and 2010. While a key difference of interest between these two years from a modeling perspective are the large reductions in emissions of NO_x (21%) and SO_2 (36%) which occurred mostly in the eastern U.S., meteorological conditions also differed significantly between these two years and these differences both confound the impact of emission reductions on ambient air quality and provide an opportunity to examine how models respond to changing meteorology. Observed summer ozone levels in many portions of the Northeast and Midwest were largely unchanged in 2010 despite reductions in precursor emissions. The authors have previously demonstrated that normalization of the ozone trend to account for differences in meteorological conditions, including warmer summer temperatures in 2010, shows that the emission reductions would have resulted in lower ozone levels at these locations if not for the countervailing influence of meteorological conditions. We present here an evaluation of the ability of models to accurately account for the impact of the 2006 – 2010 emission reductions on air quality using a synoptic weather pattern classification methodology designed to remove the influence of meteorology on the 2006 – 2010 air quality trends. The synoptic classification consists of matching groups of days between the two years on the basis of similarities in sea-level pressure patterns. Results show that the models exhibit some skill in replicating observed ozone trends between 2006 and 2010 when results are stratified by synoptic patterns. However, the pattern classification, which is based solely on sea-level pressure, does not account for other key meteorological factors influencing ozone concentration differences between 2006 and 2010 and thus does not provide for a true evaluation of the model's ability to replicate the underlying (emissions driven) ozone trend.

Keywords

AQMEII, air quality - meteorology interactions, emission trends, ozone trends, synoptic types

1. Introduction

Development of accurate models for simulating atmospheric trace gas composition is a key component of an effective air quality management program. The Air Quality Model Evaluation International Initiative (AQMEII) was developed to fulfill the need to both better understand uncertainties in regional-scale model predictions and to foster continued model improvement by providing a collaborative, cross-border platform for model development and evaluation in North America and Europe (Galmarini and Rao, 2011).

Phase 2 of AQMEII (AQMEII-2) focused on evaluation of online-coupled models capable of simulating feedbacks between atmospheric trace gas composition and meteorological conditions. AQMEII-2 included the option for participants to evaluate model performance for two individual calendar years: 2006 and 2010. As discussed by Stoeckenius et al. (2014), modeling of the North American domain by AQMEII-2 participants used emission inventories for 2006 and 2010 derived from U.S. EPA's 2008 emissions modeling platform with year-specific adjustments to activity levels and emission factors for on-road and off-road mobile sources, year-specific continuous emissions monitoring systems (CEMS) data for the large point sources where CEMS data were available, and year-specific fire emissions estimates. Updated estimates of Canadian emissions and Mexican emissions developed for 2006 were used without adjustment in the 2010 inventory (Pouliot, et al., 2014). Thus the only differences between the 2006 and 2010 modeling inventories are changes to mobile sources, CEMS point sources and fire emissions in the U.S.. Biogenic and wind-blown dust emissions were calculated on-line by each modeling group participating in AQMEII-2; the resulting emissions are based on varying methodologies developed by each group and summaries are not available.

As pointed out by Pouliot et al. (2014) and Stoeckenius et al. (2014), emissions from anthropogenic sources were reduced substantially during the interval between 2006 and 2010. Significant reductions in emissions from electric power generation occurred between 2006 and 2010 in the eastern U.S. as reflected in summaries of total U.S. sub-regional emissions (Fig. 1; sub-region definitions in Fig. 2). Seasonal reductions of 31% to 52% occurred in SO₂ and 22% to 15% in NO_x in the Midwest, Northeast, and Southeast (Table 1). Comparable NO_x reductions occurred in other subregions except for a smaller (11%) reduction in the South-Central sub-region. NO_x reductions varied seasonally for large source with continuous emissions monitors (mostly electric utilities) as shown in Table 2. Utilities in the Midwest and Northeast already had significant controls in effect during the summer season by 2006 and only minor additional reductions occurred by 2010 whereas large reductions occurred year-round between 2006 and 2010 in the Southeast where summer season controls had not previously been widely applied. Nevertheless, total NO_x emission reductions were similar in winter and summer in the Northeast as the seasonal difference in the utility emission reductions is diluted by large but seasonally invariant reductions in mobile sources and the (assumed) 0% change in area source emissions. PM and anthropogenic VOC emissions were strongly elevated in the summer of 2006 in the western sub-regions due to major wildfires: in the 13 western states, 6.7 million acres burned in 2006 as compared to 1.5 million acres in 2010 (NIFC, 2014). Apart from the influence of fires, PM_{2.5} emissions showed little change overall. Small reductions occurred in on-road and off-road mobile source VOC emissions. These data suggest that, apart from the influences of differences in boundary conditions and meteorology or chemical regime changes in secondary PM and O_3 formation, significant reductions in SO₂, sulfate and nitrate PM, and O_3 concentrations should have occurred in 2010 relative to 2006.



Fig. 1. Winter (Win; December - February) and summer (Sum; June - August) daily average emissions for 2006 and 2010 used in AQMEII-2 simulations for North America (biogenic VOC and NO_x emissions are not included).



Fig. 2. U.S. sub-regions used to summarize emissions and air quality.

	СО	NH ₃	NO _x	PM_{10}	PM _{2.5}	SO_2	VOC
Midwest	-20%	1%	-25%	5%	7%	-36%	-7%
Northeast	-25%	-1%	-22%	-1%	-3%	-44%	-11%
Plains	-6%	1%	-18%	-5%	10%	-17%	3%
South-Central	-15%	1%	-11%	1%	4%	-11%	-2%
Southeast	-23%	0%	-25%	0%	0%	-52%	-9%
West	-50%	-16%	-19%	-22%	-46%	-26%	-36%
West Coast	-37%	-13%	-25%	-36%	-50%	-25%	-33%
TOTAL:	- 2 6%	-3%	-21%	-9%	-14%	-37%	-13%

Table 1. Fractional changes in annual U.S. emissions [(2010 - 2006)/2006] by sub-region.

Table 2.	Reductions i	n NO _v	emissions	by 20	10 relative	e to 2006	levels.
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NO _x	CEM Poir	nt Sources	All Sources		
% change	Winter	Summer	Winter	Summer	
Midwest	-54%	-7%	-30%	-16%	
Northeast	-37%	-6%	-21%	-19%	
Plains	-35%	-29%	-20%	-17%	
South-Central	-8%	-13%	-11%	-11%	
Southeast	-55%	-36%	-26%	-22%	
West	-23%	-26%	-16%	-22%	
West Coast	26%	1%	-22%	-27%	

Analyses of data from U.S. monitoring networks by Stoeckenius et al. (2014) showed that summer mean daily maximum 8-hour average ozone (MDA8O3) concentrations were generally lower in 2010 than in 2006 except for the Northeast and upper Midwest where there were increases at many sites along the Washington to Boston urban corridor and in the Chicago area and near zero (+/- 2 ppb) changes away from the major urban areas (Fig. 3).

AQMEII-2 model simulations provide an opportunity to evaluate the ability of coupled models to accurately simulate the impact of the 2006 – 2010 emission reductions on air quality. However, trace gas concentrations are a function of both emissions and meteorological conditions and meteorological variations confound the attribution of 2006-2010 air quality differences to changes in anthropogenic emissions. This is particularly true for secondary pollutants such as ozone and PM which are formed via chemical reactions involving directly emitted (primary) pollutants, the reaction rates of which are sensitive to meteorological parameters. An analysis of differing meteorological influences on ozone levels in the eastern U.S. presented by Stoeckenius et al. (2014) showed that conditions in 2010 were more favorable for ozone production than in 2006 in the Midwest and Northeast, consistent with the warmer temperatures observed in the eastern U.S. in 2010 as compared to 2006. When ozone levels were adjusted to take meteorological influences into account using the statistical ozone trend adjustment methodology developed by Camalier et al. (2007), 2010 ozone levels were found to be lower in nearly all urban areas throughout the U.S. in 2010 compared to 2006 (see Stoeckenius et al., Fig. 15). This result is consistent with the lower NO_x and VOC emissions noted above.



Fig. 3. Difference (2010 – 2006) in summer (June – August) mean MDA8O3 at all U.S. monitoring sites.

Inter-annual variations in meteorological conditions may confound evaluations of the ability of photochemical models to replicate observed ozone trends. For example, simply comparing observed with predicted ozone differences between the 2010 and 2006 ozone seasons without controlling for differences in meteorological conditions may not provide an accurate indication of the model's ability to predict the impact of emission changes on ozone levels. One way around this problem is to limit comparisons of observed with predicted air quality between the two years to days characterized by similar meteorological conditions. While the trend adjustment methodology of Camalier et al. (2007) accounts for combined influences of multiple meteorological variables (temperature, humidity, wind speed, etc.), it does not easily lend itself to a straightforward classification of days into groups defined by similarities in meteorological conditions. We therefore present here an evaluation of model performance in replicating observed 2006-2010 ozone trends based on a synoptic meteorological pattern classification methodology developed by Hogrefe et al. (2014b). Comparisons of observed meteorological and air quality conditions with model predictions are not included in this paper but are the subject of several companion papers (Campbell et al., 2014; Hogrefe et al., 2014a; Wang et al., 2014).

2. Data and Methods

2.1 Air Quality Observations and Model Predictions

Hourly ozone observations and paired model predictions from all available surface monitoring sites in the U.S., Mexico and Canada falling within the AQMEII North America modeling domain for 2006 and 2010 were extracted from the ENSEMBLE system (Im et al., 2014). These data were processed into daily maximum running 8-hour average ozone concentrations (MDA8O3). EPA data completeness criteria (U.S. EPA, 1998) were used to exclude days with unrepresentative MDA8O3 due to missing observations.

2.2 Meteorology

Days during 2006 and 2010 were classified by synoptic conditions based on correlation patterns in sea level pressure (SLP) fields developed by Hogrefe et al. (2014b). Given the size of the AQMEII-2 North American modeling domain and the heterogeneous nature of weather conditions across the

full domain, Hogrefe et al. divided the domain into an eastern and a western portion along approximately W105 degrees longitude and developed separate sets of synoptic patterns for the eastern and western halves. Since the largest regional ozone precursor emission reductions between 2006 and 2010 occurred in the eastern U.S., we focus here on results for the eastern half of the domain. Of the 14 synoptic patterns identified by Hogrefe et al. in the eastern portion of the modeling domain, Patterns 1-8 and 14 occurred on at least four days in each year during the May – September warm season and were considered for inclusion in the analysis. Pattern #8 was subsequently excluded due to missing monitoring data at some sites. In addition, the 7% of days exhibited sea-level pressure patterns that could not be classified under the typing scheme were also excluded from further analysis as they do not represent a distinct synoptic pattern. Patterns #1 and #3 were by far the most common in both 2006 and 2010 (Fig. 4) with only small differences in the pattern frequencies between the two years.





Surface pressure maps for the individual days most correlated with these two synoptic patterns are shown in Fig. 5. Both warm seasons had generally similar distributions of synoptic Patterns #1 and #3 which together accounted for between 54 and 46% of all summer days. Both of these patterns include extension of the semi-permanent Bermuda High into the southeastern U.S. Pattern #1 occurs in both winter and summer but Pattern #3 is predominantly a summer pattern. The higher summer temperatures in 2010 relative to 2006 noted by Stoeckenius et al. do not appear to be associated with a significant shift in synoptic patterns, although there was a slightly higher frequency of Pattern #3 in 2010 and this pattern is associated with more intense warm surface temperature anomalies in the Midwest and Mid-Atlantic portions of the eastern U.S (Fig. 6).



Fig. 5. Surface pressure fields for days most representative of synoptic Pattern #1 (left) and #3 (right) during 2006 and 2010.



Fig. 6. Mean surface temperature anomalies based on NCEP/NCAR 40-year Reanalysis data (Kalney, et al., 1996) for Pattern #1 (left) and #3 (right) for summer days during 2006 and 2010 (image provided by the NOAA/ESRL Physical Sciences Division, Boulder Colorado from their Web site at http://www.esrl.noaa.gov/psd/).

3. Results and Discussion

Ozone "trend" statistics are defined here as the May – September mean MDA8O3 at a monitoring site in 2010 minus the corresponding mean in 2006 where means are calculated for all days belonging to a specific synoptic pattern. Observed ozone trends were compared with predicted trends where the predicted trends are calculated from model results as reported to ENSEMBLE by the four model simulations available in the ENSEMBLE system for the North American domain for both 2006 and 2010: Environment Canada GEM-MACH simulation without feedbacks (CA2), U.S. EPA WRF-CMAQ simulation (US2), Environment Canada GEM-MACH simulation with feedbacks (CA2f), and North Carolina State University's WRF-Chem simulation (US8). Evaluation of the ability of these models to reproduce observed meteorological and air quality conditions are reported by Im et al. (2014); we focus here specifically on evaluations of the model's ability to reproduce observed 2006 – 2010 ozone trends. Our analysis is focused on the eastern U.S. as that is where the largest regional

emission reductions occurred and specifically on the Midwest and Northeast sub-regions (Fig. 2) where inter-annual variations in meteorological conditions are most likely to influence ozone trends. Results are presented for the warm season (May – September) period when ozone production is most active.

Within each sub-region, observed trends vary widely by monitoring site and by synoptic pattern. Mean and median trends in the Northeast are near zero with 50% of the trends falling within ±5 ppb whereas trends in the Midwest are more homogeneous and tend to the negative (Fig. 7). For purposes of comparison, Fig. 7 also shows mean and median trends averaged over all warm season days without regard to synoptic pattern. The latter trends are 0.9 to 1.1 ppb more positive (less negative) in the Northeast (Midwest) than the composite trends based on stratification by synoptic pattern, suggesting that the stratification has accounted for some of the meteorological differences between 2006 and 2010 that resulted in little to no observed ozone reduction in 2010 despite reductions in anthropogenic precursor emissions. The lack of more significant negative ozone trends in the Northeast when stratified by synoptic pattern is in contrast to the relatively large underlying (emissions driven) negative trends revealed by applying the statistical meteorological trend adjustment approach developed by Camalier et al. (2007) to the 2006 and 2010 ozone data as described by Stoeckenius et al. (2014). Thus the synoptic patterns, which are solely based on similarities in surface pressure patterns, do not effectively account for other key meteorological factors such as temperature that are responsible for the tendency towards increased ozone in 2010 despite lower precursor emissions. In particular, as was pointed out in Sec. 2.2, the higher summer temperatures in 2010 relative to 2006 noted by Stoeckenius et al. which effectively masked the underlying ozone trend in the Northeast and portions of the Midwest are not associated with any significant shift in the relative frequencies of occurrence of the synoptic patterns.



Fig. 7. Distributions (over all monitoring sites and synoptic patterns) of observed ozone trends in May-September mean MDA8O3 stratified by synoptic pattern in the Northeast and Midwest subregions.

Standard model performance statistics as defined by U.S. EPA (2014) comparing observed with model predicted ozone trends were computed for each individual synoptic pattern (i.e., matching days by pattern between 2010 and 2006). Performance statistics included spatial correlation coefficient,

mean bias, mean error, root mean square error, normalized mean bias, and normalized mean error¹. The performance statistics were then aggregated over all patterns with sufficient data (as described in Sec. 2.1) to provide an overall summary of the model's ability to reproduce observed trends when matched by synoptic pattern. Days in the "unclassified" pattern were not included in the aggregation. There results were compared with performance statistics for trends computed over all days without matching by pattern as shown in Fig. 8 and Tables 3 and 4. Note that results aggregated over all patterns are based on equal weighting of each pattern regardless of the number of days assigned to the pattern. As expected, matching by synoptic pattern greatly increases spatial correlations and reduces the magnitude of the model bias (except in the case of the US6 model in the Northeast) although mean error statistics increase. Increases in the error statistics are likely the result of the fact that the majority of days are assigned to the same two or three patterns in both years as was shown in Fig. 4, leaving the majority of patterns with just a handful of days in each year and resulting in an increased composite variance. Examination of results for individual synoptic patterns (not shown) indicates wide variation in observed trends and in model performance in reproducing the trends. In particular, the large negative bias in the Northeast for US6 was found to be associated with large negative biases under Patterns #5, #6 and to a lesser extent Pattern #7, which were not matched by similar biases in the other models. We speculate that the significantly different performance of US6 for days assigned to these patterns may be associated with significant differences in predicted meteorology but additional detailed investigation of model results will be needed to confirm this. Nevertheless, the overall results presented here indicate all models demonstrate some skill in replicating spatial patterns in trends when matching by synoptic pattern. The US6 model exhibits the highest correlation with observed trends in both sub-regions and has the lowest bias magnitude in the Midwest. The US8 model shows a strong negative bias relative to the other models, resulting in prediction of much greater than observed negative trends, especially in the Midwest. Mean errors are roughly of similar magnitude in both sub-regions but normalized mean errors are much larger in the Northeast where the observed trend is small (Fig. 7).

¹ Since trends can be either positive or negative, the calculation of normalized mean bias and normalized mean error was modified to use the absolute value of the mean observed trend as the normalization factor.



Fig. 8. Scatter plots of observed vs. predicted ozone trends at individual monitoring sites in the Northeast (top) and Midwest (bottom) sub-regions for averages matched by synoptic pattern (left) and averages without matching (right).

Table 3.	Model	performance	statistics for	or ozone	trends,	Northeast	sub-region.
					,		

		CA2	US6	CA2f	US8
R	Matched	0.58	0.75	0.58	0.64
	Unmatched	0.23	0.41	0.23	0.46
MB	Matched	-0.09	-2.07	0.10	-3.83
	Unmatched	-1.10	-1.57	-0.78	-5.01
ME	Matched	4.98	4.57	5.06	6.00
	Unmatched	2.23	2.29	2.23	5.02
RMSE	Matched	6.51	5.73	6.53	7.13
	Unmatched	2.81	2.76	2.72	5.46
NMB(%)	Matched	-9	-201	10	-372
	Unmatched	-98	-140	-69	-447
NME(%)	Matched	484	444	492	583
	Unmatched	199	204	199	448

		CA2	US6	CA2f	US8
R	Matched	0.42	0.58	0.44	0.45
	Unmatched	0.35	0.27	0.30	-0.10
MB	Matched	0.43	-0.08	0.74	-1.48
	Unmatched	1.35	-0.46	1.63	-2.58
ME	Matched	4.46	3.77	4.40	4.48
	Unmatched	2.41	2.24	2.57	3.52
RMSE	Matched	5.56	4.78	5.51	5.47
	Unmatched	3.28	3.07	3.47	4.22
NMB(%)	Matched	18	-3	31	-61
	Unmatched	133	-45	160	-254
NME(%)	Matched	183	155	181	184
	Unmatched	237	220	252	346

Table 4. Model performance statistics for ozone trends, Midwest sub-region.

4. Summary and Conclusions

Coupled models applied to the North American domain under the Phase 2 of the AQMEII exhibit some skill in replicating observed ozone trends between 2006 and 2010 when results are stratified by synoptic patterns defined on the basis of similarities in surface pressure fields. Model bias and spatial correlation indicate better composite performance for trends stratified by synoptic pattern as compared to predictions of mean trends in the absence of such stratification. On the other hand, composite error statistics (ME, RMSE, NME) increase in magnitude when trends are stratified by pattern, most likely due to the relatively small number of days belonging to the majority of individual strata in each year. Performance in replicating trends varies from one model to the next with the US6 model exhibiting the strongest correlations, smallest errors, and (except in the Northeast) the smallest bias magnitudes.

While stratification by synoptic sea level pressure pattern somewhat limits the confounding influence of meteorological differences between 2006 and 2010 and shows that model performance for ozone trends is better than what one would conclude in the absence of matching by synoptic pattern, examination of observed trends stratified by synoptic pattern shows that the synoptic pattern classification does not account for other key meteorological factors influencing ozone concentration differences between 2006 and 2010 and thus does not fully reveal the underlying (emissions driven) ozone trend. This in turn means that the ability of models to accurately replicate the impact of 2006 – 2010 emission reductions on ozone levels is not effectively isolated by the use of the synoptic pattern stratification which is based solely on sea-level pressure. Future studies should consider using a refined, multivariate synoptic classification methodology so as to better capture the key meteorological influences on ozone within the classification system.

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