Evaluation of near surface ozone and particulate matter in air quality simulations driven by dynamically downscaled historical meteorological fields

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Abstract

In this study, techniques typically used for future air quality projections are applied to a historical 11-year period to assess the performance of the modeling system when the driving meteorological conditions are obtained using dynamical downscaling of coarse-scale fields without correcting toward higher-resolution observations. The Weather Research and Forecasting model and the Community Multiscale Air Quality model are used to simulate regional climate and air quality over the contiguous United States for 2000–2010. The air quality simulations for that historical period are then compared to observations from four national networks. Comparisons are drawn between defined performance metrics and other published modeling results for predicted ozone, fine particulate matter, and speciated fine particulate matter. The results indicate that the historical air quality simulations driven by dynamically downscaled meteorology are typically within defined modeling performance benchmarks and are consistent with results from other published modeling studies using finer-resolution meteorology. This indicates that the regional climate and air quality modeling framework utilized here does not introduce substantial bias, which provides confidence in the method's use for future air quality projections.

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1. INTRODUCTION

Air quality is strongly influenced by meteorology, and it is well established that a changing climate will affect future ambient air quality (e.g., Jacob and Winner, 2009; Weaver et al., 2009; Fiore et al., 2015). Many studies that have examined the effects of climate change on air quality have concluded that surface ozone (O_3) levels will likely increase in localized areas due to the projected changes in meteorological conditions (Leung and Gustafson, 2005; Nolte et al., 2008; Katragkou et al., 2011; Kelly et al., 2012; Trail et al., 2014; Fann et al., 2015). This has potential implications for air quality management plans, because the “climate penalty” (Wu et al., 2008) may offset some of the improvements in ambient air quality that would otherwise occur as emissions controls are implemented (Fann et al., 2016), with adverse consequences for human health. One other potential effect of climate change on air quality is a regionally dependent lengthening of the ozone season, historically classified as May through September in most of the United States (e.g., Fiore et al., 2002; Nolte et al., 2008; Weaver et al., 2009; Cooper et al., 2014; Clifton et al., 2014). Accordingly, it is critical to develop a regional modeling framework that can credibly project air quality changes resulting from future climate scenarios.

For most historical (“retrospective”) regional air quality studies using Eulerian models, meteorological conditions are obtained from a comparably scaled meteorological model. Furthermore, the meteorology developed for those retrospective air quality modeling applications assimilates observations where available to achieve the best possible representation of the three-dimensional evolution of the meteorological conditions over the time period of interest (Otte, 2008a,b). Several “dynamic evaluation” air quality modeling studies have been conducted in which the focus is not the air quality model’s predictions in comparison to observations, but rather on the model’s response to changing emis-

sions or meteorology (e.g., Gilliland et al., 2008; Foley et al., 2015). Hogrefe et al. (2011) conducted an 18-year simulation over the Northeast United States, examining the model’s ability to capture the interannual variability in O_3 levels due to meteorology as well as trends due to changes in emissions. In all of these studies, the meteorology was developed by assimilating high-resolution observational data.

When starting from a coarse-scale global modeling framework to simulate regional-scale air quality, dynamical downscaling can be used (Giorgi and Meleux, 2007). In dynamical downscaling, the coarse global data provide input to the regional model through the lateral boundaries and sometimes within the interior domain through “nudging”. Interior nudging maintains consistency in the weather patterns between the global and regional models and can reduce errors in the regional model (von Storch et al., 2000; Miguez-Macho et al., 2004; Bowden et al., 2012). Unlike traditional retrospective meteorological modeling, no fine-scale observations are assimilated when downscaling. It is challenging, however, to evaluate the effects of using meteorology downscaled from a global climate model (GCM) for simulating air quality. Fields from a GCM are representative of the climate during that era, e.g., “climate circa 2000,” but cannot accurately simulate the weather of the day. It is therefore necessary to model several years to account for interannual variability of meteorological conditions. Also, air pollutant emissions have been changing dramatically over the past few decades, and it is necessary to consider these changes when comparing to measurements taken over multiple years.

In order to evaluate the validity of using downscaled fields to drive future air quality projections, it is important to test the downscaling method using historical coarse-scale global fields without correcting toward higher-resolution meteorological observations. The downscaled meteorology may then be used to drive a chemical transport model, and the results can be compared to historical observations of air quality to ascertain whether future air quality projections based on downscaled meteorology can provide credible results. Only a few studies have been conducted using dynamically downscaled historical meteorology

59 to simulate regional air quality, all of which were focused on Europe (Katrakou
 60 et al., 2010; Zanis et al., 2011; Lacressonnière et al., 2012; Menut et al., 2013).
 61 Meteorological variables important for simulating air quality, including temper-
 62 ature, precipitation, boundary layer height, and wind speeds, can be degraded
 63 when the regional model is driven by dynamically downscaled meteorology (Ka-
 64 trakou et al., 2010; Lacressonnière et al., 2012; Menut et al., 2013). Katrakou
 65 et al. (2010) and Zanis et al. (2011) used meteorology downscaled from both
 66 the ECMWF ERA40 reanalysis and from the ECHAM5 global climate model to
 67 simulate European ozone concentrations over the decade 1991–2000. Katrakou
 68 et al. (2010) found that the modeling system adequately reproduced average
 69 decadal surface ozone levels at rural monitoring sites, while Zanis et al. (2011)
 70 reported that the performance of their modeling system using downscaled me-
 71 teorology was comparable to that obtained in previous evaluation experiments.
 72 Lacressonnière et al. (2012) used both operational meteorological analyses and
 73 global climate model data to simulate European air quality, and found that the
 74 air quality model driven by climate model data was less skillful in reproducing
 75 air pollutant concentrations. However, the differences in model performance in-
 76 dicators between the two frameworks were not substantial, and they concluded
 77 that predictions driven by the climate model were adequate to simulate future
 78 European pollutant levels and associated health impacts. These studies gen-
 79 erally did not consider changing emissions either within the modeling domain
 80 or the resulting influence on lateral boundary conditions. As a model sensitiv-
 81 ity experiment, Katrakou et al. (2010) found that differences due to plausible
 82 changes in chemical boundary conditions were comparable to biases attributable
 83 to using meteorology from ECHAM5 rather than ERA40.

84 In this study, we conduct an 11-year simulation of air quality in the United
 85 States and evaluate model performance driven by downscaled historical me-
 86 teorological fields that are not enhanced with high-resolution meteorological
 87 observations. The downscaled meteorology used here is meant to emulate the
 88 coarseness (in space and time) of a future climate scenario provided by a GCM.
 89 Year-specific emission estimates are used to capture the large changes in var-

ious precursor emissions that occurred in the United States over the modeled years both within the United States (Xing et al., 2013) and globally (European Commission, 2011). The resulting air quality fields for the historical period are compared against observations from four national networks. Biases in ozone (O_3), total fine particulate matter ($PM_{2.5}$), and speciated fine particulate matter, including sulfate (SO_4^{2-}), ammonium (NH_4^+), nitrate (NO_3^-), and organic carbon (OC), are compared to results typically attained in retrospective air quality model simulations forced by high-resolution, observation-driven dynamic analyses of the meteorological conditions. To our knowledge this work is the longest and most comprehensive air quality modeling study conducted to date using downscaled historical meteorology over the continental United States.

2. MODELING CONFIGURATION

The meteorology used for this study was generated by the Weather Research and Forecasting (WRF) model (Skamarock and Klemp, 2008) version 3.4.1. North America was modeled using a two-way nested 108-36-km configuration and 34 vertical layers up to 50 hPa. WRF was driven by $2.5^\circ \times 2.5^\circ$ reanalyses from the National Centers for Environmental Prediction-Department of Energy Atmospheric Model Intercomparison Project (“R2”) (Kanamitsu et al., 2002). WRF was initialized 00 UTC 2 December 1987 and run continuously through the end of 2010. The physics options in WRF followed Otte et al. (2012), except the present study used the Kain-Fritsch subgrid cloud parameterization with radiative feedbacks (Herwehe et al., 2014). The U.S. Geological Survey (UGSG) 24-category land use classification was used. The Meteorology-Chemistry Interface Processor (Otte and Pleim, 2010) v4.2 was used to prepare WRF outputs for CMAQ.

In WRF, two variants of nudging are available for gridded fields: analysis nudging (or grid nudging) and spectral nudging. Analysis nudging is typically used for retrospective air quality modeling applications with WRF (Otte, 2008a,b), and it is appropriate because observations can be incorporated into

120 the fields that are nudged so that they represent the spatial scales well. Spectral
121 nudging (von Storch et al., 2000; Miguez-Macho et al., 2004) is often preferred
122 by the regional climate modeling community because observations are not avail-
123 able in future climate scenarios and spectral nudging can be used to preserve
124 the characteristics of the large-scale flow. See, for example, comparisons of anal-
125 ysis nudging and spectral nudging within WRF by Liu et al. (2012), Bowden
126 et al. (2012, 2013), Otte et al. (2012), and Omrani et al. (2013). In this study
127 we employed spectral nudging at all levels above the planetary boundary layer
128 (Otte et al., 2012) of potential temperature, U- and V-component winds, and
129 geopotential toward the R2 fields. No finer-scale observational data exogenous
130 to the R2 were assimilated during the simulation.

131 The anthropogenic emissions used in this study were described by Xing et al.
132 (2013). Activity data reported by the State Energy Data System were multiplied
133 by emission factors derived from the U.S. Environmental Protection Agency
134 (EPA) National Emissions Inventory (NEI) and constrained by AP-42 emission
135 control factors. This dataset accounts for various emission drivers, such as pop-
136 ulation, economic growth, and historical pollutant controls. Simply using the
137 NEI, which is considered to be the hallmark of emissions datasets for the United
138 States, was not an option in our 11-year simulation for two reasons. First, the
139 NEI emission profiles are updated every three years. Therefore, emissions in the
140 NEI often do not account for fluctuating economic drivers which can strongly
141 dictate emission levels between reporting years. Second, the methodology to
142 develop the NEI evolves with each release, leading to inconsistencies that make
143 their use in multi-year simulations problematic (Xing et al., 2013). Biogenic
144 emissions were calculated within CMAQ and responded to changes in the me-
145 teorological conditions.

146 Air quality was simulated using the Community Multiscale Air Quality
147 (CMAQ) model version 5.0.2 (Byun and Schere, 2006; Appel et al., 2013), with a
148 36-km domain subset from the WRF 36-km North American domain and span-
149 ning the continental United States. The CMAQ simulation used the same USGS
150 land use classification and 34-layer vertical structure as the WRF simulation.

151 CMAQ was configured with the Carbon Bond 2005 gas phase mechanism with
152 updates for toluene and chlorine chemistry (CB05TUCL; Sarwar et al., 2011),
153 the AERO6 aerosol module (Simon and Bhawe, 2012; Appel et al., 2013; Nolte
154 et al., 2015), and online calculation of photolysis rates (Foley et al., 2010). Initial
155 and lateral boundary conditions for CMAQ originated from a 108-km CMAQ
156 simulation that encompassed the Northern Hemisphere (Xing et al., 2015). An
157 initial spin-up of ten days was conducted, followed by 11 years of continuous
158 simulations, spanning 1 January 2000 through 31 December 2010.

159 Retrospective air quality studies for continental-scale domains over annual or
160 shorter periods using regional-scale models like WRF and CMAQ typically use
161 higher spatial resolution than the 36-km grid spacing used here. For example,
162 Foley et al. (2010), Appel et al. (2011, 2013), and Nolte et al. (2015) all employed
163 12-km grid spacing, while studies in California typically use grid spacings of
164 4–5 km (Ying and Kleeman, 2006) to represent accurately the meteorological
165 flow in complex terrain and the strong spatial gradients in emissions. However,
166 simulations to assess air quality trends (Hogrefe et al., 2011; Gan et al., 2016) or
167 the influence of climate change on air quality (Nolte et al., 2008; Gao et al., 2013;
168 Trail et al., 2014; Gonzalez-Abraham et al., 2015) must be run for multiple years
169 and require intense computational resources that make it impractical to use as
170 fine a horizontal grid spacing. For this reason, 36-km modeling is commonly
171 used to examine health impacts under a changing climate (Bell et al., 2007; Post
172 et al., 2012; Fann et al., 2015; Wilson et al., 2016). Gan et al. (2016) reported
173 that the largest differences in PM concentrations simulated using 36-km and
174 12-km model configurations were episodic, while differences in long-term trends
175 were small. The results of Gan et al. (2016) further justify the use of 36-km
176 grid spacing, since most studies of the health effects of changes in air quality
177 attributable to climate change are based on long-term average concentrations.

178 3. EVALUATION FRAMEWORK

179 Hourly O_3 observations from EPA’s Air Quality System (AQS) were pro-
180 cessed into maximum daily 8-h average concentrations (MDA8), for comparison

181 to the U.S. National Ambient Air Quality Standard (NAAQS). $\text{PM}_{2.5}$ concen-
 182 trations were obtained from the Chemical Speciation Network (CSN), the Clean
 183 Air Status and Trends Network (CASTNet), and the Interagency Monitoring of
 184 Protected Visual Environments (IMPROVE) network. Total $\text{PM}_{2.5}$ is evaluated,
 185 as are its most abundant speciated components: sulfate (SO_4^{2-}), ammonium
 186 (NH_4^+), nitrate (NO_3^-), and organic carbon (OC). AQS and CSN are pri-
 187 marily urban networks, with monitors generally located near populated regions.
 188 In contrast, CASTNet and IMPROVE are largely rural networks. The spatial
 189 distributions of the O_3 and PM monitoring networks are shown in Fig. 1. Al-
 190 though there are differences in the testing and acceptance protocols among the
 191 networks, these differences minimally impact the measurements due to stringent
 192 quality assurance procedures (Solomon et al., 2014) and are neglected for this
 193 analysis. Additionally, it should be noted that some network monitors operate
 194 only seasonally, and that some monitors came online or went offline during the
 195 11-year simulation period. All available measurements reported by each net-
 196 work over the modeling period are used in these calculations. Accordingly, the
 197 number of sites in each network that are included in the analysis vary slightly
 198 by year and season.

199 Modeled values are taken from the grid cells and time periods corresponding
 200 to the observations. The model evaluation metrics used in this study are mean
 201 bias (MB), mean error (ME), mean fractional bias (MFB), and mean fractional
 202 error (MFE), as defined in Table 1. The MFB and MFE are normalized by
 203 the mean of each model-observation pair, and therefore do not assume that the
 204 observations are the absolute truth. Additionally, the MFB and MFE have the
 205 advantage of being bounded, symmetric metrics, meaning that they give equal
 206 weight to modeled concentrations greater than observations and to modeled
 207 concentrations less than observations, and are not unduly influenced by a few
 208 instances where the observations in the denominator are very low (Yu et al.,
 209 2006).

210 Boylan and Russell (2006) proposed a PM model evaluation framework using
 211 specified “goal” and “criteria” values for MFB and MFE, reproduced in Table 1.

212 These definitions qualitatively classify model performance within three zones:
 213 (1) meeting the goal; (2) failing the goal but meeting the criteria; and (3)
 214 failing to meet either the goal or the criteria. According to Boylan and Russell
 215 (2006), the modeling goal is “considered to be close to the best a model can
 216 be expected to achieve.” If the model performance satisfies the criteria but
 217 fails to meet the goal, the results are still considered acceptable. If neither the
 218 goal nor the criteria is met, the performance is outside the range of what is
 219 considered acceptable. Note that the Boylan and Russell (2006) specifications
 220 allow for greater relative error at lower concentrations. Conversely, at higher
 221 concentrations, the MFB goal and criteria asymptotically approach 30% and
 222 60%, respectively, while the MFE goal and criteria asymptotically approach
 223 50% and 75%, respectively. Following Boylan and Russell (2006), we assess the
 224 model performance for each constituent at each network on a monthly basis.

225 Finally, the results from this study are compared to other published air
 226 quality modeling studies. The main source for this comparison is Simon et al.
 227 (2012), who compiled statistical results from 69 contemporary modeling evalu-
 228 ations conducted between 2006 and 2012.

229 **4. RESULTS**

230 *4.1. Meteorology*

231 Since meteorological fields strongly influence the simulation of air quality, an
 232 analysis of the meteorological input to the CMAQ simulations is conducted here.
 233 Only a brief analysis of the meteorology is provided because the focus of this pa-
 234 per is on the performance of the air quality simulations. The 36-km downscaled
 235 meteorological fields simulated by WRF are compared against hourly 0.31° fields
 236 from the Climate Forecast System Reanalysis (CFSR) (Saha et al., 2010) for
 237 2-m temperature and against three-hourly 32-km North American Regional Re-
 238 analysis (NARR) (Mesinger et al., 2006) fields for precipitation. Regionally
 239 averaged monthly deviations in the downscaled WRF simulations from CFSR
 240 and NARR are shown in Fig. 2 for the 11-year historical period, 2000–2010,
 241 which was used for the CMAQ simulations in this analysis. Monthly average

242 temperature differences across the United States have a cool bias of up to 2 K.
 243 The 2-m temperatures have very high correlations (0.997–0.999), which is typ-
 244 ical for modeling diurnal and annual cycles with WRF, and the monthly mean
 245 bias in 2-m temperature ranges from -1.2 to -0.3 K within the six regions.
 246 The WRF simulation also has a wet bias of 0 – 40 mm mon $^{-1}$, and the mean
 247 regional bias in precipitation ranges from 11.0 mm mon $^{-1}$ in the Southeast to
 248 37.2 mm mon $^{-1}$ in the Northeast. As expected, the precipitation correlation
 249 coefficient with respect to NARR is lower for precipitation than for temper-
 250 ature, ranging from 0.796 in the Southeast to 0.933 in the Northwest. The
 251 correlations in precipitation tend to be higher in regions that are dominated by
 252 large-scale precipitation and lower in regions that are dominated by convective
 253 precipitation.

254 The levels of bias in temperature and precipitation in this WRF simulation
 255 are consistent with those obtained for 1988–2007 using a similar configuration
 256 of WRF with spectral nudging (see simulation “SN” in Figs. 4–6 of Otte et al.
 257 (2012) and Fig. 3 of Spero et al. (2014)). A comprehensive evaluation of that SN
 258 simulation appears in Otte et al. (2012), where means, variability, and extremes
 259 in meteorological fields over a 20-year period are simulated with reasonable
 260 accuracy. The regional correlation coefficients and mean biases in Fig. 2 of the
 261 present study are comparable to the SN simulation shown in Fig. 3 of Spero
 262 et al. (2014), which provides confidence that the WRF fields are of sufficient
 263 quality to be used in driving a CMAQ simulation.

264 *4.2. Ozone*

265 To examine air quality, we first compare the central portion of the observed
 266 and modeled O_3 distributions. Figure 3 shows the interquartile range (IQR, i.e.,
 267 the 25th–75th percentiles) of the observed and modeled MDA8 O_3 concentra-
 268 tions for each season and year from 2000 to 2010. A few trends are apparent in
 269 the observed MDA8 O_3 levels throughout this period. First, the observations
 270 show a decreasing trend in the 75th percentile summer (JJA) O_3 . Also evident
 271 is an increasing trend in 25th percentile winter (DJF) values. These trends were

discussed by Cooper et al. (2014) and Simon et al. (2015), with both attributing these trends primarily to recent declines in precursor NO_x emissions in the U.S. over this time period.

The median modeled MDA8 O_3 is essentially unbiased during the spring (MAM), but is positively biased by approximately 10 ppb during the summer and fall (SON) and by around 7 ppb during the winter. Despite this positive bias, CMAQ accurately captures the summer and winter O_3 trends, responding to the decreasing precursor emissions, as well as interannual meteorological variability, as shown by the anomalously low observed and modeled O_3 levels during the summer of 2004. The spread in the distribution (as represented by the magnitude of the IQR, or the height of the bars in Fig. 3) is well simulated by the model for the summer, but is slightly compressed relative to the observations for spring and fall, and is substantially underrepresented during the winter. Additionally, the biases in the 25th percentiles, medians, and 75th percentiles for each season show consistency from year to year (Table 2). The standard deviation of the bias in each percentile is less than the absolute average, providing coefficients of variation consistently below 1, indicating low variability.

Simon et al. (2012) analyzed CMAQ predictions from Appel et al. (2011) and calculated the mean bias for four regions of the U.S. over the May–September O_3 season between 2002 and 2006. The median of their mean bias calculations was positive 2.9 ppb. Across the modeling studies compiled by Simon et al. (2012), the IQR of O_3 mean bias spanned approximately 0–6 ppb, with the upper end of the distribution extending to about 14 ppb. As shown in Table 3, the annual mean biases in the present study, when considering all seasons, range from 5.8–8.0 ppb, at or slightly above the Simon et al. (2012) 75th percentile but well within the range of prior studies. Eder and Yu (2006) reported an MDA8 O_3 NMB of 8.1% and NME of 19.6% for April–September 2001. Here, the analogous statistics for the same modeled period were 14% and 23%, respectively. The IQR of NMB and NME reported by Simon et al. (2012) spanned approximately 0–8% and 15–22%, respectively, with the distributions extending to 20% and 32%. For the 11-year period in the present study, annual NMB and NME values

range from 14–17% and 22–25%, respectively. Again, these are higher than the 75th percentiles from the Simon et al. (2012) compilation, but are within their reported range.

CMAQ generally underestimates the dynamic range of O_3 concentrations compared to measurements, with a positive bias at the low end of the distribution, a negative bias at the high end of the distribution, and performing best at predicting mid-range O_3 levels (Foley et al., 2010; Simon et al., 2012). Additionally, U.S. EPA modeling guidance for O_3 attainment demonstration (U.S. EPA, 2007) suggests excluding data points where the observed MDA8 O_3 falls below a specified minimum threshold, due to the high frequency of low O_3 concentrations and the greater relevance of higher values for human health and NAAQS compliance. Accordingly, model performance statistics for each yearly dataset were recalculated after applying thresholds of 60 ppb, corresponding to a moderate level of O_3 , and 70 ppb, equal to the 2015 revised NAAQS. Table 3 shows improved model performance statistics for higher O_3 concentrations. When a 60 ppb threshold is applied, the model bias is slightly negative (MB = -1.2 ppb; MFB = -2.7%), and with a 70 ppb threshold, the model is more strongly negatively biased (MB = -4.2 ppb; MFB = -6.4%). In comparison, Simon et al. (2012) reported larger negative biases at high O_3 levels: the median bias and median fractional bias were -4.95 ppb and -7.6% when using a 60 ppb threshold and -9.0 ppb and -12.3% when using a 75 ppb threshold. Thus, the somewhat higher positive bias of the model configuration used in this study results in lower biases at the higher end of the O_3 distribution.

4.3. $PM_{2.5}$

Figures 4 and 5 show the IQR of the observed and modeled total $PM_{2.5}$ concentrations for each season and year from 2000 to 2010 for the CSN and IMPROVE sites, respectively. CASTNet is not included in this portion of the analysis since it does not report total $PM_{2.5}$ concentrations. Observed concentrations at CSN sites are highest during the summer and winter and lowest during the spring. Median concentrations show a negative decadal trend for

all seasons except spring, while there is a strong decline in 75th percentile concentrations during the fall and winter. Modeled $\text{PM}_{2.5}$ concentrations at CSN sites are negatively biased throughout the year, with the largest negative bias during the summer, which has been attributed to underpredictions of secondary organic aerosol (Carlton et al., 2010; Nolte et al., 2015). Despite the negative bias, however, Fig. 4 shows that the model captures both the declining decadal trends in medians and the interannual variability. The distribution of modeled values in the spring features almost no bias and is the best predicted season.

Relative to measurements at the more urban CSN sites, $\text{PM}_{2.5}$ concentrations at IMPROVE sites are lower throughout the year. As with CSN, concentrations in the IMPROVE network are highest during the summer. Unlike observations at CSN locations, however, Fig. 5 shows little to no trend in median concentrations, with the possible exception of a negative summer trend since 2007. The absence of a negative trend is attributable to the fact that IMPROVE sites are primarily in the western U.S., while the majority of the NO_x and SO_2 emissions reductions have occurred in the eastern U.S. Modeled concentrations are slightly positively biased during the winter and negatively biased the rest of the year, with the negative bias largest during the summer. The magnitude of the IQR is well simulated during spring and fall, but is underestimated during summer and overestimated during winter.

Total $\text{PM}_{2.5}$ MFB and MFE model performance statistics are presented in Fig. 6 in comparison to the Boylan and Russell (2006) curves. For these “bugle plots”, statistics are computed on a monthly basis, resulting in 132 data points (11 modeled years $\times$ 12 months per year) for each monitoring network. The model always meets the less-stringent criteria for both MFB and MFE, and it often meets the goal. Also evident in Fig. 6 are the higher concentrations across the CSN sites in comparison to IMPROVE.

The same model evaluation methods are also carried out for the major $\text{PM}_{2.5}$ components at the CSN, IMPROVE, and CASTnet sites, when available. As shown in the bugle plots for sulfate, nitrate, ammonium, and organic carbon (Fig. 7), model performance varies by constituent and network. Performance is

best for sulfate, which usually meets the performance goal for both MFB and MFE across all three networks. The goal is not met for only a few data points from CSN sites, where the model negative bias is too large, but the results still comfortably satisfy the performance criteria. The model also generally performs well for ammonium, always satisfying the performance criteria and nearly always satisfying the goal, particularly at CASTNet locations where the concentrations are lower. For CSN sites, however, the negative bias in ammonium concentrations is sometimes large enough to cause the MFB and MFE to fail the model performance goal, though the criteria is met.

Nitrate performs somewhat worse than sulfate and ammonium, with more points falling outside the MFB and MFE performance goals and a few failing to meet the performance criteria. Model performance is further degraded for organic carbon, which consistently underpredicts when observed concentrations exceed $3 \mu\text{g m}^{-3}$, resulting in MFB and MFE values that frequently do not meet the performance goal and often do not meet the performance criteria. Similar performance for OC was reported by Boylan and Russell (2006) in their original application of this evaluation method.

Figure 8 expands on the analysis by comparing the distribution of monthly MFB and MFE statistics for total $\text{PM}_{2.5}$ and its constituents from this study to the results from other contemporary modeling studies summarized in Simon et al. (2012). Boxes show the IQR of monthly MFB and MFE computed across all modeled years, with the whiskers on each box extending to the 10th and 90th percentiles. It should be noted that MFB improves as the absolute value approaches zero. The distribution of monthly MFB in this study was comparable to the distribution in Simon et al. (2012), or better. The median MFB at CSN locations was -16.8% and -6.9% at IMPROVE locations. The median MFB reported in Simon et al. (2012) for total $\text{PM}_{2.5}$ was -12.0% , with their IQR spanning -33.5 to -3.0% . As seen in the bottom panel of Fig. 8, the IQR of the MFE values for total $\text{PM}_{2.5}$ from this study were 42.5 – 48.9% at CSN locations and 44.1 – 51.1% at IMPROVE locations. In comparison, the IQR of the median MFE in Simon et al. (2012) was 34.0 – 54.0% .

Also shown in Fig. 8 are the IQR of the speciated $\text{PM}_{2.5}$ MFB and MFE results from this study for each of the observation networks and the results compiled in Simon et al. (2012). The IQR of the MFB results in this study is generally comparable to or better than the results from contemporary retrospective modeling studies for sulfate, ammonium, nitrate, and organic carbon. An exception is for organic carbon predictions at CSN sites, which is more strongly underpredicted in this study. However, the results are comparable when considering the 10th and 90th percentile results from Simon et al. (2012). Also included in Fig. 8 is the IQR of the MFE for the same datasets. The MFE for sulfate and nitrate perform well, while ammonium and organic carbon feature slightly higher IQR values of MFE at CSN sites, when compared to Simon et al. (2012). As previously discussed, the speciated portions of $\text{PM}_{2.5}$ that were simulated most poorly, according to the Boylan and Russell (2006) performance metrics, were nitrate and organic carbon. The results in Fig. 8 indicate that the regional climate and air quality modeling framework used in this study produces levels of MFB and MFE for nitrate and organic carbon at levels consistent with other published studies.

5. DISCUSSION AND CONCLUSIONS

Historical $2.5^\circ \times 2.5^\circ$ global meteorological reanalyses from R2 were used as a proxy for a GCM and downscaled to 36-km resolution using the WRF model. The WRF simulations were constrained using spectral nudging toward the R2, but were intentionally run without assimilating fine-scale observations, because such observations are not available to constrain regional climate simulations for future periods. The downscaled meteorological fields from WRF were then used to drive CMAQ for an 11-year period to evaluate the effects of these downscaling techniques on the simulated concentrations of various pollutants. The analysis of air quality focused on O_3 , total $\text{PM}_{2.5}$, and speciated fine PM, including sulfate, ammonium, nitrate, and organic carbon. Concentrations simulated by CMAQ were compared against data from four national networks spanning the continental United States. These networks included the

425 AQS, CSN, IMPROVE, and CASTNet monitoring sites. The modeling period
426 spanned 2000–2010, which also corresponded with significant decreases across
427 the U.S. in precursor emissions for O_3 and $PM_{2.5}$ (U.S. EPA, 1998; Xing et al.,
428 2013). This signal was evident in statistical measures of pollutant levels at the
429 monitoring sites used in this study.

430 Seasonal O_3 trends were well simulated by this regional climate and air
431 quality modeling configuration. CMAQ simulated the decrease in 75th per-
432 centile summer O_3 and the increase in 25th percentile winter O_3 . In addition
433 to capturing the overall O_3 trends, CMAQ was able to maintain consistent bias
434 between the observed and modeled percentiles. Analysis was also performed
435 in which we excluded data points where observations were below a specified
436 threshold, to analyze how CMAQ performed in simulating higher levels of O_3 ,
437 which are of most concern for human health and NAAQS compliance. After
438 applying thresholds of 60 ppb and 70 ppb to the observed data, mean model
439 bias from all years simulated dropped to -1.2 ppb and -4.2 ppb, respectively.
440 After applying a 60 ppb and a 75 ppb threshold to a previous CMAQ study,
441 Simon et al. (2012) reported a median model bias of -4.95 ppb and -12.3 ppb,
442 respectively. However, the median model bias from their complete analysis was
443 2.9 ppb while the mean model bias from the present study was 6.8 ppb. This
444 indicates that the regional climate and air quality modeling configuration used
445 here simulates somewhat higher levels of O_3 when compared to other published
446 air quality modeling results, but these higher levels actually improve CMAQ’s
447 negative bias at the higher end of the O_3 concentration distribution.

448 Observations of total $PM_{2.5}$ also showed some negative trends over the mod-
449 eling period, particularly a decrease in 75th and 50th percentile concentrations
450 during the fall and winter months at CSN sites. Total $PM_{2.5}$ concentrations
451 at IMPROVE network locations featured only a modest negative trend during
452 the summer. CMAQ was negatively biased at CSN sites, with the largest ab-
453 solute bias occurring in the summer when the concentrations were highest. At
454 IMPROVE sites, CMAQ was typically biased low. An exception was during
455 winter months, when the three analyzed percentiles of the modeled results were

positively biased compared to the same percentiles of the observations. When evaluating the predicted total $\text{PM}_{2.5}$ concentrations using the performance metrics of Boylan and Russell (2006), the results demonstrate that this modeling configuration is capable of simulating total $\text{PM}_{2.5}$ concentrations at levels “considered to be close to the best a model can be expected to achieve.” The results from this study were also comparable to the results from other contemporary modeling studies. The median monthly MFB from this study spanned -4.1% to -24.7% at CSN sites and 0.6% to 11.2% at IMPROVE sites while the median MFB from the Simon et al. (2012) compilation study was 12.0%.

As expected, some components of $\text{PM}_{2.5}$ were predicted better than others. Of the evaluated components, which included sulfate, ammonium, nitrate, and organic carbon, model performance was best for sulfate and ammonium, which typically met the modeling goals for mean fractional bias and mean fractional error and always met the modeling criteria. Inter-network trends also existed for these two components. For example, while the average observed concentrations of sulfate and ammonium were comparable at CSN and CASTNet locations, the levels of MFE at CASTNet locations for both these components were typically lower.

Nitrate and organic carbon were not predicted as well as sulfate and ammonium by CMAQ. Both components featured individual months during the 11-year period where the MFB and MFE statistics failed to meet the performance criteria. Aerosol nitrate is principally formed via condensation of gas-phase nitric acid (HNO_3). The amount of nitrate in the particle phase depends not only on the amount of total nitrate available, but also on the concentrations of other species, including sulfate and ammonium, as well as temperature and relative humidity. As a consequence, nitrate concentrations are particularly challenging to simulate accurately (Yu et al., 2005). Nitrate is also difficult to measure, as it is subject to revolatilization from a filter during sampling. For organic carbon, it has been previously reported that CMAQ frequently underestimates its mass, particularly in summer months (Carlton et al., 2010). To determine whether these levels of model bias for nitrate and organic carbon were attributable to

487 using downscaled meteorology or represented typical levels of bias, the MFB
488 and MFE results from this study were again compared to the results from the
489 Simon et al. (2012) compilation study. The results demonstrate that the MFB
490 and MFE levels in this study were generally comparable to results from other air
491 quality studies, except organic carbon at the typically urban CSN sites, where
492 it was further underestimated.

493 The results demonstrate that this air quality modeling configuration, in
494 which coarse-scale meteorological fields are dynamically downscaled with a re-
495 gional climate model, simulates O_3 , total $PM_{2.5}$, and speciated $PM_{2.5}$ compo-
496 nents at a level of performance that is generally considered acceptable based
497 on published performance metrics. Moreover, the model performance is statis-
498 tically comparable to results obtained in air quality modeling studies that are
499 driven by fine-scale meteorological observations. Therefore, when applying this
500 downscaling methodology to simulate regional air quality in a future climate
501 scenario, additional modeling bias strictly from this framework should not be
502 substantial. The differing biases obtained in this study compared to other air
503 quality modeling studies using WRF and CMAQ may be due to the use of mete-
504 orology downscaled from coarse global fields, or they may be attributable to the
505 use of slightly different physics options and land use representations in WRF. It
506 should be noted that the model performance obtained here likely represents an
507 upper bound on currently attainable model performance in a downscaling ap-
508 plication, because a GCM cannot be expected to be as accurate as a reanalysis,
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Table 1: Metrics used in this study for evaluation of model performance for O₃ (MB, ME) and PM (MFB, MFE).

Metric	Equation ^a
Mean Bias	$MB = \frac{1}{N} \sum (M_i - O_i)$
Mean Error	$ME = \frac{1}{N} \sum M_i - O_i $
Mean Fractional Bias	$MFB = \frac{2}{N} \sum \frac{M_i - O_i}{M_i + O_i} \times 100$
Mean Fractional Error	$MFE = \frac{2}{N} \sum \frac{ M_i - O_i }{M_i + O_i} \times 100$
MFB Goal ^b	$MFB \leq \pm 170 e^{-(\overline{M_i} + \overline{O_i})} + 30$
MFE Goal	$MFE \leq 150 e^{-\frac{2}{3}(\overline{M_i} + \overline{O_i})} + 50$
MFB Criteria	$MFB \leq \pm 140 e^{-(\overline{M_i} + \overline{O_i})} + 60$
MFE Criteria	$MFE \leq 125 e^{-\frac{2}{3}(\overline{M_i} + \overline{O_i})} + 75$

^a O_i are observed O₃ or PM values; M_i are modeled values for the corresponding grid cell and time period; sums are taken across all sites for a given network and across all days for a given period.

^bGoal and criteria definitions from Boylan and Russell (2006); $\overline{M_i}$ and $\overline{O_i}$ denote mean PM concentrations in $\mu\text{g m}^{-3}$.

Table 2: Seasonal bias (ppb) of modeled 25th percentile, median, and 75th percentile MDA8 O₃ compared to the AQS network. Mean is expressed as the average of each absolute value. CV = coefficient of variation.

Year	MAM			JJA			SON			DJF		
	25th	median	75th	25th	median	75th	25th	median	75th	25th	median	75th
2000	3.8	2.2	0.6	10.8	11.3	10.4	12.1	11.5	11.0	10.0	8.3	5.5
2001	1.5	0.2	-0.3	9.7	10.3	9.9	11.3	10.2	9.4	10.0	7.9	5.3
2002	2.1	0.8	0.5	9.5	9.0	7.1	12.8	11.3	10.3	8.7	7.1	4.7
2003	3.0	0.7	-0.1	9.9	10.2	9.8	11.1	10.8	9.5	8.2	6.2	4.6
2004	2.2	0.4	-0.8	10.4	10.5	9.6	12.9	11.4	10.4	9.1	7.1	4.2
2005	1.2	-0.3	-1.5	11.1	10.5	9.1	11.9	10.9	9.7	9.2	6.9	4.7
2006	1.2	-0.2	-1.3	9.2	8.3	7.2	11.0	10.9	10.7	9.2	6.9	4.3
2007	1.9	0.7	-0.7	10.1	9.0	8.0	12.4	11.6	9.7	8.0	6.6	4.9
2008	-0.3	-2.0	-3.6	8.4	7.3	6.4	12.4	11.7	10.1	8.1	6.1	4.2
2009	1.0	-1.3	-2.6	10.1	10.0	9.9	12.9	11.3	10.0	7.4	5.6	3.7
2010	2.0	0.5	-0.8	10.2	10.3	9.5	9.7	8.3	7.0	5.9	4.7	3.4
<i> mean </i>	1.8	0.9	1.2	10.0	9.7	8.8	11.9	10.9	9.8	8.5	6.7	4.5
<i>Std.Dev.</i>	0.98	0.70	1.07	0.75	1.16	1.36	1.00	0.96	1.06	1.20	1.01	0.64
<i>CV</i>	0.53	0.82	0.90	0.08	0.12	0.15	0.08	0.09	0.11	0.14	0.15	0.14

Table 3: Annual model performance statistics for MDA8 O₃ computed only using days on which observations exceed a specified threshold

Threshold	Metric	Units	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010	Total
0 ppb	MB	ppb	8.0	7.1	6.7	7.1	7.3	6.8	6.6	6.9	5.8	6.9	6.2	6.8
0 ppb	ME	ppb	11.5	11.1	11.0	11.1	10.9	10.6	10.2	10.1	9.9	10.2	9.3	10.5
0 ppb	MFB	percent	20.0	17.4	16.9	16.8	18.6	16.6	16.1	16.6	15.0	17.7	14.9	16.9
0 ppb	MFE	percent	26.9	25.1	25.0	24.8	25.9	24.0	23.4	23.0	23.2	24.7	21.2	24.3
0 ppb	n	–	298160	287162	298858	274813	274142	277290	317105	322632	329276	333769	319170	3332377
60 ppb	MB	ppb	–0.1	0.0	–1.0	–0.7	–2.2	–0.8	–0.8	–1.2	–3.9	–1.8	–1.4	–1.2
60 ppb	ME	ppb	10.0	10.3	10.8	10.2	9.7	9.5	9.1	8.8	9.8	9.8	8.3	9.7
60 ppb	MFB	percent	–1.1	–1.1	–2.3	–1.9	–4.4	–2.2	–2.3	–2.6	–6.9	–4.1	–3.0	–2.7
60 ppb	MFE	percent	14.2	14.5	14.9	14.6	14.4	13.7	13.3	12.9	15.0	14.8	12.5	14.1
60 ppb	n	–	52499	53231	57180	48400	34830	50546	50294	52955	41527	26169	36342	503923
70 ppb	MB	ppb	–3.4	–2.9	–4.2	–4.3	–5.8	–3.9	–3.1	–4.7	–7.5	–4.5	–4.2	–4.2
70 ppb	ME	ppb	10.6	10.9	11.5	11.3	11.1	10.3	10.0	9.9	11.9	11.0	10.0	10.8
70 ppb	MFB	percent	–5.2	–4.8	–6.2	–6.5	–8.9	–6.1	–5.0	–7.1	–11.0	–7.0	–6.6	–6.4
70 ppb	MFE	percent	13.8	14.1	14.6	14.7	15.2	13.8	13.2	13.4	16.4	14.9	13.6	14.2
70 ppb	n	–	22456	22737	28654	19059	11421	19827	17676	18632	12313	6680	8663	188118

Figure 1: Locations of O₃ AQS sites (green) and PM_{2.5} CSN (gold), CASTNet (red), and IMPROVE (blue) sites.

Figure 2: Monthly area-averaged precipitation difference (mm) from NARR (solid line) and monthly area-averaged 2-m temperature difference (K) from CFSR (dotted line) for the WRF simulation for 2000–2010. Correlation coefficient (“R”) and mean bias (“MB”) are provided for average monthly precipitation with respect to NARR and for average monthly 2-m temperature with respect to CFSR. Regions are as defined in Spero et al. (2014).

Figure 3: Seasonal distributions of MDA8 O_3 at AQS sites, where “MAM” is March-April-May, “JJA” is June-July-August, “SON” is September-October-November, and “DJF” is December-January-February of the same year. Dark boxes show the interquartile range (IQR; 25th through 75th percentiles) of the observations, and red lines are the associated medians; light boxes show the IQR of modeled mixing ratios, and blue lines are the associated medians.

Figure 4: Seasonal distributions of total PM_{2.5} concentrations at CSN sites. Dark boxes show the IQR of the observations, and red lines are the associated medians; light boxes show the IQR of modeled concentrations, and blue lines are the associated medians.

Figure 5: Seasonal distributions of total PM_{2.5} at IMPROVE sites. Dark boxes show the IQR of the observations, and red lines are the associated medians; light boxes show the IQR of modeled concentrations, and blue lines are the associated medians.

Figure 6: Total $PM_{2.5}$ mean fractional bias (top) and mean fractional error (bottom) calculated for each month from 2000 to 2010, compared to the Boylan and Russell (2006) goal and criteria performance curves described in Table 1.

Figure 7: Speciated PM_{2.5} mean fractional bias (top) and mean fractional error (bottom) calculated for each month from 2000 to 2010, compared to the Boylan and Russell (2006) goal and criteria performance curves described in Table 1.

Figure 8: Modeled fractional bias (top) and fractional error (bottom) in PM_{2.5} and its constituents for 2000–2010, grouped by observation network and compared to the compilation study of Simon et al. (2012). The bars represent the IQR of each respective dataset and the whiskers extend to the 10th and 90th percentiles.