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Dynamic evaluation of CMAQ part I: Separating the effects of changing emissions and changing meteorology on ozone levels between 2002 and 2005 in the eastern US

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HIGHLIGHTS

- A dynamic evaluation is conducted of ozone predictions from the CMAQ model.
- Impacts on ozone from emissions and meteorology are separately quantified.
- Decreases in ozone levels due to decreasing emissions were not fully captured.
- Causes for model biases are investigated.

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ABSTRACT

A dynamic evaluation of the Community Multiscale Air Quality (CMAQ) modeling system version 5.0.1 was conducted to evaluate the model's ability to predict changes in ozone levels between 2002 and 2005, a time period characterized by emission reductions associated with the EPA's Nitrogen Oxides State Implementation Plan as well as significant reductions in mobile source emissions. Model results for the summers of 2002 and 2005 were compared to simulations from a previous version of CMAQ to assess the impact of model updates on predicted pollutant response. Changes to the model treatment of emissions, meteorology and chemistry had substantial impacts on the simulated ozone concentrations. While the median bias for high summertime ozone decreased in both years compared to previous simulations, the observed decrease in ozone from 2002 to 2005 in the eastern US continued to be underestimated by the model. Additional "cross" simulations were used to decompose the model predicted change in ozone into the change due to emissions, the change due to meteorology and any remaining change not explained individually by these two components. The decomposition showed that the emission controls led to a decrease in modeled high summertime ozone close to twice as large as the decrease attributable to changes in meteorology alone. Quantifying the impact of retrospective emission controls by removing the impacts of meteorology during the control period can be a valuable approach for communicating to policy makers the net benefit of national control measures.

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1. Introduction

A key regulatory use for air quality models is to estimate the impact of different emission reduction strategies on ambient concentration levels. Dynamic model evaluation, concerned with comparing observed and simulated changes in pollutant concentrations (Dennis et al., 2010), is thus an integral step in establishing credibility in the model predicted pollutant response. Several previous dynamic evaluation studies have focused on 2002 and 2005 to evaluate the model's ability to predict ozone changes due to decreasing NO_x emissions (e.g., Gilliland et al., 2008; Godowitch et al., 2011). Emission reductions during this time period were driven by the EPA's Nitrogen Oxides State Implementation Plan Call (NO_x SIP Call) rule, implemented in 2003 through 2004 and aimed at reducing NO_x emissions from power plants in the eastern US, as well as substantial reductions in mobile

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source emissions. The NO_x SIP Call rule was designed to reduce the interstate transport of ozone and was implemented using a NO_x Budget Trading Program for states to track and trade NO_x allowances from their individual ozone season NO_x "budget". The NO_x SIP Call is an attractive natural experiment for dynamic model evaluation because the emission reductions that occurred over this time period contributed to a substantial decrease in observed ozone levels (20–30%) and because Continuous Emissions Monitoring System (CEMS) measurements were available for major electrical generating units (EGUs), allowing for accurate quantification of the emission reduction from this sector within an air quality modeling system.

Gilliland et al. (2008) provided a dynamic evaluation of the Community Multiscale Air Quality modeling system version 4.6 (CMAQv4.6) using model simulations of the summers before (2002) and after (2004, 2005) the NO_x SIP Call was implemented. The analysis found that the air quality model predictions underestimated the ozone reductions observed after the implementation of the rule. An evaluation of CMAQ version 4.7 spanning the summers of 2002–2006 presented in Godowitch et al. (2010) showed a similar underestimation of the decrease in max 8hr average ozone levels compared to observations. Their analysis of modeled and observed weekday morning NO_x levels in urban areas suggested this was due, in part, to an underestimation of the decreasing trend in non-mobile surface NO_x emissions (i.e. area, low-level non-EGU point sources and nonroad sources) in the model inputs. This hypothesis was investigated further using sensitivity analysis of CMAQv4.7 by Napelenok et al. (2011), Zhou et al. (2013), and Kang et al. (2013). Overall, these studies found that modeled ozone response could be improved by adjusting ground-level NO_x emission inputs, but the predicted ozone change still under-estimated observed reductions.

Used as-is, this under-estimate in response would be conservative when designing pollution control scenarios (i.e. it would lead to more improvement in air quality than predicted). However, dynamic evaluation results have been used to justify designing less-stringent control plans. For example, Pegues et al. (2011) cited three states that used dynamic evaluation studies to justify increasing model ozone responsiveness in the weight of evidence determinations of their respective State Implementation Plans for reaching ozone attainment in 2009. Such adjustments ended up leading to incorrect attainment predictions. Therefore, addressing the potential causes of the muted model response found in previous dynamic evaluation studies is a key priority for ongoing model developments. Here we assess the impact of model updates included in CMAQv5.0.1 on the dynamic evaluation of ozone predictions for the 2002 – 2005 time period.

An additional challenge in a dynamic evaluation is that the observed air quality changes over time are also driven by meteorological variability. To separate the impacts on ozone levels due to emission changes from changes in meteorological conditions across the summers of 2002 and 2004, Godowitch et al. (2008) constructed modeling scenarios that combine meteorological inputs from one summer with the CEMS data for NO_X and SO_2 emissions from point sources from the other summer time period. Emissions in other sources categories were left unchanged for these "cross" simulations. The cross simulations demonstrated how the cooler wetter conditions in 2004 compared to the 2002 base year had as large an impact on the ozone mixing ratios as the emissions reductions associated with the control program. Here we used simulations for the summers of 2002 and 2005 because 2005 had more similar meteorological conditions to 2002 in terms of the number of days with favorable versus non-favorable conditions for ozone production. We expand on the idea of using cross simulations by creating simulations that more fully quantify the effect of meteorology on emissions from several sectors, including both point and mobile sectors, the main sources of precursor NO_x emissions for ozone formation in this region. Specifically, a method was developed to adjust CEMS data to account for differences in electricity demand driven by year-specific meteorology. Removing the impacts of meteorology during the control period provides a valuable approach for quantifying the net benefit of specific control measures and can be used to better diagnose the biases identified in a dynamic evaluation study.

2. Model approach and observational datasets

All simulations were conducted using CMAQv5.0.1 (Appel et al., 2013) including the inline photolysis option and the Carbon Bond version 2005 chemical mechanism (Yarwood et al., 2005) updated with toluene reactions (CB05-TU; Whitten et al., 2010). The simulations were performed over the continental US for June 1 through August 31 2002 and 2005 using a grid with 12 km horizontal resolution and 35 vertical layers. Meteorological inputs were based on WRF3.3 with MCIPv4.0 (see Appel et al., 2013 for further details). Emission inputs were developed based on 2002 and 2005 National Emission Inventory data (http://www.epa.gov/ttn/chief/eiinformation.html) using the Sparse Matrix Operator Kernel Emissions (SMOKE; Houyoux et al., 2000) processing system version 3.1 and included year-specific data from CEMS for large combustion and industrial processes (mainly EGUs). Year specific mobile emissions were derived from EPA's Motor Vehicle Emission Simulator (MOVES; www.epa.gov/otaq/models/moves) version 2010b and the Biogenic Emissions Inventory System (BEIS) version 3.1.4 was used for inline biogenics (Carlton and Baker, 2011). Emission inputs also included inline NO produced from lightning using year specific flash rate data from the National Lightning Detection Network (Allen et al., 2012). Boundary conditions were based on 2005 monthly median values from a GEOS-Chem version 9-01-02 simulation (http://wiki.seas.harvard.edu/geos-chem/) using version 8-02-01 chemistry, GEOS-5 meteorology and ICOADS shipping emissions (Henderson et al., 2014).

Ozone observations from 444 monitoring stations from the EPA's Air Quality System (AQS; http://www.epa.gov/ttn/airs/airsaqs) and 42 sites from the Clean Air Status and Trend Network (CASTNET; http://www.epa.gov/castnet) were used to evaluate model predicted ozone for the 21 states, including DC, that took part in the NO_x SIP Call rule. In addition, NO_2 observations from 45 urban AQS sites were used to diagnose potential errors in emission inputs. A map of the eastern US domain that identifies the NO_x SIP Call states and a map of the AQS and CASTNET monitor locations are shown in Supplemental Fig. S1. The model metric used for attainment demonstrations is based on an average of high summertime max 8-hr average ozone days (MDA8 O_3). Here we focus on the average of the top ten summer MDA8 O_3 values (top 10% of days in the simulation). A comparison of different metrics for ozone attainment demonstrations is discussed in Part II of this study. In Section 3 we discuss the updates in the CMAQv5.0.1 system related to dynamic evaluation of ozone, including changes to the meteorological and emission inputs compared to a previous version. Section 4 presents the development and analysis of the cross simulations, followed by a discussion in Section 5.

3. Evaluation of CMAQv5.0.1 simulations for 2002 and 2005

Following the results from the dynamic evaluation of the CMAQv4.6 system, several hypotheses were proposed to explain the underestimation of the change in ozone during this time period (Pierce et al., 2010; Godowitch et al., 2011). Underestimation of nocturnal low level jet wind speeds in the mid-Atlantic were

shown to lead to spatial displacements of peak ozone (Godowitch et al., 2011). Updated data assimilation in the WRF meteorological inputs as described in Gilliam et al. (2012) were used in the current simulations and have been shown to improve ozone transport in the Northeast US (Godowitch et al., 2014). In addition, mobile emissions in the new simulation were estimated using the new mobile emissions model MOVES which incorporates the latest emissions data and more sophisticated algorithms compared to the previous mobile emissions model, Mobile6 (Vallamsundar and Lin, 2011). NO_x emissions from both light- and heavy-duty trucks were found to be underestimated in the previous mobile emissions model and the new estimates are now significantly higher (CRC, 2010). Table 1 shows NO_x emissions totals for June–August 2002 and 2005 by sector for the same eastern US domain that was used in Gilliland et al. (2008) and shown in Fig. 2. NO_x mobile emissions increased by 33% while non-road emissions decreased by ≈ 10% in both years compared to Gilliland et al. Total NO_x emissions increased by roughly 10% in both years. However the percent change in total emissions is within 1% of the summary reported in Gilliland et al. Table 1. Nonetheless, as discussed further below, the increase in NO_x emissions may have affected the chemical regime for ozone production.

In terms of chemistry issues, updates to the CMAQ photolysis algorithm incorporated effects of aerosol loading on photolysis rates and year-specific satellite-derived ozone column data, which together led to decreased ozone in urban areas compared to the previous model version. Alternate chemical mechanisms were also explored to investigate specific chemistry issues that could potentially account for the lack of response in the modeled ozone values to NO_x emissions (Gilliland et al., 2008). At the time of the simulations, no alternate mechanisms were found to improve upon the standard configurations of the CB05 mechanism (Henderson et al., 2011). Finally, in contrast to the coarse vertical resolution used in the v4.6 simulations (14 layers with a 40 m surface layer) the current simulations used 35 vertical layers with a 20 m surface layer.

As a result of all of the changes to the modeling system from v4.6 to v5.0.1, ozone increased an average of 4% (≈2 ppb) across the eastern US in both 2002 and 2005, although there was localized decreased ozone in several urban areas. This net increase was primarily driven by the switch from MM5 to WRF meteorology (Appel et al., 2010), a new modeled NO_x source aloft through the inclusion of NO from lighting, and the use of GEOS-Chem boundary conditions rather than fixed profiles. The increased NO_x emissions from MOVES increased ozone across the region as well, but also contributed to the decreased ozone in urban areas. Overall, the increase in ozone improved the median bias in the average of the top ten MDA8 ozone values for both years (see Fig. 1a). The RMSE for the v5.0.1 predictions was lower than the v4.6 predictions in 2002 (10.5 ppb versus 11.7 ppb) and slightly worse in 2005 (7.7 ppb versus 7.5 ppb). A key issue highlighted by the boxplots in Fig. 1a is that the difference in the bias across these years impacts the dynamic evaluation of both models. The modeling system would be better able to capture the change in ozone if the bias was consistent across these summers. Instead the high summertime ozone levels in the base year summer were underestimated more than the high summertime ozone levels after the controls were in place.

Fig. 2 shows the observed and modeled 2005–2002 change in the average of the top 10 MDA8 ozone values (ppb). The CMAQv5.0.1 simulations predicted a larger decrease in ozone for much of West Virginia, Virginia, North Carolina and South Carolina compared to the v4.6 simulations. However the new simulation did not predict the 15–25 ppb decrease in observed ozone along the eastern coast. In addition, the location of the largest ozone decreases in the Ohio River Valley in the v5.0.1 simulation were different from the v4.6 simulation results. Fig. 1b provides the observed, v4.6 predicted, and v5.0.1 predicted change in the top ten MDA8 ozone days at rural (CASTNET and AQS), suburban (AQS) and urban (AQS) monitoring locations within the 21 NO_x SIP Call states. (Monitor locations and types are shown in Supplemental Fig. S1.) At rural locations, particularly the CASTNET sites, the v5.0.1 predicted change was somewhat closer to the distribution of the observed change than the v4.6 predicted change. At the suburban and urban AQS sites, the distributions of the v4.6 predicted change was closer to the observed distributions than the v5.0.1 predictions. A possible explanation for the difference in urban and rural performance is that the increase in mobile source NO_x from MOVES made some urban areas more VOC limited, reducing the modeled impact of the NO_x controls at urban sites in the new simulation compared to the predictions using the MOBILE6 emissions in the v4.6 simulation.

To diagnose potential errors in emission inputs we investigated morning NO_2 mixing ratios which have been shown to be strongly related to ground level NO_x emission levels (e.g., Godowitch et al., 2010). Fig. 3 shows the modeled minus observed bias and the observed and modeled change in 2005–2002 morning and daytime NO_2 levels at 45 AQS urban monitoring locations within NO_x SIP Call States. The v5.0.1 predicted decrease in NO_2 mixing ratios underestimated the observed change in early morning hours (Fig. 3b). Fig. 3a shows that morning (6 am–9 am) NO_2 levels were over estimated by the model, with the bias increasing in 2005 compared to 2002. This increase in the overestimation of 2005 NO_2 levels is likely not driven by problems with meteorological processes, such as too little vertical mixing due to incorrect boundary layer height. Misspecification of the boundary layer height would be expected to produce similar bias in both years. During the rest of the day when there is greater vertical mixing and stronger horizontal transport, the model and observed values were much more similar, suggesting that chemistry and transport are not the main causes of the NO_2 discrepancies. This supports the hypothesis in Godowitch et al. (2010) that the underestimation of the change in morning NO_2 levels could be due to an underestimation in the decrease in surface NO_x sources.

Table 1
Domain-wide NO_x emission estimate summaries (ktons) for the eastern US domain shown in Fig. 2 for June, July and August, 2002 and 2005. These sums only include the major emission sectors listed here. Other sectors that are less than 5% of the total emissions are included in the emission inputs to CMAQ but not in the table. Emission totals and the % difference from Gilliland et al. (2008). Table 1 are included in parentheses.

	EGUs (CEM,ptday)	Mobile (onroad)	Non-EGUs (ptnonipm)	Non-road[a]	Total
Summer 2002 NEI	853 (853)	1531 (1148)	348 (373)	671 (734)	3403 (3108)
Summer 2005 NEI	504 (493)	1214 (911)	329 (370)	646 (722)	2693 (2496)
% Difference (2005–2002)/2002	−41% (−42%)	−21% (−21%)	−5% (−1%)	−4% (−2%)	−21% (−20%)

[a] Non-road included emissions from the ALM (aircraft/locomotive/marine) sector to be consistent with Gilliland et al. (2008) Table 1.

4. Development and analysis of cross simulations

Four additional sensitivity simulations were developed to decompose the model predicted change in ozone into the change due to emissions and the change due to meteorology. As part of this process, a method was developed to adjust CEMS data to account for differences in electricity demand driven by year-specific meteorology. New base year simulations for 2002 and 2005 (Sim02e02m, Sim05e05m) were created that only used CEMS data from point sources that had continuous hourly data available for

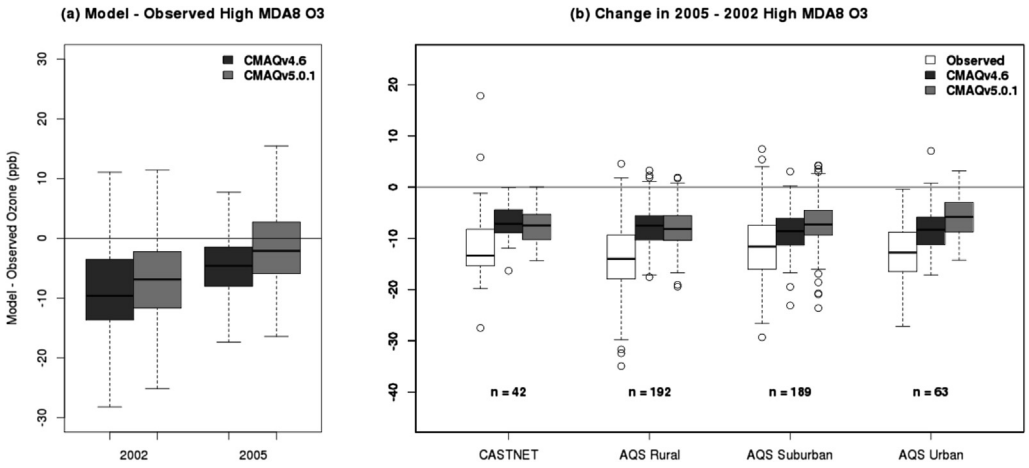

Fig. 1. (a) Model − observed bias in the average of the top 10 MDA8 O_3 values (ppb) at 486 AQS and CASTNET monitors within the NO_x SIP Call states for CMAQv4.6 (dark gray) and CMAQv5.0 (light gray) predictions; (b) observed and modeled change in 2005–2002 average of the top 10 MDA8 O_3 values (ppb) at CASTNET sites and at AQS sites grouped by rural/suburban/urban designations. The boxes indicate the 25th, 50th and 75th percentiles of the data, while the whiskers extend to 1.5 times the interquartile range of the data. Data points falling beyond the whiskers are shown as open circles.

both years. The matched units accounted for more than 96% of the tons per year of NO_x and SO_2 measured with CEMS in both 2002 and 2005. The remaining sources with CEMS data were re-categorized as non-CEMS point sources (i.e. ptnonipm). The ozone predictions for Sim02e02 m and Sim05e05 m were very similar (within ±1%) to the simulations used for evaluation in the previous section. In addition to the new 2002 and 2005 simulations, two "cross" sensitivity simulations were used to simulate air quality under 2005 emissions with 2002 meteorology (Sim05e02m) and 2002 emissions with 2005 meteorology (Sim02e05m).

The processing of emissions from EGUs with available CEMS data was based on unit specific adjustments of the emissions to account for the impact of different meteorological influences in a different year. Summertime 2005 NO_x emissions were generally lower than 2002 emissions due to the implementation of the NO_x SIP Call, but the temporal fluctuations were different due to differences in electricity demand which is heavily influenced by year-specific meteorology (see Supplemental Fig. S2). For example, applying 2002 CEMS data from a very cool wet day in 2002 that had hot stagnant conditions in 2005 would result in very low NO_x emissions being used on a high demand day. To more accurately estimate 2002 emissions with 2005 meteorological patterns for a particular EGU_i, ($EMIS02e05m_i$) the hourly 2005 CEMS emissions ($CEMS2005_i$) were scaled based on the ratio of summer total CEMS emissions from that unit ($SY1_i/SY2_i$) in 2002 versus 2005:

$$EMIS02e05m_i = CEMS2005_i \times (S2002_i/S2005_i) \quad (1)$$

An analogous calculation was made to estimate 2005 emissions with 2002 meteorological patterns. Fig. 4 shows daily total NO_x for the continental US based on the original CEMS data and the meteorology-adjusted data for 2002 and 2005. Summertime change in NO_x emissions from 2002 to 2005 was driven by NO_x SIP Call emission reductions rather than by changes in meteorology (i.e. the two blue lines and two green lines track each other closely throughout the summer). In contrast, peak NO_x emissions in mid-to late January were driven by colder temperature in 2005 rather than by seasonal emission totals (i.e. the dark blue dashed line and solid light green line track each other in January).

Careful attention was also given in creating the emission inputs for the remaining emission sectors. Emissions from point sources with no CEMS data (including the unmatched units that did not have data for both 2002 and 2005) were based on annual total emissions from the NEI that were scaled to daily totals using ratios based on average CEMS data within a state associated with the meteorology year. In this way, the seasonal emissions totals corresponded to the NEI ptnonipm emissions of the emissions year, while the daily temporal fluctuations followed the pattern of the point sources with CEMS data during the meteorology year. Additional details on the calculation of the scaling ratios is provided in Supplemental material S3.

Mobile emissions for the cross runs were based on MOVES simulations using emission factors and activity data from the designated emission year with meteorological inputs based on the meteorology year (e.g. 2002 emission factors and activity with 2005 meteorology). Mobile source emissions are sensitive to meteorology since increased temperature will increase evaporative VOC emissions and NO_x emissions factors in MOVES are also temperature dependent. Emissions from nonroad (e.g. construction), industrial point and commercial marine sectors were based on the emissions year but shifted to match the day-of-the week and the holidays of the meteorology year. Emissions from fertilizer application, biogenic sources, fires, dust and NO produced from lightning were tied to the meteorological year. All other sectors had the same inventory for all four simulations, except modified for the day-of-the-week and the holidays of the meteorology year. Thus the cross simulations were able to capture the impact of meteorology on ozone levels through direct effects such as advection and dispersion, and through indirect meteorological effects on emissions such as the effect of temperature on electricity demand or evaporative emissions from motor vehicles.

Table 2 shows NO_x emissions totals from EGUs and mobile sources for June–August 2002 and 2005 for the eastern US domain for the four sensitivity simulations. Note mobile emissions for Sim02e02 m and Sim05e05 m are the same as what was shown in Table 1 and EGU emissions for these runs are very similar to Table 1 results as discussed earlier in the section. Meteorological conditions in 2005 compared to 2002 did not change the seasonal totals from the mobile and EGU sectors in any substantial manner. The large decrease in total emissions from these sectors can be attributed directly to the emission controls that occurred during this time period.

Fig. 5 shows the impact of the emissions and meteorology changes on ozone across the domain. The top row of Fig. 5 is the

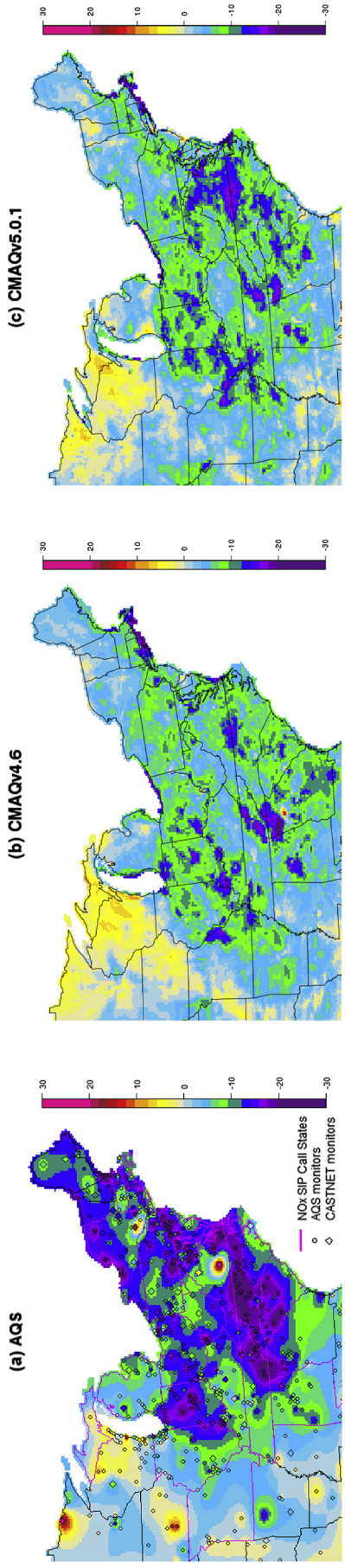

Fig. 2. 2005–2002 change in the average of the top 10 MDA8 ozone values (ppb) based on (a) AQS and CASTNET observations (b) CMAQv4.6 simulations and (c) CMAQv5.0.1 simulations. Note that the interpolated field underlying the ozone observations in (a) is not intended to represent the "true" observed ozone field. The interpolation (created through universal kriging) is simply provided to help the reader better distinguish the spatial patterns at the clusters of AQS and CASTNET sites.

2005–2002 change in the top 10 MDA8 ozone days based on (a) the change in emissions with 2002 meteorology (Sim05e02m − Sim02e02m) and (b) the change in meteorology with 2002 emissions (Sim02e05m − Sim02e02m). As expected, emissions reductions led to a homogenous regional decrease in ozone, with the largest decreases in the Ohio River Valley and mid-Atlantic states. Changes in ozone due to meteorology are spatially much more heterogenous with many areas showing an increase in ozone from 2002 to 2005.

Fig. 5c shows the change in ozone that is not explained by the individual changes in emissions or meteorology represented in the cross simulations. Specifically this is calculated by summing the values in 5(a) and 5(b) and then subtracting this from the total change in high summertime ozone shown in Fig. 5d. This left-over term reflects any nonlinear interactions between meteorology and emissions which were not accounted for in the cross simulations. For example, differences in how temperature affects ozone chemistry under different emissions loadings would not be included since the meteorology change in Fig. 5b is based on fixed 2002 emission inputs. This additional interaction term had a very small impact on ozone levels in this application (±2 ppb).

Fig. 5d shows the total change in high summertime ozone (Sim05e05 m − Sim02e02 m), which, by definition, is equivalent to the sum of (a), (b) and (c). We see how much the meteorological-driven change in (b) influences the final spatial pattern, changing the locations with the largest ozone decrease compared to the emissions-only response represented in (a). As stated above, these two summers were very similar in terms of the number of days with ozone-conducive meteorology versus the number of days with less favorable conditions for ozone formation. Even after selecting two summers that had similar meteorological conditions, the modeled change in meteorology resulted in changes in ozone levels up to ±15 ppb. This highlights the advantage of using a modeling approach such as described here to be able to remove the impact of meteorology when quantifying and evaluating the net impact of retrospective emission controls. It also highlights the difficulty in performing dynamic evaluation studies aimed at isolating the effect of emission reductions given the non-negligible effect of meteorological variability.

Thus far we have focused on the upper end of the ozone distribution. Fig. 6 shows the change in MDA8 ozone (ppb) by percentile. Each boxplot shows the distribution of the change in ozone at the given percentile across the 444 AQS sites. The 2005–2002 difference in the observed ozone levels becomes steadily more negative with increasing percentile. The modeled difference follows the same trend but does not show the change as steeply at the upper percentiles. The decomposition of the total modeled change into the emissions, meteorology and remaining interaction terms are also included for each percentile. At the upper end of the modeled ozone distribution, meteorological conditions in 2005 compared to 2002 tend to mostly decrease ozone in the NO_x SIP Call states. However, the average decrease in ozone attributed to emission reductions is more than twice as large as the decrease attributable to changes in meteorology alone. The decrease in the lower end of the ozone distribution ($\leq$10th percentile) is driven almost completely by meteorology changes.

Ideally we would like to have an observation-based decomposition of the change in the ozone in order to better diagnose if the differences in the observed and modeled total ozone change are influenced more by the emissions or the meteorological inputs. Camalier et al. (2007) provided an approach for estimating meteorologically adjusted observed ozone trends based on a generalized linear model (GLM) of ozone using a set of meteorological parameters, including multiple parameters for observed temperature, wind, humidity, pressure, stability, 24 h transport trajectories and

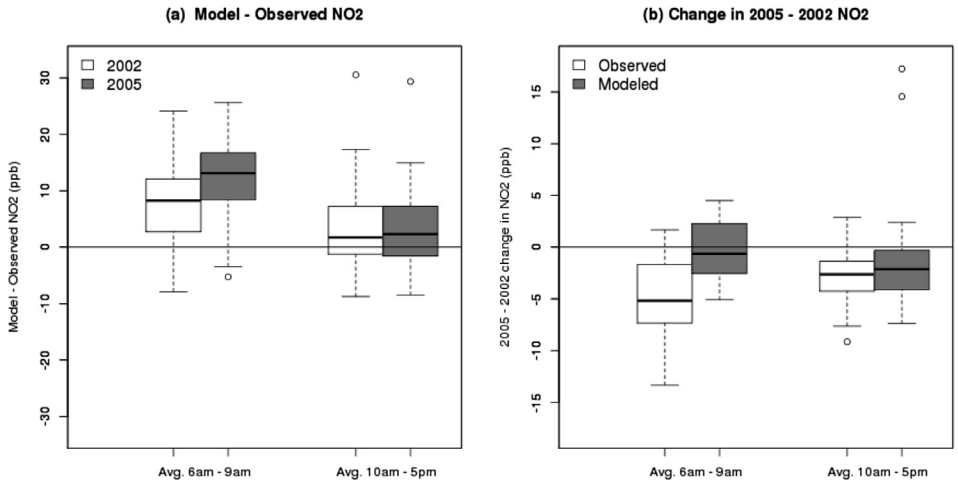

Fig. 3. (a) Model − observed NO_2 bias (ppb) and (b) observed and modeled change in 2005–2002 NO_2 (ppb) for morning and daytime hours at 45 urban AQS sites within NOx SIP Call States. Model predicted values are from the v5.0.1 simulations. The boxes indicate the 25th, 50th and 75th percentiles of the data, while the whiskers extend to 1.5 times the interquartile range of the data. Data points falling beyond the whiskers are shown as open circles.

synoptic weather conditions. The magnitude of the meteorology adjustment can then be estimated by calculating the difference between the raw and met-adjusted ozone values at a given site. Maps of the annual met adjustments for 2000 to 2012 are available online (http://www.epa.gov/airtrends/weather.html).

The data from this statistical approach can be used as a way to evaluate the model-based estimates for the change in ozone due to meteorology by subtracting the estimated meteorology adjustment in 2002 from the estimated adjustment in 2005 (e.g. [Raw 2005 − Adj. 2005] − [Raw 2002 − Adj 2002]). The meteorologically-adjusted observations were provided at a limited set of AQS and CASTNET stations (see Fig. S1). The statistical model was estimated using ozone data from May through September and provided information on the mean concentrations at each site. Thus far our analysis has focused on June through August ozone data in order to be consistent with Gilliland et al. (2008). Model simulation output was available for September 2002 and 2005 and this additional month is included in the following comparison. Similar to the 50th percentile portion of Fig. 6, Fig. 7 shows the observed and modeled change in the summer (June–September) mean ozone across the AQS and CASTNET sites. However, Fig. 7 also includes the estimated change in observed ozone attributed to meteorology (shown in the dark green box plot and calculated over the May–September period). The model and observed distribution of the change in ozone attributed to meteorology are similar, showing both increases and decreases in ozone within approximately 5 ppb. The spatial distribution of these changes are provided in Supplemental Fig. S4. This comparison again suggests that the observed decrease in ozone across these summers is primarily driven by emissions changes and that these changes are not fully captured by the modeled emission inputs (cf., Zhou et al., 2013; Kang et al., 2013). However, there are some differences between the two maps in Supplemental Fig. S4, suggesting that additional diagnostic work aimed at quantifying and evaluating the effects of changing meteorology on simulated ozone concentrations is warranted. Furthermore, an extension of the GLM approach to provide met-adjusted ozone for different percentiles is needed to more fully evaluate the results in Fig. 6.

5. Discussion

The system updates in the CMAQv5.0.1 simulations improved the median bias in high summertime ozone predictions and led to larger predicted ozone decreases from 2002 to 2005 in mostly rural areas but less ozone decreases in more urban locations, compared to v4.6 simulations. While the meteorological, emissions and chemistry changes in the CMAQv5.0.1 system had a substantial impact on the spatial pattern of the change in ozone, there remain observed ozone decreases that the model did not fully capture. Additional model developments suggested in Pierce et al. (2010) (e.g. increased horizontal grid resolution, more realistic boundary conditions, improvements in surface NO_x emissions in urban areas) could lead to improvements in the dynamic evaluation of the modeling system. Recall that Fig. 2b showed that model performance was worse at more urban locations compared to surrounding suburban and rural sites. Using finer horizontal grid resolution could better resolve urban gradients in locations where the model tends to predict ozone increases from 2005 to 2002 due to NO_x titration over too large of a spatial area.

Year specific hourly boundary conditions could impact the model predicted ozone response particularly in coastal areas in the northeast and the Gulf where seasonal mean ozone has been shown to be most heavily influenced by the boundary conditions for ozone (Napelenok et al., 2011). Monthly median boundary conditions from a 2005 GEOS-Chem simulation were used here because appropriate meteorological inputs for the global model were not available for 2002 at the time of simulation. Sensitivity simulations showed that using monthly medians in 2005 rather than the original hourly boundary conditions did increase high summertime ozone by 0.5–1.5 ppb along the northeastern and Gulf coasts, although it is unclear if this had any impact on the predicted change in ozone across the two summers. Analysis of May–September average mid-tropospheric ozone mixing ratios from a recently completed WRF-CMAQ simulation over the northern hemisphere (Xing et al., 2014) show both positive and negative differences on the order of a few ppb between 2002 and 2005 in regions where inflow would be expected to influence the regional-scale simulations analyzed in this study. Future research is needed to quantify the impact of inter-annual variability in large-scale background mixing ratios on the dynamic evaluation of regional-scale simulations. However, an analysis of recent WRF-CMAQ simulations performed for 2006 and 2010 utilizing year-specific boundary conditions showed qualitatively similar results to the ones presented in this study for the comparison of observed and modeled high ozone mixing ratios between the two years (Hogrefe et al., 2014), suggesting that the choice of boundary conditions in

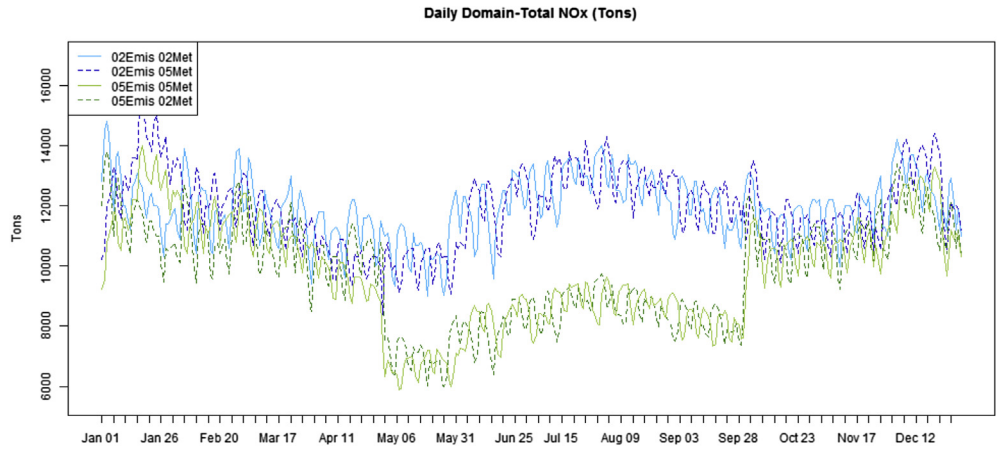

Fig. 4. Daily total NOx (tons) from CEMS data for 2002 (blue) and 2005 (green) across the continental US. The original CEM totals are shown as solid lines. The meteorology-adjusted CEMS totals are shown as dashed lines: 2002 emissions with 2005 meteorology (dashed blue) and 2005 emissions with 2002 meteorology (dashed green). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 2
Domain-wide NO_x emission estimate summaries (ktons) for June, July and August, 2002 and 2005 for four cross simulations. The percent change from the 2002 emissions levels is shown in parenthesis for each simulation.

	EGUs (CEM,ptday)	Mobile (onroad)
2002 emissions, 2002 meteorology	848	1531
2002 emissions, 2005 meteorology	851 (+0.4%)	1589 (+4%)
2005 emissions, 2002 meteorology	480 (−43%)	1255 (−18%)
2005 emissions, 2005 meteorology	487 (−43%)	1214 (−21%)

the current study may not be the main contributor to the underestimated model response.

Emissions clearly have a large influence on the ability of the modeling system to reproduce observed air quality trends, however fully diagnosing errors in the emissions inputs remains very challenging. For example, the increase in MOVES mobile source NO_x emissions relative to MOBILE6 may have been too strong, thus shifting the photochemical regime towards VOC-limited, making the modeling system less responsive to NO_x controls. While it is

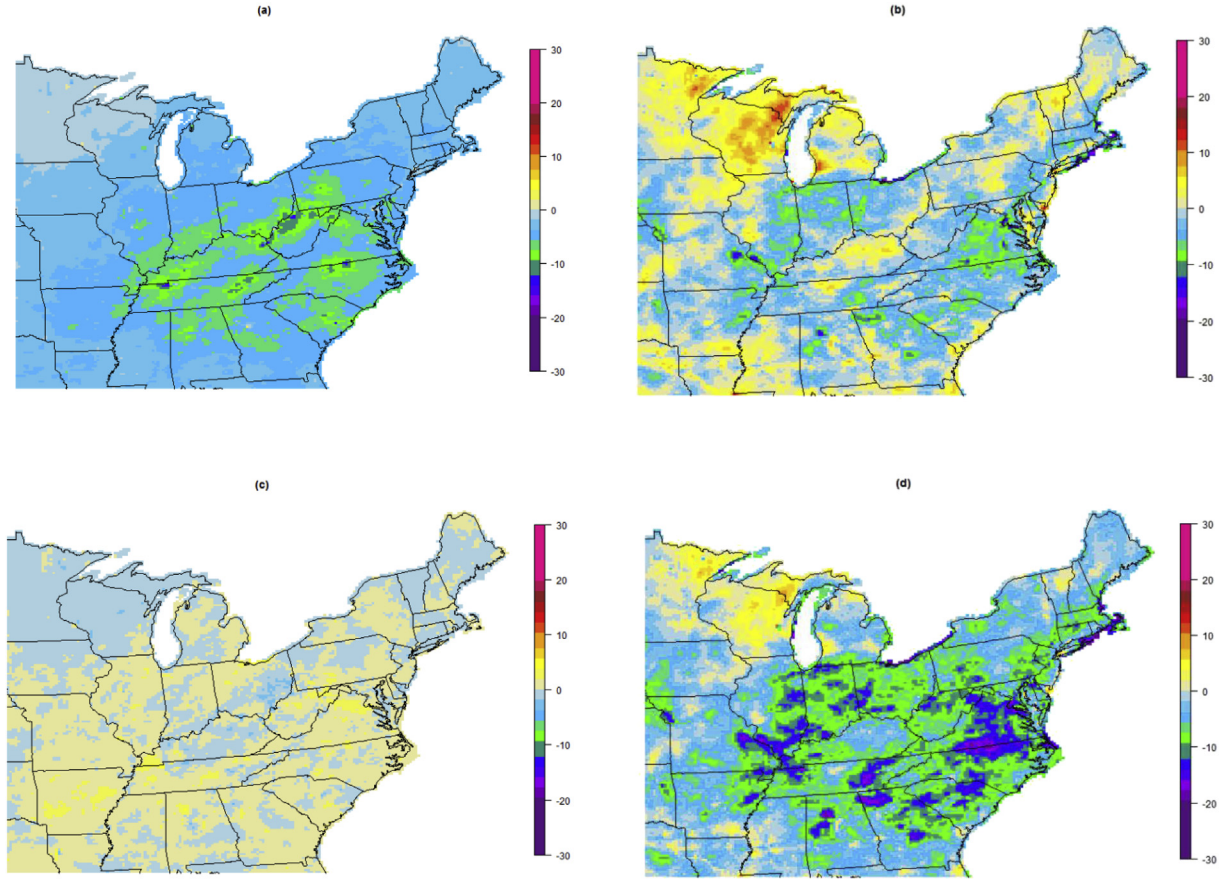

Fig. 5. The 2005–2002 change in high summertime ozone due to (a) the change in emissions (with 2002 meteorology), (b) the change in meteorology (with 2002 emissions), (c) the change not explained by the emissions or meteorology changes represented in the cross simulations and (d) the total modeled change across the two summers.

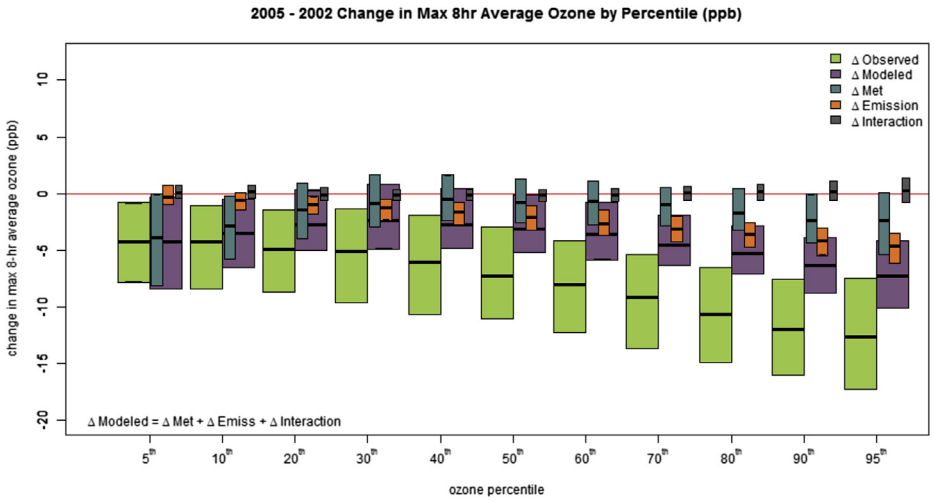

Fig. 6. 2005–2002 change in MDA8 ozone (ppb) by percentile at 444 AQS monitoring stations. The observed change is shown in green. The modeled change (purple outline) is decomposed into the change due to emissions (orange), meteorology (blue), and the remaining change that is not explained by the emissions or meteorology components (gray). The boxes indicate the 25th, 50th and 75th percentiles of the data. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

beyond the scope of this study whether the photochemical regime simulated with MOBILE6 or MOVES is more realistic, the emission study of a traffic tunnel in CA presented in Fujita et al. (2012) suggested that MOVES NO_x emissions may indeed be overestimated. Future work is needed to test this hypothesis and its impact on dynamic evaluation. Finally, as discussed in Section 3, downward trends in surface NO_x emissions may be underestimated, affecting both the chemical regime and the dynamic response of the modeling system.

This study demonstrated how dynamic evaluation studies can be enhanced by better quantifying and accounting for the effects of interannual meteorological variability. One avenue for accomplishing this goal is on-going work directed at analyzing decadal scale model simulations to determine whether the underestimation of observed changes between 2002 and 2005 is specific to these particular years or reflects a more fundamental issue with the modeling system or available emission inputs. Additional on-going work for this type of dynamic evaluation includes an extension of the statistical model presented in Section 4 that uses additional meteorological parameters and applies quantile regression methods in order to estimate observation-based meteorology-adjusted ozone for different percentiles, rather than just seasonal means. Such an approach would allow for further diagnostic evaluation of the model predicted change in ozone that can be attributed to changes in meteorology. The role of dynamic evaluation should be to gauge the modeling system's ability to capture observed changes in air quality, and to help guide model improvement and direct resources to areas needed for additional data collection.

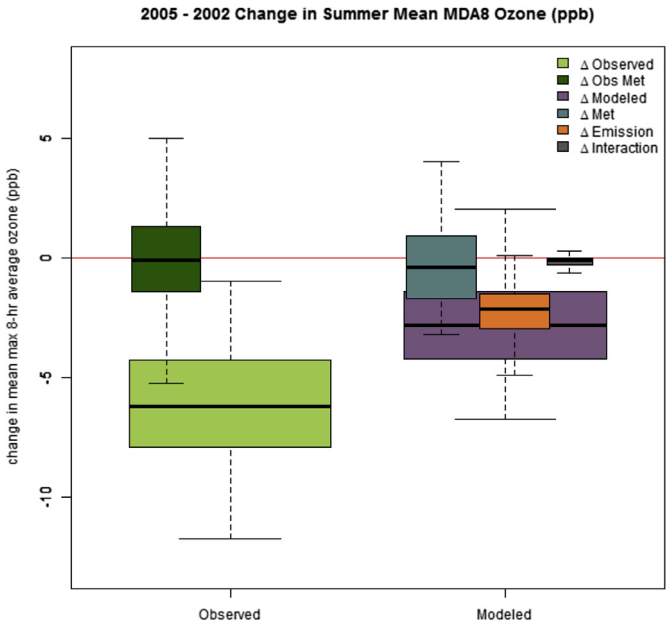

Fig. 7. 2005–2002 change in mean MDA8 ozone (ppb) at 53 AQS and 7 CASTNET locations. Note that the met-adjusted observations are based on the mean over May through September of each year. The other boxplots are based on June through September averages. The boxes indicate the 25th, 50th and 75th percentiles of the data, while the whiskers extend to 1.5 times the interquartile range of the data.

Disclaimer

Although this work was reviewed by EPA and approved for publication, it may not necessarily reflect official Agency policy.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2014.12.038.

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