Investigating the Impact on Modeled Ozone Concentrations Using Meteorological Fields
From WRF With an Updated Four-Dimensional Data Assimilation Approach

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ABSTRACT

The four-dimensional data assimilation (FDDA) technique in the Weather Research and Forecasting (WRF) meteorological model has recently undergone an important update from the original version. Previous evaluation results have demonstrated that the updated FDDA approach in WRF provides more accurate wind fields aloft than the original approach, particularly during the nocturnal period when low level jets are a common feature in the eastern United States. Due to the importance of WRF/FDDA meteorological fields in retrospective air quality applications, a modeling study with the Community Multiscale Air Quality (CMAQ) model was undertaken to ascertain if the improved wind flow fields translate into better performance for ozone. To undertake this objective, separate CMAQ model simulations were performed with meteorological inputs generated by WRF using the original and the updated FDDA approaches for a three month summer period. The evaluation effort focused on observed and modeled surface ozone from a mid-morning hour (10 LDT). Comparisons of modeled results against concentrations aloft from an instrumented tall tower and from available morning vertical profile measurements were also examined. Surface concentrations near 10 LDT are desirable for evaluating the transport process since they are often representative of ozone that has been transported aloft overnight and has undergone downward entrainment in response to convective mixing the following morning. Statistical results from surface observed and modeled concentration pairs indicated modeled ozone from the CMAQ simulation using the updated FDDA meteorology displayed smaller biases and lower absolute errors at 88% and 80% of monitoring sites, respectively, in the eastern United States. The CMAQ results with the updated FDDA generally exhibited smaller biases and lower absolute errors at monitoring sites across the northern states than in the southeastern states. The results provide evidence that the more
accurate wind flows generated with the updated WRF/FDDA approach improved CMAQ model performance based on the statistical results from 10 LDT ozone concentrations.

**Keywords:** four-dimensional data assimilation, ozone model evaluation, horizontal transport, WRF, CMAQ

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1. Introduction

The horizontal transport process strongly governs the spatial ozone pattern and its temporal variability in air quality model simulations. Consequently, accurate three-dimensional (3-D) wind fields over the entire diurnal cycle are critical to realistically simulating the horizontal distribution of ozone on regional scales. Modeled wind speed and/or direction errors cause increasingly larger spatial displacements in modeled ozone, which negatively impact model performance by contributing to the scatter found between modeled and observed ozone concentration pairs in evaluation studies. The use of four-dimensional data assimilation (FDDA) in dynamic meteorological modeling has greatly improved the characterization of modeled wind flows and other meteorological parameter fields in the lower troposphere for retrospective air quality modeling applications (Otte, 2008a,b). The FDDA technique, or Newtonian nudging as applied in a meteorological model simulation, continuously adjusts the modeled state variables of wind, temperature, and moisture toward 3-D model analysis fields modified with available observations and greatly reduces the accumulation of model error during the course of a simulation (Seaman, 2000).

Developing analysis fields of winds and other meteorological variables for FDDA has recently evolved as more observational data sets, especially from remote-sensing systems, have become readily accessible (Gilliam et al, 2012). After Godowitch et al. (2011) found that wind speeds aloft, particularly in the nocturnal low level jet and overlying residual layer were underestimated in comparisons to wind profiler observations in the eastern US during nighttime hours based on meteorological simulations with the existing FDDA technique, a concerted model testing and evaluation effort was undertaken to develop an improved FDDA approach.
that would generate more accurate 3-D wind fields for characterizing flows aloft during the nighttime and post-sunrise periods. Based on extensive testing and evaluation with the Weather Research and Forecasting (WRF) model, an updated FDDA approach that took advantage of additional archived observational winds obtained by different measurement platforms, as well as a key revision in the FDDA method, was adopted (Gilliam et al., 2012). In particular, wind flow errors at different heights at night with the updated FDDA technique were reduced compared to the original FDDA approach. Simulating the development and evolution of the nocturnal low level jet, a frequent feature from the mid-Atlantic region into New England in the summer (Zhang et al., 2006), is important since it can serve as a mechanism for transporting pollutants hundreds of kilometers during the nocturnal period. Realistic modeling of the nocturnal evolution of the wind field in the overlying residual layer of the lower troposphere is also essential to accurately simulating the horizontal transport of ozone and other pollutants in parts of the eastern United States during the summer season.

After the adoption of the updated WRF/FDDA technique, a follow up modeling study applying the Community Multiscale Air Quality (CMAQ) model was necessary to investigate whether improvements in simulated wind fields also translate into better model performance for ozone concentrations. Therefore, photochemical simulations with the CMAQ model using meteorological fields generated using WRF/FDDA with the updated and original approaches were performed for a three-month summer 2002 period. Other model inputs, including emissions, boundary conditions, and the model configuration remained the same in both modeling scenarios. Results will consist of statistical metrics and various analyses of modeled and observed surface ozone (O$_3$) concentration pairs from a mid-morning hour (10 local daylight time (LDT)). At morning hour, it will be demonstrated that surface ozone levels reflect the magnitudes of ozone found aloft which have been subjected to overnight transport in the eastern United States (US). Hence, our hypothesis is that more accurate wind fields, that provide a more representative characterization of the horizontal ozone distribution aloft in the region, should produce improved CMAQ performance for ozone in statistical measures determined with modeled and observed concentration pairs from this mid-morning hour. Further information about the rationale for selecting this time to assess model results is provided in section 4. The summer of 2002 was selected since additional ozone measurements aloft were also available from tower and aircraft profile measurements and a broad range of O$_3$ concentrations occurred during this summer period including several high ozone episodes (Godowitch et al, 2011). Additionally, testing results in Gilliam et al. (2012) revealed the updated
FDDA approach performed better than others for the summer 2002 episode. Comparisons of modeled and observed O₃ profiles from selected experimental case studies during the summer of 2002 are also examined to provide evidence in distinguishing between the ozone performance of the two model simulations.

2. Model Description and Simulation Details

The CMAQ chemical transport model (version 5.0.1) with the updated Carbon Bond (CB05TU) chemical mechanism including toluene chemistry (Whitten et al., 2010) was applied in the simulations for this study. Other key process components of the CMAQ model base configuration included the Asymmetric Convective Model version 2 (ACM2) vertical mixing scheme, the Pleim-Xiu (PX) land surface model, piece-wise parabolic (PPM) horizontal advection method, and deposition velocity approach for dry deposition (Byun and Schere, 2006).

The modeling domain extended beyond the continental United States and contained a 458 X 299 horizontal grid with a 12-km grid cell size. There were 35 vertical layers from the surface to 50 mb with 13 layers below 1 km. The thickness of layer 1 was ≈ 20 m. The model simulations spanned the three-month period from June 1 through August 31, 2002. A 10-day spin-up period prior to June 1 was also simulated to minimize the effect of initial conditions. The lateral boundary conditions were prescribed by concentrations generated from a Goddard Earth Observing System global chemistry (GEOS-Chem) model simulation. The CMAQ model configuration and these inputs were the same for both modeling scenarios.

Hourly meteorological parameter fields were generated by WRF version 3.3 with the same 12-km horizontal resolution as CMAQ. The same physics options as described in Gilliam et al. (2012) were applied in both WRF simulations with the exception of a different FDDA approach.

The WRF/FDDA technique continuously adjusts the modeled variables with 3-D model analysis fields archived from the initial conditions as well as 3-h forecasted fields of US weather forecast models (Gilliam and Pleim, 2010). The original FDDA technique that was applied for many years relied on routine hourly surface observations and the twice-daily rawinsonde profile measurements. These measurements were introduced into a WRF utility program which incorporated the observations and modified the 3-D analyses fields to produce a closer fit to the observations. In the original approach, surface 2-D analysis fields were also applied for
adjusting modeled winds with the weighting diminishing with height over the lowest model layers. Otte (2008a) and Gilliam et al. (2012) provide additional details about the original FDDA technique. The updated FDDA approach described in Gilliam et al. (2012) takes advantage of additional archived data sets from remotely sensing platforms, which include wind profiler measurements and WSR-88 Doppler radar wind data for use in the reanalysis procedure. Additionally, a revision in the FDDA technique involved the complete elimination of surface analysis nudging. Both FDDA approaches applied analysis nudging to the state variables in model layers above the PBL height during the entire diurnal cycle.

Since our CMAQ modeling study was performed independently (uncoupled mode) of the WRF runs, the Meteorology-Chemistry Interface Processor (MCIP) utility program was exercised with the WRF output in order to generate format-compatible meteorological data sets to drive the CMAQ simulations.

Hourly gridded emission data sets were generated by the Sparse Matrix Operator Kernel Emissions (SMOKEv2.2) processing system. Anthropogenic emissions were extracted from the U.S. EPA National Emissions Inventory (NEI) for 2002 to generate gridded surface area and minor point source emissions. The hourly pollutant emissions for elevated major point sources were specified from Continuous Emissions Monitoring System (CEMS) data sets. Gridded on-road vehicle emissions were generated by the MOBILE6.2 model. Natural surface emissions of NO_x, isoprene, and other biogenic VOC species were generated by the Biogenic Emissions Inventory System (BEISv3.14) model. Additional emissions from ship traffic and wildfires from both periods were also included. The same emission data sets were applied in both model simulations.

3. Measurements and Analysis Techniques

The surface ozone observations from the Air Quality System (AQS; USEPA (2002a)) network sites and the Clean Air Status and Trends Network (CASTNET; USEPA (2002b)) monitoring sites were matched in space and time with the modeled concentrations. While the CASTNET sites are found in agricultural and forested locations at considerable distances from cities, the AQS network sites are located in a variety of land uses environments within urban areas as well as rural locations of the United States. Specifically, the observed O_3 at 10 LDT from each site was paired against the 10 LDT hour-average modeled layer 1 concentration in the CMAQ ACONC file from the grid cell containing the monitoring site location. In addition, hourly ozone measurements made at 4 different heights (z = 3 m, 77 m, 149 m, 433 m AGL) on a TV tower
(i.e., WRAL-TV located near Raleigh, NC) were also available from the AQS data base since a
different AQS site identification number was assigned to each level. Modeled O$_3$ values from
layers containing these measurement heights were paired with the corresponding height-
specific hourly observations. Unfortunately, it was discovered that due to a problem with the
sampling tube the ozone data at the 433 m level was found to be unreliable.

Observed vertical O$_3$ profiles were obtained by a University of Maryland research aircraft at
selected small airport locations in the mid-Atlantic states during morning periods in conjunction
with an experimental field study during June and July 2002 (Castellanos et al., 2011). Details
about the aircraft instrumentation and sampling flights consisting of spiral ascents/descents are
discussed in Hains et al. (2008). The aircraft’s latitude/longitude coordinates and altitude in the
vertical spirals were used to match up observed O$_3$ values with model concentrations (i.e.,
CMAQ 3-D CONC file) from the appropriate grid cell and vertical layer. Due to the high
resolution of the measurements, all observations within each model layer were averaged and
the mean observed values were paired with the model’s layer-average concentrations that were
temporally interpolated to the time of the observed profile.

4. Results

4.1 Examination of Ozone Aloft

The time variations of mean observed ozone concentrations from two levels aloft and at the
surface from the WRAL-TV tower and modeled concentrations from vertical layers containing
the measurement heights are displayed in Figure 1. These observed and modeled results are
presented in separate figures since the intent is to depict the temporal behavior in the
observations and modeled concentrations aloft rather than to directly compare absolute
concentrations because the layer-average model values do not correspond to the same heights
as the observations. The results in Figure 1b reveal that the modeled mean values closely track
the temporal evolution in the observed O$_3$ in Figure 1a at each level. Concentrations exhibit a
gradual decline during the nocturnal hours after midnight, followed by a rapid rise that typically
occurs during the morning period. The decrease of ozone overnight is attributed to dry
deposition and titration with existing nitrogen oxide (NO). Weak vertical mixing within the
nocturnal stable boundary layer also causes the concentrations at other heights to be affected
by the near-surface loss processes. Once the convective mixing layer begins to grow after
sunrise, higher O$_3$ concentrations aloft are steadily mixed downward as the vertical mixing
process is a major contributor to the rapid increase in surface ozone concentrations during the
morning period (Zhang and Rao, 1999). The observed and modeled results both show that
surface $O_3$ rapidly increases and eventually attains the magnitude of concentrations at levels
aloft. The results indicate the concentrations at all levels are quite comparable by 10 LDT.
Around this time the nocturnal inversion layer has generally been eroded and the strong vertical
ozone gradient that had existed earlier has been eliminated. The upward extent of vertical
mixing finally reaches into the residual layer, where the vertical distribution of $O_3$ is generally
much less variable as will be noted later. Once the mixing into the residual layer occurs, the
rate of rise in surface $O_3$ concentrations tends to become more gradual and the role of
photochemical processes becomes more relevant in influencing the evolution of $O_3$
concentrations within the PBL.

(Insert Figure 1)

In contrast, vertical mixing within the well-mixed planetary boundary layer (PBL) declines in
the evening and winds aloft begin to accelerate which can transport the leftover $O_3$ contained
within the residual layer considerable distances during the nocturnal period. Vukovich and
Scarborough (2005) have given a thorough description of the nocturnal evolution of ozone
transport. During the summer months, the residual layer extends from the top height of the
surface-based nocturnal inversion layer (e.g., 300-500 m AGL) to the preceding day’s PBL
height (e.g., 1 500 - 2 500 m AGL). The ozone concentrations within the residual layer
generally exhibit little change during the nocturnal period and early morning hours, however, the
wind flow differences between these simulations is expected to produce notable spatial
displacements in the horizontal ozone pattern aloft. An example case in Figure 2a depicts the
modeled spatial $O_3$ pattern after an extended period of nocturnal transport. High $O_3$
concentrations aloft in the mid-morning (i.e., 10 LDT) are evident in various areas of the eastern
United States in these CMAQ results using the updated FDDA meteorology which were also
evident in the surface layer $1 O_3$ field by this time. In fact, notable high ozone plumes are
apparent in the mid-Atlantic (MA) states, Ohio River Valley (ORV) region and northeastern (NE)
states. Figure 2b shows notable $O_3$ differences at this time between the two model simulations
of up to $\pm 20$ ppb in the vicinity of the high concentration plumes where large horizontal
gradients exist in this case. A generally southwesterly (SW) wind flow, a common pattern
occurring in the eastern United States during summer (Godowitch et al., 2011), prescribed the
orientation of notable $O_3$ plumes. In particular, the higher $O_3$ areas and $O_3$ differences along
the NE coast and MA regions primarily exhibit the signatures of major urban plumes, while
major point sources in the ORV region (Godowitch et al., 2008) contribute to $O_3$ variations in the
vicinity of the ORV and further downwind into northern NE in this case. These spatial concentration differences were a common feature in these model simulations and these displacements are attributed to speed and direction variations in the wind flows generated by the original and updated FDDA approaches. It is apparent that in some areas in Figure 2b there are also small O₃ differences of a few ppb.

( Insert Figure 2 )

The lack of spatially-dense and temporally-resolved observed ozone profile measurements has greatly limited attempts to distinguish which ozone pattern is closer to reality. However, the results in Figure 3 give some evidence that the CMAQ O₃ aloft using the updated meteorology provides better overall agreement to the morning observed profile at this central Virginia site during this field study case than the modeled profile generated using the original FDDA meteorology, especially in the residual layer above about 500 m. Mean observed O₃ from the aircraft spirals and modeled profiles were also determined from 30 sites over 10 morning cases. Results in Figure 4a indicate the mean modeled profiles are quite similar to each other and both model results are very comparable to O₃ concentrations aloft in the observed mean profile. The small differences between the modeled mean ozone profiles from these simulations suggest that many of the aircraft profiles were obtained at sites in areas where ozone was rather spatially uniform, which does not help to distinguish between the model results. Figure 4a reveals that both modeled results overestimate observed mean values in the lowest few hundred meters, which will also be assessed from analysis of the surface observed MODELED pairs.

( Insert Figure 3 )

Additional results of grouping the modeled O₃ values between 500-1,500 m over 10 ppb intervals of observed O₃ are shown in Figure 4b. At lower observed concentrations, these modeled results tend to overestimate observed ozone, while at the highest concentrations the modeled results slightly underestimated observed values. Both modeled results appear to mimic evaluation results of afternoon maximum ozone concentrations at the surface (Appel et al. 2007; Mao et al., 2010), which might not be unexpected since ozone aloft in the morning can be traced back to the previous day's ozone levels. Figure 4b also indicates the modeled results using the updated FDDA meteorology exhibit slightly less variability at the higher concentration levels. Next, comparisons of ozone from 10 LDT from the broader spatial coverage and higher
density of surface monitoring sites are expected to provide a better opportunity to distinguish between the model performance of these simulations.

( Insert Figure 4 )

**4.2 Comparative Results of Modeled and Observed Surface Ozone Concentrations**

The statistical metrics shown in Table 1 were determined from observed and modeled 10 LDT ozone pairs from AQS and CASTNET monitoring sites located in the eastern United States over the 92 days of the modeling period. The results revealed that the model simulation using the updated FDDA meteorology exhibited better model performance with a smaller mean bias (MB) by about 20% and a lower mean absolute error (MAE) by about 8% than those for the model results using the original FDDA meteorology for each group of sites.

( Insert Table 1 )

An additional metric examined for this study was the frequency factor (Fp) defined as the percentage of cases that each model simulation value was closer to an observation than the other simulation result. To determine Fp, the absolute difference (|M – O|) between each modeled and observed O₃ concentration at 10 LDT was determined and the simulation result exhibiting the smaller absolute difference was selected. Table 1 reveals that the CMAQ results with the updated FDDA meteorology were more frequently closer to observations with an Fp of 58% versus only 42% for simulation results using the original FDDA meteorology. A typical case showing which model value was closer to the observed 10 LDT O₃ value at each AQS site is depicted in Figure S1. In this case, the modeled results using the updated FDDA meteorology more accurately simulated observed values at 62% of the sites and it’s modeled values were especially in better agreement at numerous sites along the northeast urban corridor stretching from Washington, DC to Connecticut. A notable result was that Fp for the updated FDDA simulation was higher on each day than Fp for the results using the original FDDA meteorology.

Since differences in the bulk statistical metrics were somewhat modest, further analysis was performed in an effort to identify more definitive differences from site-specific statistical metrics. A comparison of MAE between both simulations in Figure 5 reveals that the modeled results using the updated FDDA meteorology exhibited less error at over 80% of AQS sites with MAEs lower by 10% or more at sites where higher model errors existed. The spatial distribution of MAE from the simulation using the updated FDDA meteorology is shown in Figure S2. Lower
model errors generally occurred at sites in the northern states, while higher MAEs were more
often found in the southeastern states. This result is also borne out in Table 1 which indicates a
lower MAE for the AQS/NE site group than in the overall AQS/EUS sites that also contains the
southeastern sites.

( Insert Figure 5 )

The site-specific MB results are compared between these simulations in Figure 6. Results
indicate that MB values for model results from the updated FDDA meteorology were less than
those for the simulation results using the original FDDA meteorology at 88% of the sites. The
spatial distribution of MB is displayed in Figure S3 at all AQS sites from the updated FDDA
simulation results. Small positive model biases are generally found at sites in the northern
states and underestimates by the model occur at relatively few locations. On the other hand,
the highest positive biases, where the largest model overestimates of 10 LDT O3 occurred, at
sites in the southeastern states. Additionally, a closer examination of the difference in MB
between these simulations in Figure 7 indicates that MB for modeled results using the updated
FDDA meteorology was quite close to the bias in the other simulation at AQS sites in the
southeastern US, while exhibiting much lower MB values at the vast majority of sites in the
northern areas of the domain. A possible cause for the greater error and more bias in both
model simulations in the southeastern region of the domain is overestimated ozone boundary
conditions. Wind flows in the summer more frequently transport air westward across the
Atlantic Ocean or from the Gulf of Mexico into the southeastern US and modeled O3 can be
overestimated since chemical mechanisms currently do not account for various halogen species
emissions over large water bodies that may destroy O3 and dry deposition velocities need to be
increased over water bodies in CMAQ (Sarwar et al, 2014).

( Insert Figure 6 )

( Insert Figure 7 )

5. Summary

In this model evaluation study, CMAQ simulations were conducted with meteorological data
sets generated by WRF using the original and updated FDDA approaches to investigate
whether more accurate wind fields from the latter produces better model performance for mid-
morning ozone in the eastern United States. The rationale for focusing on 10 LDT ozone was
that concentrations at this time reflect transported ozone which in an evaluation could better
isolate the impact of differences in wind fields on model performance. Statistical results based on modeled and observed 10 LDT ozone pairs revealed that modeled results with the updated FDDA meteorology exhibited less bias and smaller absolute errors at a large majority of monitoring sites. Comparative results of morning ozone profiles indicated that modeled concentrations closely matched observed values in the residual layer. Comparisons of modeled ozone against tower measurements showed that CMAQ replicated the temporal variation of ozone after midnight and through the morning period. The notable positive model ozone bias in the southeastern US may be attributable to overestimated ozone in the southern boundary conditions, which deserves further investigation.

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Supporting Material Available
Example of modeled ozone concentration closest to observed values (Figure S1), mean absolute error at AQS sites (Figure S2), and mean bias at AQS sites (Figure S3). This information is available free of charge via the Internet at http://www.atmospolres.com.

References


Figure Captions

Figure 1. a) Hourly variation of ozone at 3 levels on the WRAL-TV tower near Raleigh, NC and b) hourly mean modeled ozone from 3 corresponding layers containing the measurement levels. Measurements heights are imbedded within model layers 1, 3, and 5, respectively, and layer 8 is near 400 m AGL. Data are missing at specific hours in the tower measurements due to routine instrument calibrations.

Figure 2. a) Spatial ozone pattern from the updated simulation on June 11 at 10 LDT in layer 10 (≈ 600 m AGL) and b) ozone differences (updated and original).

Figure 3. Observed and modeled O₃ profiles at a central Virginia site (Louisa, VA) at 0915 LDT on June 11.

Figure 4. a) Mean observed and modeled mid-morning ozone profiles are derived from June and July 2002 cases. Dashed lines represent ± 1σ from the mean observed values. b) Modeled original (red) and updated (blue) ozone based on values from 500-1 500 m AGL in all morning profiles.

Figure 5. Comparison of mean absolute error (MAE) between the CMAQ simulation results at individual AQS sites. Updated MAE is lower than the original MAE at 80% of all sites.

Figure 6. Comparison of mean bias (MB) between the updated and original simulation results at individual AQS sites. MB from the updated simulation is lower at 88% of sites.

Figure 7. Difference in mean bias values (updated - original) at AQS sites. Negative values indicate less bias in the simulation results with updated FDDA meteorology.
Figure 1
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\(^a\) N = number of observed and modeled pairs
\(^b\) S1 = simulation with original FDDA meteorology
\(^c\) S2 = simulation with updated FDDA meteorology
\(^d\) MB = mean bias
\(^e\) MAE = mean absolute error
\(^f\) Fp = percentage of cases that results of a simulation were closer to observations
\(^g\) EUS = 619 sites in 27 states east of the Mississippi River
\(^h\) NE = sites in 11 northeastern states
\(^i\) CNET = 34 CASTNET eastern sites
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Captions for Supporting Figures

Figure S1. Example illustrates which modeled ozone concentration (blue; updated, red; original) is closer to the observed value at AQS sites on June 11 at 10 LDT.

Figure S2. Mean absolute error at AQS sites from the CMAQ simulation results using the updated FDDA meteorology.

Figure S3. Mean bias (MB) at AQS sites from the updated FDDA simulation.
Figure S2.
Figure S3.