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The impact of chemical lateral boundary conditions on CMAQ predictions of tropospheric ozone over the continental United States

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Abstract A sensitivity study is performed to examine the impact of lateral boundary conditions (LBCs) on the NOAA-EPA operational Air Quality Forecast Guidance over continental USA. We examined six LBCS: the fixed profile LBC, three global LBCs, and two ozonesonde LBCs for summer 2006. The simulated results from these six runs are compared to IONS ozonesonde and surface ozone measurements from August 1 to 5, 2006. The choice of LBCs can affect the ozone prediction throughout the domain, and mainly influence the predictions in upper altitude or near inflow boundaries, such as US west coast and the northern border. Statistical results shows that the use of global model predictions for LBCs could improve the correlation coefficients of surface ozone prediction over the US west coast, but could also increase the ozone mean bias in most regions of the domain depending on global models. In this study, the use of the MOZART (Model for Ozone And Related chemical

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Tracers) prediction for CMAQ (Community Multiscale Air Quality) LBC shows a better surface ozone prediction than that with fixed LBC, especially over the US west coast. The LBCs derived from ozonesonde measurements yielded better O_3 correlations in the upper troposphere.

Keywords Air quality model · CMAQ · Boundary condition · Ozonesonde · AIRNOW · Chemical transport model · Ozone prediction

1 Introduction

The National Air Quality Forecast Guidance (NAQFG) run at the United States' National Centers for Environmental Prediction (NCEP) [13,20] uses time-invariant profiles as the chemical lateral boundary conditions (LBCs) for air quality predictions over the continental United States. The profile LBCs are often used even in retrospective air quality modeling when influences of transport outside of a limited modeling domain are relatively weak compared to the impact of emissions within the model domain. Over the continental United States, the major inflows are seasonal and sporadic such as pollutants emitted from Asia transported across the Pacific which are usually enhanced during springtime [7,8], pollutants from Mexico that affect the southern border, and northern influx from Canadian emissions, and occasionally Canadian and Alaskan wildfire plumes (e.g., [12]) and Saharan dust. The static LBCs obviously cannot reflect the chemical influences related to intermittent events. Here, we explore alternative approaches to defining the chemical LBCs for the NAQFG, including LBCs derived from global model predictions and from observational data, with the goal of better capturing the day-to-day variability in the chemical fields to provide improved ozone forecasts. In this study, we do not test the top boundary condition as [32] and CMAQ currently uses gradient-zero top boundary conditions. If at the top layer the vertical velocity is upward, it is just an outflow and no LBC is needed. For cases when there an inflow (downward motion), we just use the concentration at the top-most layer to estimate in advective flux in.

2 Models and lateral boundary conditions

The NOAA-EPA NAFQS is based on a one-way "offline" coupling of the North American Mesoscale (NAM) Model, which is currently the Weather Research and Forecasting Model–Non-hydrostatic Mesoscale Model (WRF-NMM; [11]), and Community Multiscale Air Quality (CMAQ) Modeling System [1]. The NAM is run for the full North American continent on a latitude–longitude domain with ~12-km horizontal grid spacing, and it contains 60 sigma-pressure hybrid layers up to 2 hPa. The primary physics options in the NAM include the NOAH (NOAA/Oregon State/AFWA /Office of Hydrology-NWS) unified 5-layer land and surface model [2], the Mellor-Yamada-Janjic planetary boundary layer (PBL) closure scheme [10], Ferrier cloud microphysics [3] and the Betts-Miller-Janjic convective mixing scheme [9]. In the NAQFG, CMAQ is run over the continental United States with 12-km horizontal grid spacing and uses 22 common WRF-NMM terrain following vertical layers from surface to 100 hPa. The CMAQ model employs the Carbon Bond 4 (CB4) chemical mechanism [4], vertical diffusivity and dry deposition based on [26], scale J-table for photolysis attenuation due to cloud, and Asymmeric Convective Model for the PBL (ACM2) [25]. The National Emission Inventory (NEI) with base year 2001 with recent updated point

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Table 1 Global models and their configurations in this study

	MOZART	RAQMS	GFS O_3
Horizontal Resolution	2.8° × 2.8°	2° × 2°	0.31° × 0.31°
Meteorology	GFS analysis	GFS analysis	GFS forecasts
Anthropogenic emissions	[5]	GEIA/EDGAR inventory with updated Asian emission [30]	Not applicable
Biomass burning emissions	GFED-v2 [35]	Ecosystem/severity based	Not applicable
Stratospheric ozone	Synthetic ozone constraint [16]	OMI/TES assimilation [24]	Initialized by SBUV-2

and mobile emissions is used in this study. We use biogenic Emission Inventory System (BEIS) version 3.13 [22] to generate hourly biogenic emissions for the model simulations. Additional details on the NAQFG configuration can be found in [20] and [14].

In this study, six simulations are conducted with the NAQFG to assess the impacts of various sources of LBCs. The first simulation, "Fixed LBCs", uses the time-invariant chemical profile LBCs that are employed in the operational system. Three additional simulations are performed to assess the impact of using LBCs derived from global models (Table 1) for the time-varying chemical boundary conditions. These global models incorporate satellite-based chemical data, but with different methodologies. Among the global models, the Model for OZone And Related chemical Tracers (MOZART) model (version 4, updated from [6]) has the most detailed reactions and related chemical species (97 species), including bulk sulfate, ammonium, organic and soot aerosols, and size-resolved dust and sea salts (e.g., [21]). The Real-time Air Quality Modeling System (RAQMS) [23] model used in this study has only gaseous chemistry. A statistical digital filter analysis system [28,29] was used in RAQMS data assimilation to perform a univariate assimilation of stratospheric profile and total column ozone observations from Aura OMI (Ozone Monitoring Instrument) satellite data [24]. In the simulations with LBCs derived from MOZART and RAQMS fields, the time-varying chemical LBCs include full-profile O_3, CO, sulfur oxidants, nitrogen oxidants and volatile organic compounds (VOCs). Table 2 shows the species mapping tables which is used to convert the LBC species concentrations of RAQMS and MOZART to CMAQ's implementation of CB4. MOZART and RAQMS have similar inorganic gaseous species. RAQMS's mechanism is modified from CB4 by including explicitly treated ethane (C_2H_6) and a different lumping method for alkenes. The MOZART chemical mechanism has more explicit VOCs, which are simply split into paraffin (PAR) and olefin (OLE) carbon bonds. The third simulation initializes CMAQ LBCs with 3D prognostic O_3 fields from NCEP's operational Global Forecast System (GFS) [17,18]. The O_3 prediction in GFS is initialized from assimilation of Solar Backscatter Ultra-Violet (SBUV-2) satellite observations, and O_3 is advected as a trace species with simple zonally averaged climatological derived production and depletion mechanism [27]. Since SBUV-2 can only provide O_3 data above 250 hPa, GFS-O_3 LBC is applied to the CMAQ simulation above 11 km, and time-invariant chemical boundary conditions are used for O_3 below 11 km and for all other chemical species. In this study, we import GFS-O_3 LBC every hour while MOZART and RAQMS LBCs are updated every 3 and 6 h, respectively.

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Table 2 Hydrocarbon species mapping tables between RAQMS and MOZART species and CMAQ CB4

RAQMS species	CMAQ CB4
C2H6	2*PAR
OLET (terminal alkenes)	OLE1 + PAR
OLEI (internal alkenes)	OLE2 + 2*PAR
MOZART species	**CMAQ CB4**
CH3CHO	ALD2
C2H6	2*PAR
C3H8	3*PAR
BIGALK (higher alkanes)	4*PAR
C3H6	OLE + 2*PAR
BIGENE (higher alkenes)	OLE + 3*PAR
C10H16 (terpene)	OLE + 9*PAR

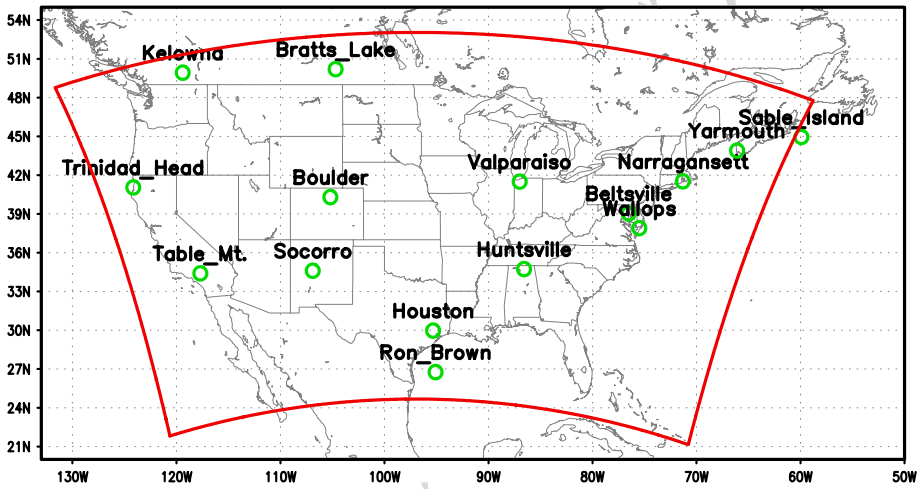

Fig. 1 IONS ozonesonde sites during August 1–5, 2006, and the model domain

The final two simulations in this study include the ozone boundary conditions derived from ozonesondes (see Fig. 1 for the station locations) from the INTEX Ozonesonde Network Study (IONS; available online at http://croc.gsfc.nasa.gov/intexb/ions06.html; [33,34]) averaged from August 1 to 5, 2006 as the IONS data are not available on earlier dates from which the simulations started. The IONS balloon-borne ozonesonde data were sampled by electrochemical concentration cell sensors with an accuracy of about 10% in the troposphere, but that accuracy can degrade to 15% when O_3 is lower than 10 ppbv [19]. For the first of these simulations, "IONS-LBC1", the values from the nearest ozonesondes (based on latitude or longitude) to each boundary are assigned and distributed without spatial interpolation along the lateral boundaries (11 of total 15 ozonesonde sites were used). For example, the IONS ozonesonde profiles from Table Mountain, Trinidad Head, and Kelowna (Fig. 1) are distributed from south-to-north (Fig. 2). It should be noted that NOAA Research Ship Ron Brown (Fig. 1) was not anchored in the same location for this study, so its influence along the southern boundary varies as a function of its location in the Gulf of Mexico. In the last of these simulations, "IONS-LBC2", only the western boundary, a major inflow boundary,

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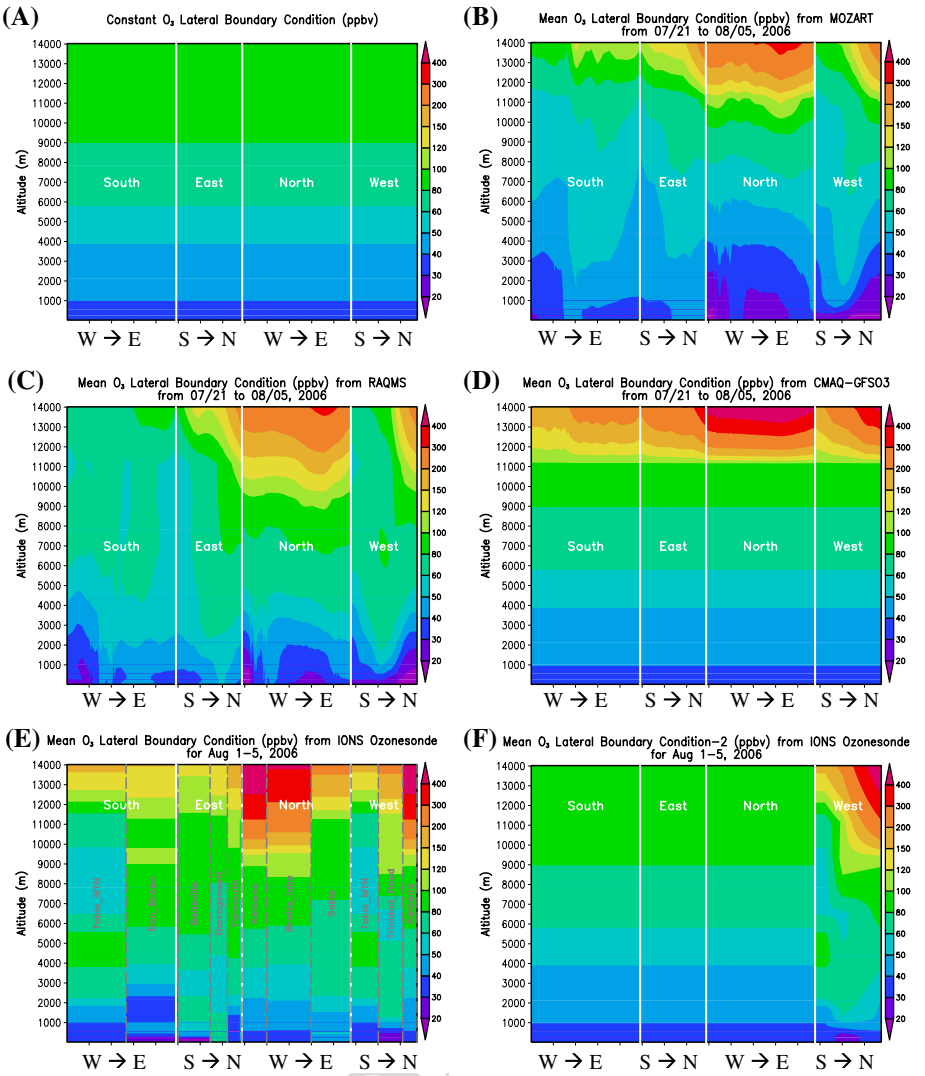

Fig. 2 Mean O_3 lateral boundary conditions along the boundaries of CMAQ 442 × 265 (12-km horizontal grid spacing) CONUS domain. The values are list from west to east for northern and southern boundaries, and from south to north for western and eastern boundaries

is changed. In this case, the ozone observations are interpolated (rather than assigned, as in IONS-LBC1) to the boundary cells. For the boundary cells south of Table Mountain and cells north of Kelowna, the assignments used in IONS-LBC1 are applied. On the other three lateral boundaries, the fixed chemical LBCs are used.

The simulations in this study are initialized with the CMAQ operational prediction at 12 UTC 21 July 2006 and run for 16 days, driven by daily updated WRF-NMM prediction initialized at 12 UTC of each day. The simulation period of July 21 to August 5, 2006 includes several near-surface high-ozone events due to regional and local photochemical activities. It is neither a particularly strong inflow period, nor a typical scenario for Asian pollutants

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or biomass burning plumes to reach North America. During the summertime, the long-time (>4 days) trans-Pacific transport cannot bring plumes of short-lived NO_x, SO_x or very active VOCs [31] to North America. Relatively long-lived CO could show some impact if the air mass encountered sufficient NO_x during daytime. However, as more comprehensibly studied by [32], the influence of CO on O_3 is one order of magnitude weaker than the influence of O_3 on O_3 in per mole base. So we mainly focus on varying the ozone lateral boundary conditions in this scenario. All the lateral boundary conditions for the sensitivity simulations are implemented throughout the simulation period.

Figure 2 shows the mean O_3 LBCs from July 21 to August 5, 2006. Above 11 km, GFS O_3 LBC has the highest mean O_3 concentration. RAQMS and MOZART provide similar O_3 LBCs in the upper troposphere, while the major difference appears in the middle troposphere as the RAQMS has more highly concentrated ozone bands extended from upper layers to lower layers. Below 1 km, RAQMS and MOZART have lower mean O_3 LBC than the fixed LBCs.

3 Comparison to ozonesondes

Ozone predictions from the NAQFG with different chemical LBCs are compared with ozonesondes from IONS (see Fig. 1). Figure 3 shows the model predictions compared to ozonesonde measurements at six sites on August 3, 2006. At Beltsville, Maryland, all six simulations overpredicted the near-surface O_3 by 10–20 ppbv. The fixed, MOZART, and RAQMS LBCs led to significant underpredictions of the tropospheric O_3 above 4 km and by as much as ~50 ppbv above 9 km. Use of the GFS and the ozonesonde-derived LBCs improved the O_3 predictions aloft. The IONS LBC1 simulation yielded the best performance over Beltsville, although the fluctuations in the vertical (e.g., between 4 and 9 km) are not captured in any of the simulations due to coarse NAQFG vertical resolution aloft. At Huntsville, Alabama and Boulder, Colorado, GFS O_3 LBC and two IONS LBCs also yielded higher O_3 concentration at high altitudes (e.g., above 6 km) than the simulations with the "fixed", MOZART, and RAQMS LBCs. However at Boulder, the use of GFS O_3 LBC continued to overestimate O_3 above 7 km by 20–30 ppbv, which was much higher than any of the other five methods. Below 6 km at Boulder, differences among the simulations except for the IONS LBCs are less than 10 ppbv, and the ozonesonde-derived LBCs tend to overpredict O_3 between 4 and 6 km. At Trinidad Head, California, near the domain's western boundary, using RAQMS LBC and two IONS LBCs resulted in better agreement than the other methods between 1 and 6 km, while the other simulations are about 20 to 40 ppbv lower than the observations at those altitudes.

At Bratt's Lake, Saskatchewan, Canada, which is near the model's northern boundary, all simulations have comparably good O_3 prediction below 4 km. Above 10 km, a significant ozone intrusion of more than 400 ppbv is observed, which did not appear at the ozonesonde sites except Kelowna on August 3. Here, the importance of using day-specific LBCs is demonstrated as most experiments yield higher and more accurate ozone predictions aloft than using a static profile. In fact, using the ozonesonde data on the lateral boundaries enabled the NAQFG to better capture the ozone enhancement when the balloon entered stratosphere, though it still under predicts the peak values near 12 km by more than 100 ppbv. Winds at Bratt's Lake were westerly at the time of the observation (Fig. 4a), and hence the source of the increased ozone at Bratt's Lake likely came from the western boundary, further evidence in the comparison of IONS-LBC1 and IONS-LBC2 at Bratt's Lake. The observed O_3 aloft at Kelowna, Canada (Fig. 3f) is ~400 ppbv, which suggests that the ozone peak at Bratt's Lake may have originated from an earlier, transient event under westerly wind (Fig. 4) because

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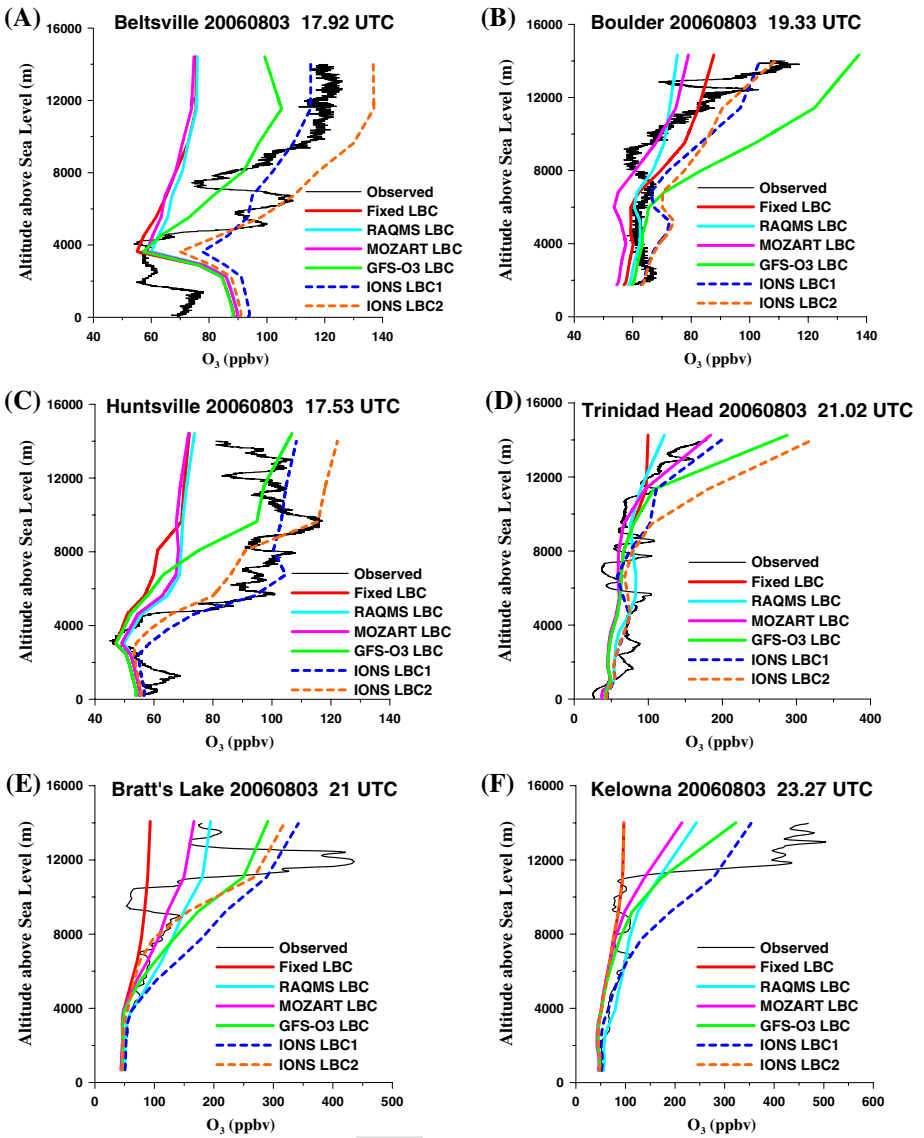

Fig. 3 The model simulations compared to IONS ozonesonde measurements at Beltsville (39.0°N, 76.5°W), Boulder (40.3°N, 105.2°W), Huntsville (35.3°N, 86.6°W), Trinidad Head (40.8°N, 124.2°W), Bratt's Lake (50.2°N, 104.7°W) and Kelowna (49.9°N, 119.4°W) on August 3, 2006

changing LBCs significantly affected the prediction at Bratt's Lake. However, none of these simulations could replicate the ozone peak values above 12 km altitude at Bratt's Lake and Kelowna. On August 3 at Kelowna, the northern boundary was the major inflow boundary (Fig. 4), which is also reflected by IONS-LBC2 results similar to the fixed LBC.

Figure 4 shows a spatial comparison of the relative change in O_3 prediction aloft (e.g., ~9.6 km above ground level, the CMAQ's 20th layer) for each of the simulations compared with the time-invariant LBCs. Figure 4a shows that using the fixed LBCs rarely leads to

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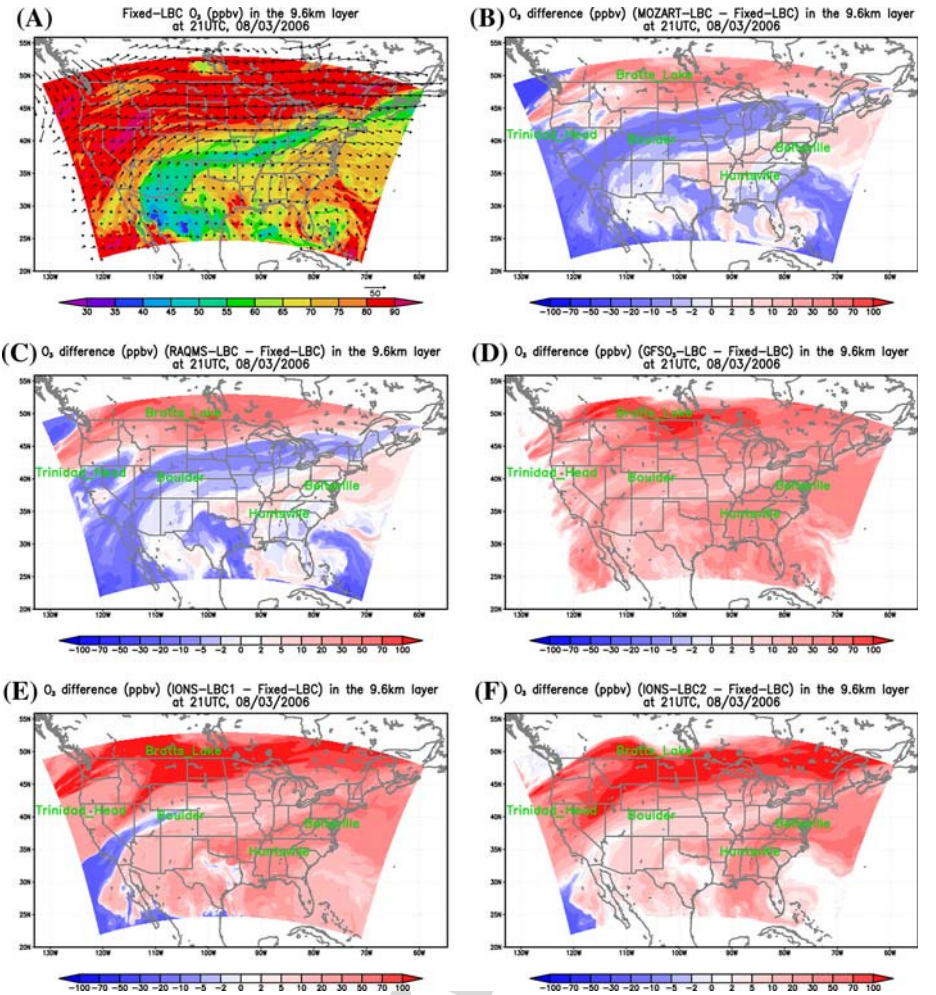

Fig. 4 Model simulated ozone (**a** for Fixed-LBC) and their differences (**b–f**) in the model's 9.6 km layer at 21Z, August 3, 2006

a prediction of more than 90 ppbv aloft anywhere in the domain, though the ozonesondes in Fig. 3 (which are distributed throughout the simulation domain) have strong variations (50 ∼ 500 ppbv) at that altitude and above. Figure 4b and c show that using the chemical LBCs from MOZART and RAQMS results in large reductions of O_3 aloft, particularly south of the jet stream (refer to Fig. 4a) and in the subtropical regions as compared to the simulation using fixed LBC. MOZART and RAQMS LBCs have lower O_3 than the Fixed LBCs (Fig. 2) aloft along the western boundary (south of the jet steam) but higher O_3 along the northern boundary. Figure 4d shows that in most areas of continental United States, the GFS O_3 LBC increases O_3 prediction aloft compared with using Fixed LBCs, which is a direct result of increasing the O_3 on the lateral boundaries (Fig. 2). The two simulations with IONS LBCs (Fig. 4e, f) increase the O_3 predictions by more than 100 ppbv (compared with the Fixed LBCs) in a swath along the jet stream from the western boundary across the northern portion

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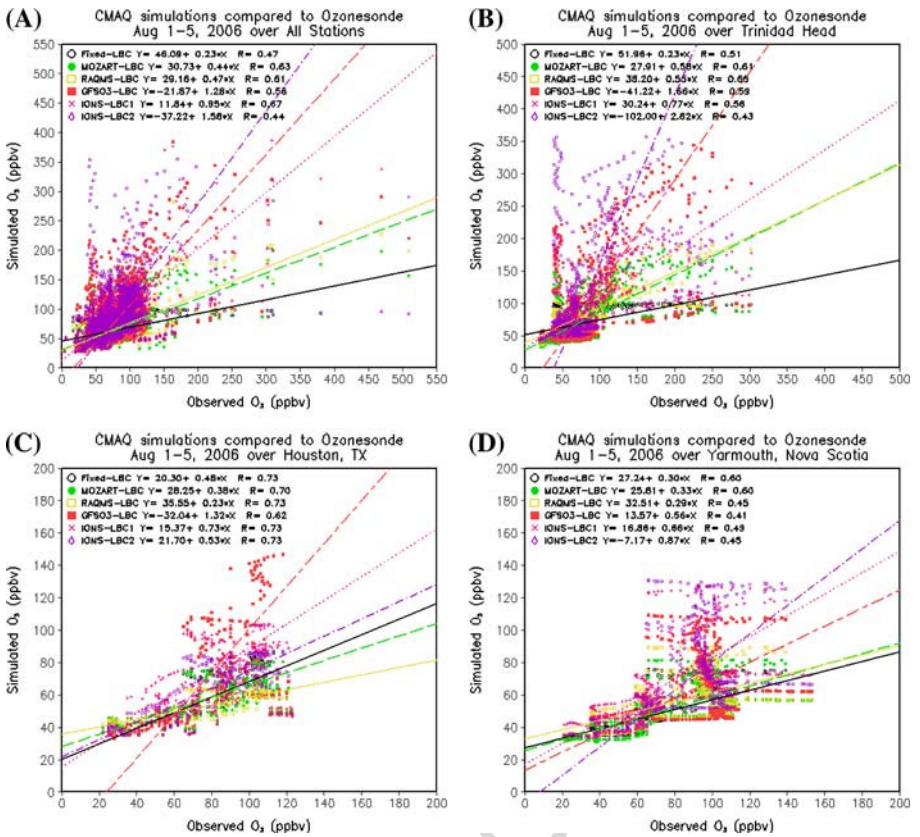

Fig. 5 The correlations between the simulations and measurement over IONS stations from August 1 to 5, 2006

of the domain. As noted above, this increase in O_3 aloft is consistent with measurements at Bratt's Lake (Fig. 3e). The IONS LBCs correctly reduce the O_3 aloft in the southwestern USA, since the LBCs from IONS data use have relatively lower ozone observed in Table Mountain (not shown). The increases in O_3 prediction aloft and north of 40°N emanate from the western boundary in this event (compare Fig. 4e, f) since IONS LBC2 has the same northern boundary values as the Fixed LBC but uses ozonesonde data along the western boundaries.

Figure 5 compares observed and predicted O_3 values throughout the troposphere for the six simulations at the 15 IONS ozonesonde sites (Fig. 1) during 1–5 August 2006. The IONS-LBC1 simulation has the highest correlation coefficient R in Fig. 5a, which suggests that the observed profiles can provide valuable influences along lateral boundaries. The IONS-LBC2 case tends to overpredict O_3 in the mid-troposphere (Fig. 3) and has a lower correlation coefficient. The O_3 overprediction of IONS-LBC2 compared with IONS-LBC1 over Trinidad head (Fig. 5b) reflects the result difference of using linear interpolation versus segmented assignment along the west boundary. The major overprediction of IONS-LBC2 compared to IONS-LBC1 occurred above 9 km over Trinidad Head (Fig. 3d), which was caused by their difference along the western boundary (Fig. 4e, f). However, since we only have three stations along the west coast, more studies are needed to evaluate which method

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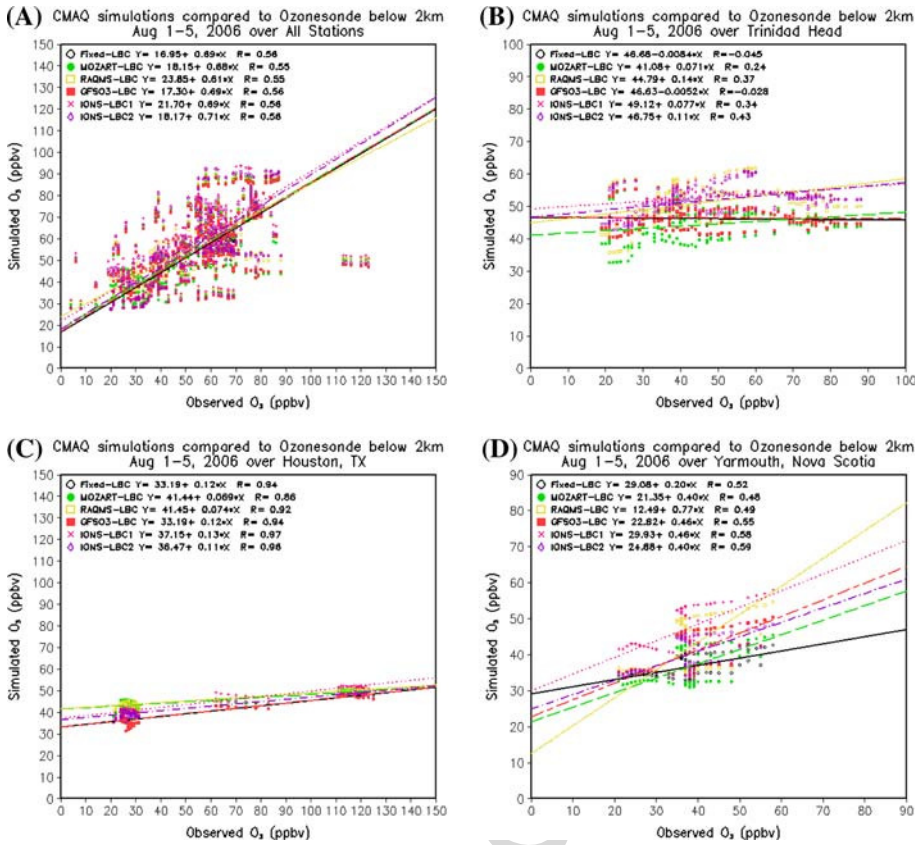

Fig. 6 Same as Fig. 5 but only for the altitudes below 2 km

is better for using ozonesonde data. Among the simulations using global model LBCs, the MOZART and RAQMS LBCs yielded similar correlations, and the GFS-O_3 LBC had the more reasonable regression slope, reflecting its skill for predicting O_3 variability aloft. The simulation with the Fixed LBCs yielded the lowest prediction skill for O_3 variations in the upper troposphere, as expected, as seen by its low regression slope (0.23 for all stations). In the lower troposphere (i.e., below 2 km), the differences between the simulations are small (Fig. 6). Therefore, in general, varying the LBCs most directly affects the O_3 concentrations in the middle and upper troposphere, but over time the specification of the LBCs can influence the surface O_3 concentrations on the interior of the forecast domain through transport and vertical advection processes (e.g., due to convection). Near the surface, regional or local emissions often play a more important role in dictating day-to-day O_3 prediction. However, the relative importance of each influencing factor also depends on the locations and scenarios. For instance, using time-varying LBCs from RAQMS improved all-altitude and low-altitude O_3 prediction at Trinidad Head (Figs. 5b, 6b) because it is located near the western boundary of the domain (Fig. 1) and it was used in the specification of the O_3 concentrations along the western boundary. On the other hand, the LBCs derived from global models did not show significant improvement over Houston at high altitudes (except for GFS O_3 that has better slope) or low altitudes (Figs. 5c, 6c). The use of time-varying LBCs had a moderate impact

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at Yarmouth, Nova Scotia, (Figs. 5d, 6d) as it is located in the northeastern quadrant of the NAQFG domain (close to Eastern and Northern outflow boundaries). The use of real-time global-model LBCs and IONS LBCs yielded better regression slopes over Yarmouth, but did not show an advantage in term of the correlation coefficient (Figs. 5d, 6d).

4 Comparison to surface observations

In the NAQFG, verification with surface measurements is a primary performance indicator because the target forecasts are for near-surface O_3, which most directly affects the human population. The Environmental Protection Agency's Air Quality System (AQS) (available from http://www.epa.gov/air/data/aqsdb.html; http://airnow.gov) network provides hourly surface ozone measurement across the United States and largely in urban areas, where the human population is concentrated. During this period, NAQFG surface O_3 prediction had a high bias, and using global LBCs may exaggerate the NAQFG overprediction of O_3 (Lee et al., this issue). In this study, 1635 AQS/AIRNOW stations within the NAQFG domain are used for near-surface verification. Table 3 shows the statistical results for the hourly data over these surface stations. Global models and IONS lateral boundary conditions yield better regression slopes when compared against all AIRNOW sites, implying that these LBCs could improve the model prediction for ozone variation magnitudes, though these LBCs could cause higher mean biases. The impact of LBCs becomes more evident when the evaluation is limited to stations west of −115°W (i.e., near the western boundary of the domain). Figure 7 shows the surface O_3 mean biases for the six simulations superimposed on their predicted mean O_3 (for Fixed LBCs only) or on mean predicted O_3 differences from Fixed LBCs (all other simulations). The use of MOZART LBCs shows the best improvement relative to the Fixed LBCs in this region by reducing the mean bias in surface O_3 by 2.5 ppbv and increasing the correlation coefficient/slope (Table 3). The most significant improvement occurred in Pacific Northwest and California coastal regions (Fig. 7b). The RAQMS LBCs simulation has

Table 3 CMAQ simulations compared to AIRNOW hourly O_3 data from August 1 to 5

	All stations	West of −115°W	North of 43°N	East of −85°W
Fixed LBC	S = 0.887	S = 0.804	S = 0.873	S = 0.844
	R = 0.714	R = 0.691	R = 0.737	R = 0.742
	MB = 8.0 ppbv	MB = 4.7 ppbv	MB = 7.5 ppbv	MB = 11.3 ppbv
RAQMS LBC	S = 0.911	S = 0.914	S = 0.942	S = 0.823
	R = 0.718	R = 0.703	R = 0.742	R = 0.744
	MB = 10.0 ppbv	MB = 7.1 ppbv	MB = 10.0 ppbv	MB = 12.8 ppbv
MOZART LBC	S = 0.941	S = 0.872	S = 0.985	S = 0.872
	R = 0.716	R = 0.730	R = 0.743	R = 0.737
	MB = 8.2 ppbv	MB = 2.2 ppbv	MB = 6.9 ppbv	MB = 11.8 ppbv
GFS O_3LBC	S = 0.935	S = 0.820	S = 0.922	S = 0.858
	R = 0.714	R = 0.697	R = 0.724	R = 0.736
	MB = 9.2 ppbv	MB = 4.8 ppbv	MB = 9.0 ppbv	MB = 12.9 ppbv
IONS LBC1	S = 0.947	S = 0.912	S = 0.905	S = 0.817
	R = 0.718	R = 0.692	R = 0.709	R = 0.742
	MB = 12.4 ppbv	MB = 10.4 ppbv	MB = 12.9 ppbv	MB = 15.3 ppbv
IONS LBC2	S = 0.952	S = 0.910	S = 0.922	S = 0.858
	R = 0.723	R = 0.698	R = 0.736	R = 0.742
	MB = 10.1 ppbv	MB = 9.0 ppbv	MB = 9.2 ppbv	MB = 13.0 ppbv

S is regression slope; R is correlation coefficient; MB is mean bias

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Fig. 7 Mean surface O_3 prediction (plot **a** for Fixed LBC), mean O_3 prediction differences (**b** for MOZART LBC, **c** for RAQMS LBC, **d** for GFS O_3 LBC, **e** for IONS LBC1 and **f** for IONS LBC2) and their mean biases from AIRNOW observations during August 1 to 5, 2006 ►

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the best regression slope, but it also tends to increase the mean bias (Table 3). It should be noted that the greatest impact from the RAQMS LBCs occurred not along the coast but further inland in Idaho (Fig. 7c). The simulations with IONS LBC also show similar features. It could be caused by overpredictions of PBL height and the corresponding vertical mixing as indicated by Lee et al. (this issue) that WRF-NMM/CMAQ tends to overpredict PBL height for the 2006 simulations. Overpredicted PBL can bring high ozone from middle layers to surface while RAQMS and IONS LBCs have higher ozone concentration between 2 and 6 km along the western boundary than that in MOZART (Fig. 1). The impact of GFS O_3 LBC near surface is relatively small over this region since it is used only above 11 km and needs longer transport time/distance to affect surface ozone (Table 3; Fig. 7d). The GFS O_3 LBC simulation has a stronger impact in the region north of 43°N. The two IONS LBCs simulations resulted in the highest surface O_3 bias over the whole domain (Table 3). Along the west coast, the differences between two IONS LBCs and the fixed LBCs range from 0 to 10 ppbv (Fig. 7e, f). Near the western boundary, all of the time-varying LBCs improve the regression slopes relative to the fixed LBCs, but only the MOZART LBC reduces the mean bias and has the highest correlation coefficient/slope. Table 3 also shows the statistical comparison of surface O_3 predictions over the eastern United States. The LBC influence in east coast during this period is relatively insignificant since it is the prevailing outflow region. In east coast, the simulations with various LBCs did not show significant difference for O_3 prediction on the correlation statistics while the mean biases are less than 4 ppbv. So the LBCs mainly affect the background concentration of predicted ozone, not its variation, as the variation and gradient from western and northern inflow boundaries have been diffused during the long-distance transport to the east coast.

5 Conclusion

This study compares the impacts on tropospheric ozone prediction over continental United States from using various LBCs derived from global models and from ozonesondes. Although this period is not a noticeable intrusion event, these boundary conditions still show a strong impact on ozone prediction at most altitudes. Depending on locations and scenarios, the simulations with these LBCs did not always yield better results than that with the fixed LBCs. For global-model LBCs, the error could be caused by global model performance or CMAQ regional prediction. When ozonesonde LBCs are used, CMAQ performance depends on ozonesonde location relative to the regional-model boundary locations, spatial/temporal resolutions, the treatment of ozonesonde data and how CMAQ evolves the observed information. Lee et al. [13] showed that high ozone aloft can often be mixed downward too quickly by CMAQ, thereby increasing model errors in the lower troposphere. In this study, we found a similar behavior: the simulation with IONS LBC1 shows good agreement with 15 IONS ozonesonde data aloft, but leads to O_3 overestimation near surface. The performance inconsistency implies that CMAQ could overestimate the vertical mixing and bring too much ozone downward. This problem could be related to the operational CMAQ's coarse vertical resolution in the free troposphere (with top layer's thickness of 4 km). The operational CMAQ uses only 22 vertical layers for computation efficiency. This relatively coarse vertical resolution could not resolve the tropopause boundary, and erroneously mix too much stratospheric ozone into the free troposphere. In lower troposphere, boundary layer mixing can

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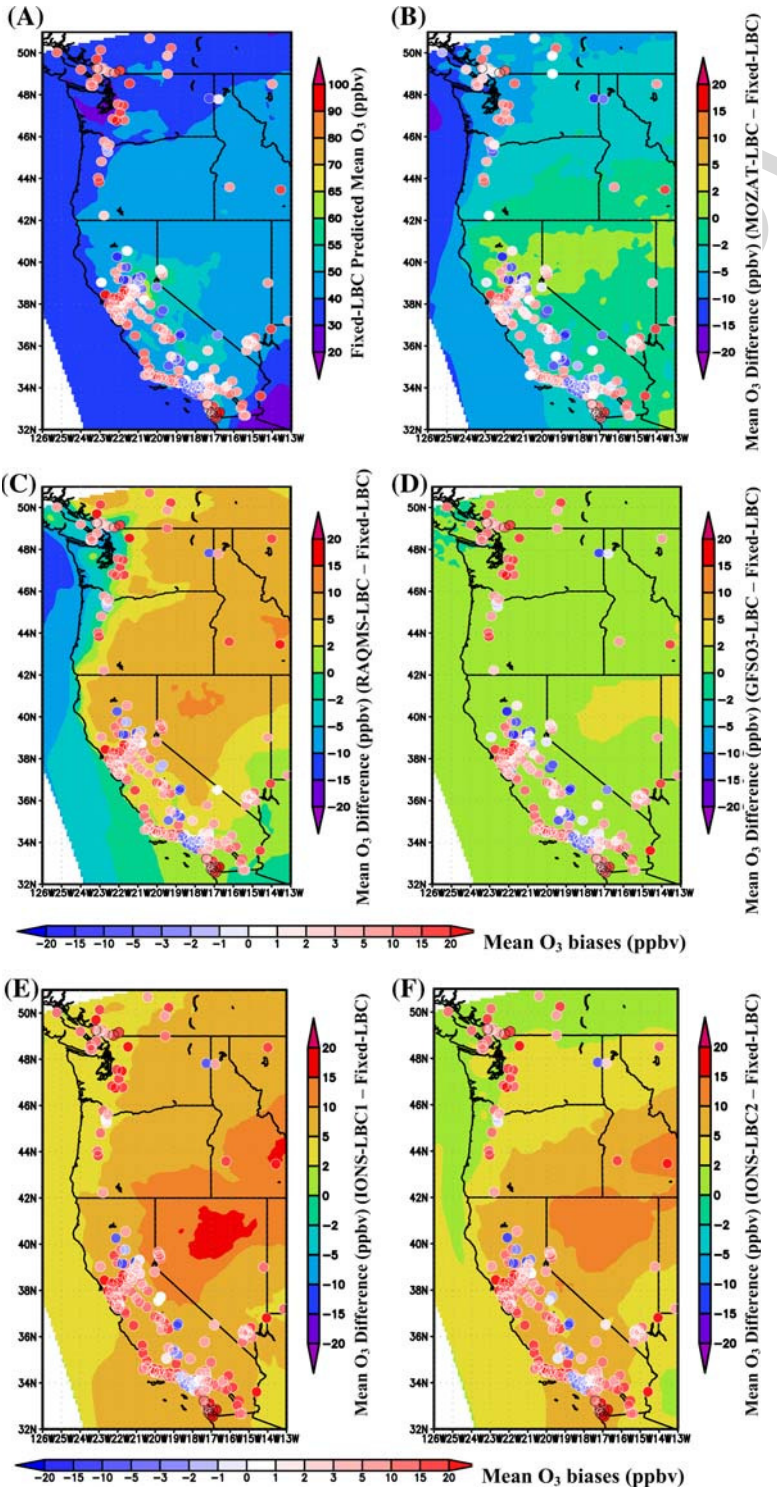

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also transfer the ozone to the surface [13]. We are investigating this issue. The ozonesonde site locations can be far from the domain boundaries, which introduces additional possible errors in IONS LBCs simulations. Current 15 IONS ozonesonde sites are not sufficient for LBCs due to their spatial and temporal resolution, though the preliminary test in IONS LBC1 shows encouraging results in middle and upper troposphere.

At high altitudes, ozone predictions are mainly related to stratospheric influence, in which using GFS O_3 for LBCs and ozonesonde-derived LBCs can capture strong transport events, perhaps due in part to their high spatial resolution. For some events, GFS O_3 also tends to overpredict upper-troposphere ozone, possibly due to insufficient vertical resolution of assimilated SBUV-2 satellite data. In this study, we did not test the use of global simulations for the top boundary condition [32], or use potential vorticity as the indicator of stratosphere–troposphere exchange, which could yield even stronger impacts on upper atmospheric prediction. Compared to GFS O_3 LBC, RAQMS and MOZART can provide height-resolved LBCs for additional species. These species include long-lifetime CO and NO_y, which can be transported from distant sources and possibly affect regional ozone production. In some events, such as the biomass burning near the domain boundary, very active species, like NO_x, could also cause great impacts. The chemical transformation of LBCs needs to be evaluated in the future.

This study demonstrated the potential for using time-varying chemical LBCs for air quality predictions of tropospheric O_3. The gains in skill from using LBCs derived from global models are promising. Although each of the global models used in this study has different strengths, it is clear that time-varying chemical LBCs derived from those models can contribute toward improved simulations of O_3 above the surface layer compared to simulation with fixed profile LBCs. Additional simulations should be conducted with more representative values for the fixed profile LBCs (particularly in the upper troposphere) to determine the full advantage of using chemical LBCs from global models. However, even if the time-invariant LBCs are increased aloft, only LBCs derived from daily conditions can capture the transient pollutant events that are critical to predicting air quality over a long period. In addition, this study shows that the LBCs derived from ozonesonde measurements yielded more skillful O_3 predictions, particularly in the upper troposphere. While we recognize that the NAQFG cannot use these data in real time because they are not centrally reported, this study can advocate the assembly and reporting of a real-time 3D air quality network that could be used, in part, to improve air quality predictions. Finally, this study has highlighted how improvements in global air quality models could yield better results for regional air quality prediction. Lateral boundary conditions are essential for regional air quality prediction, even for the scenario without strong intrusions. However, once the regional air quality model adopts the global-model LBCs, uncertainties from the global models could impact errors in the regional predictions, and we may need to overcome more uncertain factors than the regional models do to achieve improvement.

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