The Impact of Lateral Boundary Conditions on CMAQ Predictions over the Continental US: a Sensitivity Study Compared to Ozonsonde Data

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1. Introductions

The offline coupled WRF-Non Hydrostatic Mesoscale model (NMM)/ Community Model for Air Quality (CMAQ) system is the current operational air quality forecast system (AQFS) used at NOAA/NWS/NCEP. This system uses a static profile as the lateral boundary condition (BC) for air quality prediction over the continental USA, since real-time boundary conditions are not commonly available. The simple-profile BC has been widely used in the case when external influences from transport outside the CMAQ domain are relatively weak compared to emissions within the model domain. However, the validity of using a static BC is difficult to assess since the importance of the external influence is very uncertain. Over the continental USA , the major inflows include Asian pollutants transported across the Pacific which usually becomes strong during springtime (Jacob et al., 2001; Jaffe et al., 2003), Mexican pollutants impacting the south border, and Northern influx from Canadian emissions and occasionally Canadian and Alaskan wildfire plumes. Sahara desert dust storm could also reach the U.S. crossing the Atlantic. The fixed BC cannot reflect these influences related to certain events. An alternative approach for addressing this uncertainty is to use global model predictions as the lateral boundary conditions. It should be noted that the global models may also introduce uncertainties from their own model errors. In this study, we perform sensitivity studies with 3 global boundary conditions and existing fixed boundary conditions to evaluate the influences due to BCs.

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2. Models and Boundary Conditions

The operational CMAQ system with Carbon Bond Mechanism-4 (CBM4) chemical mechanism (Gery et al., 1989) at 12km horizontal resolution covering continental US is used in this study, with 22 vertical layers up to 100hPa. It uses vertical diffusivity and dry deposition based on Pleim and Xu (2001), scale J-table for photolysis attenuation due to cloud, and Asymmeric Convective Scheme (ACM) (Pleim and Chang, 1992). The detailed operational CMAQ configuration can be found in

Lee et al (2007b). This air quality prediction is driven by hourly meteorological forecasts from the operational North America Mesoscale (NAM) WRF-NMM prediction system. The NAM prediction is run with 60 sigma-pressure hybrid layers up to 2hPa; Noah unified 5-layer land and surface model; Mellor-Yamada-Janjic planetary boundary layer closure scheme; Ferrier cloud microphysics; and Betts-Miller-Janjic convective mixing scheme.

In this sensitivity study, all simulations use the same emissions, meteorology and other configurations except for lateral boundary conditions. To investigate the influence of BC, we employ 3 global models (Table 1) for the time-varied boundary conditions. It should be noted that all these global models integrate satellite data, though in different methods. Among these global models, the MOZART (Model for OZone And Related chemical Tracers) model (version 4, updated from Horowitz et al., 2003) 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., Pfister et al., 2005). The RAQMS (Real-time Air Quality Modeling System) (Pierce et al., 2003) model used in this study has only gaseous chemistry. GFS (Global Forecast System) O_3 boundary condition is provided by NCEP's operational GFS that treats O_3 as a 3-D prognostic variable (Moorthi and Iredell, 1998; NCEP, 2004). The O_3 prediction in GFS is initialized with Solar Backscatter Ultra-Violet (SBUV-2) satellite observations, and advected as a trace species with simple zonally averaged climatological derived production and depletion mechanism (Rood et al. 1991). Since SBUV-2 can only provide O_3 data above 250hPa, GFS-O_3 BC is applied to the CMAQ simulation above 10km, while the other two global models provide CMAQ with full-profile O_3, CO, sulfur oxidants, nitrogen oxidants and volatile organic compounds (VOCs). Table 2 shows the species mapping tables used in this study from RAQMS/MOZART to CMAQ's CBM-IV chemical mechanism. These models have similar inorganic gaseous species. RAQMS's mechanism is modified from CBM-IV. It added explicitly treated ethane (C2H6), and uses different lumping method for alkenes. MOZART chemical mechanism has more explicit VOCs. For most of them, we simply split into paraffin (PAR) and olefin (OLE) carbon bonds.

Figure 1 shows the mean O_3 BCs from July 21 to Aug 5, 2006. Above 10 km, GFS O_3 BC has the highest mean O_3 concentration, but its values below 10km are same as those with static BC. RAQMS and MOZART provide similar O_3 BCs in the upper troposphere, and their major difference appears in the middle troposphere as the RAQMS has more high-concentrated ozone bands extended from upper layers to lower layers. Below 1 km, RAQMS and MOZART have lower mean O_3 BC than the static BCs. Four simulations with different BCs are performed starting from 12Z, July 21, with the same initial conditions and other configurations. Simulated results are compared with IONS (INTEX Ozonesonde Network Study http://croc.gsfc.nasa.gov/intexb/ions06.html) ozonesonde data and EPA AIRNOW surface O_3 data. IONS sites for our research period are shown in Figure 2. It should be noted that NOAA Research Ship Ron Brown cruised over Gulf of Mexico, and had time-varied locations.

3. Comparison to Ozonesondes

Figure 3 shows the model prediction compared to IONS ozonesonde measurements over 5 sites on August 3, 2006. These site locations can also be seen in Figure 4. Over Beltsville, Maryland, most simulations yielded similar ozone profiles, except when using GFS O_3 BC, which captured the high ozone event in the upper troposphere, with less underprediction. Similar behaviors are also evident at Huntsville, Alabama and Boulder, Colorado. However at Boulder, the use of GFS O_3 BC led to overestimation of O_3 above 7km. Below 6km, differences between the simulations are less than 10 ppbv over Boulder. These four simulations usually yielded similar result in low altitudes, except in Trinidad Head, the site facing Pacific inflow. Here, using RAQMS BC (Figure 3) resulted in better agreement below 8km, especially between 1km and 6km. Other simulations are about 20 to 40 ppbv lower than the observations. Bratt's Lake, Saskatchewan, Canada is near the model's north boundary. Over this site, all simulations have similar O_3 prediction below 4km. In the altitudes above 10km, using GFS O_3 yielded higher ozone than RAQMS BC which yielded higher ozone than MOZART BC, with all experiments yielding larger ozone than using a static profile. Winds at Bratt's Lake were mainly westerly (Figure 4A). Figure 4 shows that in most areas of continental USA, global model BCs tend to cause higher O_3 prediction than that using Fixed BC in upper troposphere, which is simply due to their boundary condition difference (Figure 1). In other stations near the domain's north and northwest boundaries, such as Kelowna, British Columbia , Canada, we also found GFS O_3 >

RAQMS > MOZART > Fixed BC during this period. Most high differences appeared north of 40°N driven by the west wind. It should be noted that these time-varied BC sometimes also yielded lower ozone concentrations than that with fixed BC, such as the outflow near middle Atlantic coast. In general, these global BCs brought more time-dependent variations to CMAQ.

During August 1 to 5, 2006, there are 15 IONS ozonesonde profiles available for comparison within the CMAQ domain (Figure 2). Figure 5 shows observed vs predicted diagrams between the four simulations for all of these 15 profiles during the 5 days. The MOZART-BC simulation has the highest correlation coefficient R in Figure 5, indicating that this configuration best captured variation trends from lateral boundaries. The RAQMS BC yielded similar statistical correlations. Among these simulations, GFS-O_3 BC had the most reasonable correlation slope, reflecting its good skill for predicting variation in magnitudes. The Fixed BC yielded the worst performance in the upper troposphere, as expected. In the lower troposphere, the differences between experiments are small (Figure 6). Therefore, for most cases, the BC variations mainly affect the model predictions for the middle and upper troposphere. Near the surface, some regional or local factors may play a more important role on ozone. However, the relative importance of each influencing factor also depends on the locations and scenarios. For instance, using time-varied BCs from RAQMS did improve overall and low-altitude O_3 prediction (Figure 5B & 5B), over Trinidad Head, because it is located at the west edge of Northern California and facing Pacific inflow. On the other hand, global BCs did not show significant advantage over Houston at either high altitudes (except for GFS O_3) or low altitudes (Figures 4C, 5C) as other factors probably dominated. The use of different BCs had a moderate impact over Yarmouth, Nova Scotia as it is located in the Northeast of the CMAQ domain and is affected by both emitted pollutants over the continent and less frequently by BCs when transport is from the northwest. The use of real-time global BCs yielded better correlations slopes (Figure 6D) below 2km over Yarmouth, but did not show an advantage at upper levels (Figure 5D).

4. Comparison to AIRNOW data

The previous section emphasized that the importance of lateral boundary conditions depended on site location and weather conditions. Also, choice of boundary conditions on model predictions was more important at higher altitudes. For air quality prediction, verification with surface measurements is a primary performance indicator. EPA AIRNOW (http://airnow.gov) data provides operational surface ozone measurement in hourly temporal resolution. In this study, we selected 1635 AIRNOW stations within the CMAQ's CONUS domain to compare BC impacts. Table 3 shows the statistical results. Real-time boundary conditions do yield statistical improvements when compared against all AIRNOW sites except correlation slopes, implying that real-time BCs could improve the model prediction for ozone variation magnitudes. In this scenario, CMAQ prediction had a high bias. It is possible that the use of global BCs sometimes could exaggerate the CMAQ overprediction (Lee, et al., 2007a). The impact becomes stronger when evaluations are done for stations just west of -115°W (includes California, Oregon, Washington, Nevada, and western Idaho). Figure 7 shows the O_3 mean biases of the 4 simulations superposed on their predicted mean O_3 or O_3 differences. The use of MOZART BC shows the best improvement by reducing the mean bias by 2.5 ppbv and increasing the correlation coefficient/slope (Table 3). The major improvement occurred in Northwest of this domain (Figure 7B) and California coastal regions. The RAQMS BC experiment has the best correlation slope, but it also tends to increase the mean bias (Table 3). It should be noted that the highest RAQMS impact of high biases occurred not along the coast, but further inland, like Idaho (Figure 7C), which could be caused by transport from upper layers. That transport also depends on WRF-NMM and CMAQ's prediction for vertical exchange, affected by boundary layer height and convection etc. The change caused by GFS O_3 BC is relatively small over this region it is used only above 10 km and needs longer transport time/distance to affect surface ozone (Table 3 and Figure 7D). The GFS O_3 BC experiment has a stronger impact when evaluated in a region north of 43°N where tropopause folds around fronts are more prevalent. For this area, all global boundary conditions improve the correlation slopes, but only the MOZART BC reduces the mean bias and has the highest correlation coefficient/slope.

5. Conclusion

Boundary conditions can have an impact on any regional model performance. This study investigated the impacts of lateral boundary conditions from global models on ozone prediction over continental USA. The simulation period from late July to early August has several high-ozone events due to regional and local photochemical activities. It is not a typical strong inflow period, nor a noticeable scenario for Asian pollutants or biomass burning plumes to reach North America. However, time-varying global boundary conditions still show a strong impact on ozone prediction at all altitudes. Depending on locations and scenarios, the simulations with global BCs did not always yield better results than that with static BC. The error could be caused by global model performance, or CMAQ regional prediction. Previous studies (Lee, et al., 2007a) have shown that high ozone aloft can often be mixed downward too quickly by CMAQ, thereby increasing model errors in the lower troposphere.

At high altitudes, ozone predictions are mainly related to stratospheric influence, in which using GFS O_3 for LBCs show reasonable performance on capturing strong transport events, perhaps due in part to its high spatial resolution. For some events, GFS O_3 also tends to overpredict upper-troposphere ozone. It could be due to that its simplified ozone treatment does not consider photolysis loss, or the SBUV-2 satellite assimilation lacks of vertical resolution. In this study, we did not test the use of global simulations for the top boundary condition as Tang et al. (2007) did, which could yield even stronger impacts on upper atmospheric prediction. Compared to GFS O_3 BC, RAQMS and MOZART can provide full-layer BCs with more species. These species include long-lifetime CO and NO_y, which can be transported from long ranges and possibly affect regional ozone production. Therefore, the more detailed chemical boundary conditions provided from MOZART and RAQMS tended to have strong impacts on surface ozone prediction than GFS O_3 BC, especially in U.S west coast. The chemical transformation of boundary conditions needs to be evaluated in the future. The MOZART BC experiments also yielded the largest improvements for reducing the CMAQ ozone high bias, which could be due to its lower O_3 BC (Figure 1B) in low altitudes at the north and west boundaries. Finally, this study has highlighted how improvements in global air quality models could yield better results for regional air quality prediction

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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	Granier et al., 2004	GEIA/EDGAR inventory with updated Asian emission (Streets et al. 2003)	Not applicable
Biomass burning emissions	GFED-v2 (van der Werf, 2006)	ecosystem/severity based	Not applicable
stratospheric ozone	synthetic ozone constraint (McLinden et al., 2000)	OMI/TES assimilation (Pierce et al., 2007)	Initialized by SBUV-2

Table 2, Species mapping tables between RAQMS (left)/MOZART (right) species and CMAQ CBM-IV

RAQMS Species	**CMAQ CBM-IV**
CH3OOH	UMHP
HNO4	PNA
C2H6	2* PAR
OLET (terminal alkenes)	OLE1+PAR
OLEI (internal alkenes)	OLE2 + 2*PAR

MOZART Species	**CMAQ CBM-IV**
CH3OOH	UMHP
HNO4	PNA
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

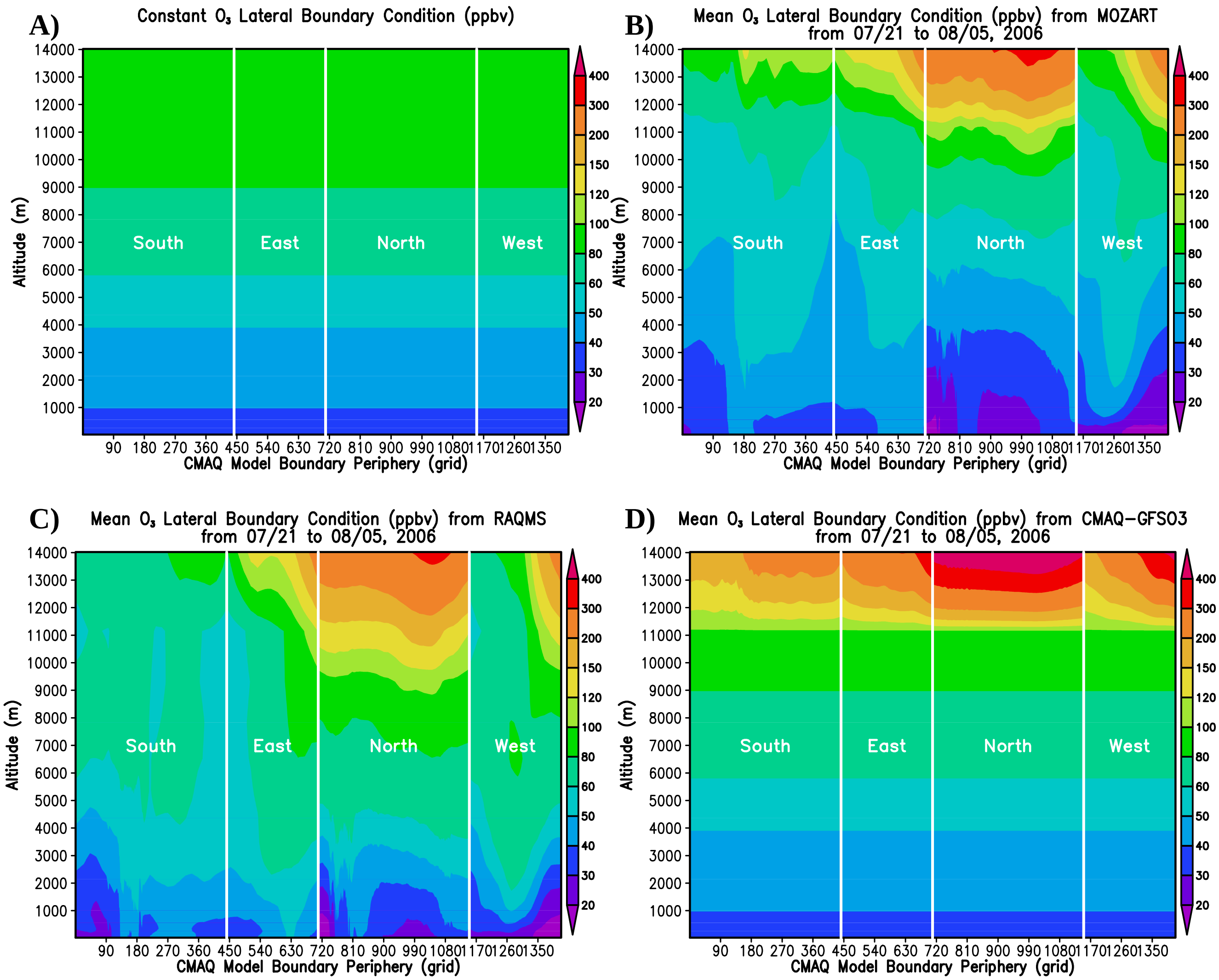


Figure 1, Mean O_3 lateral boundary conditions along the boundaries of CMAQ 442×265 (12km horizontal resolution) CONUS domain.

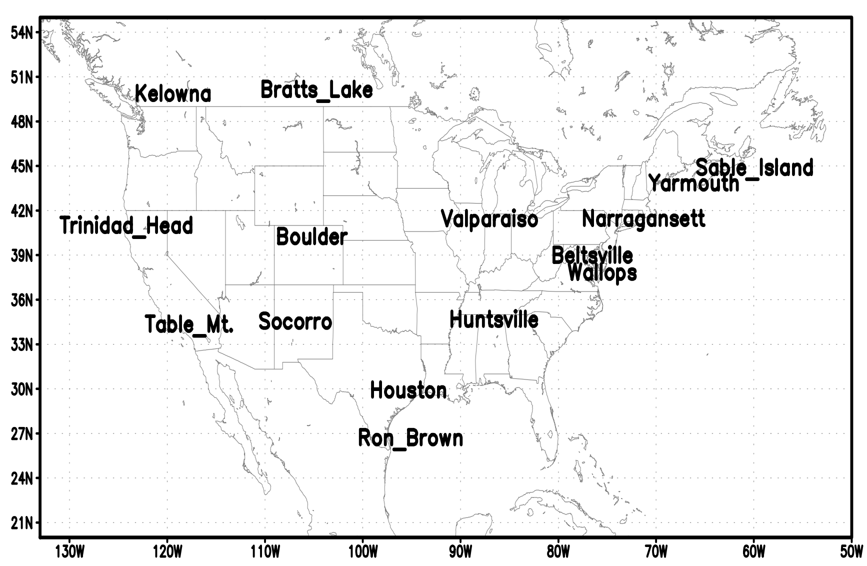
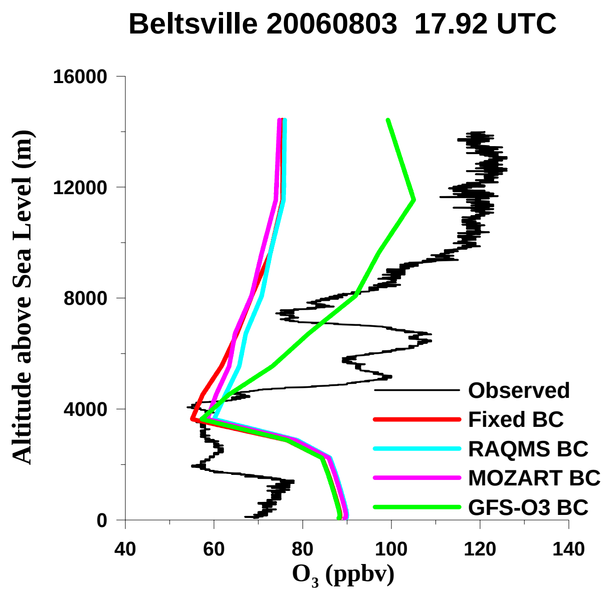
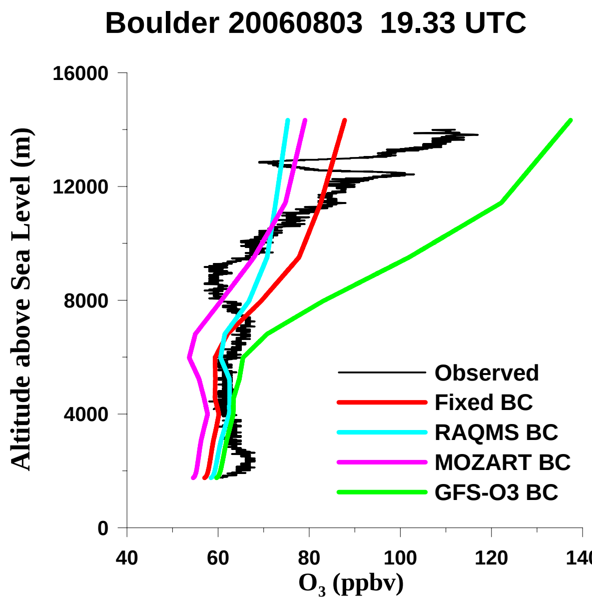
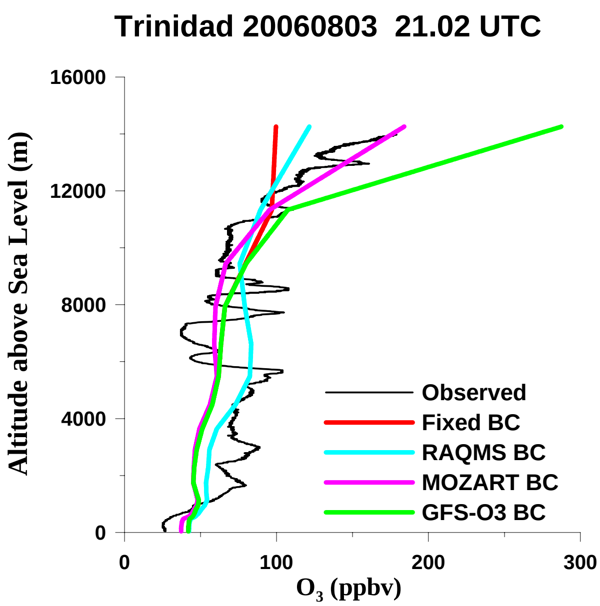
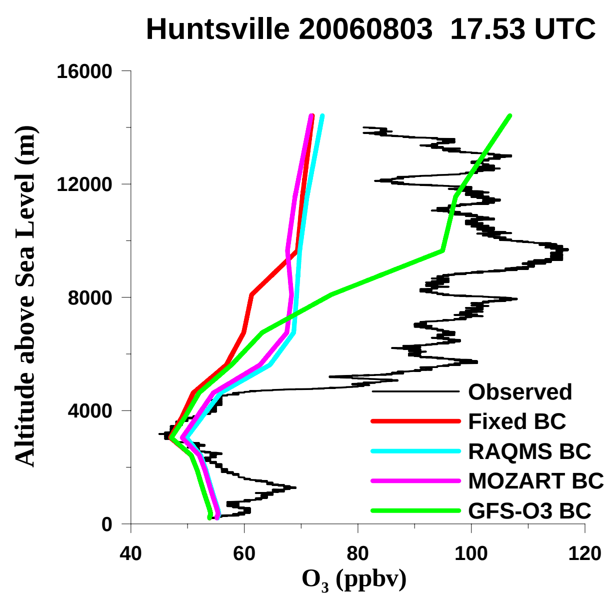


Figure 2. IONS ozonesonde sites during Aug 1-5, 2006.









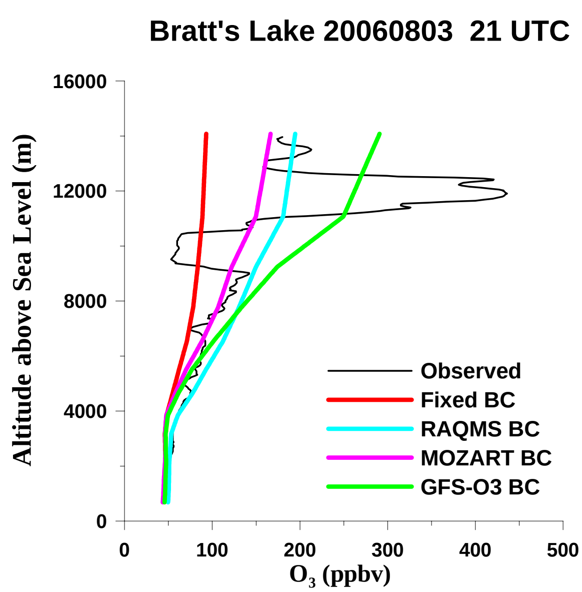


Figure 3, Four model simulations compared to IONS ozonesonde measurements at Beltsville (39.0N, 76.5W), Boulder (40.3N, 105.2W), Trinidad Head (40.8N, 124.2W), Huntsville (35.3N, 86.6W) and Bratt’s Lake (50.2N, 104.7W) on Aug 3, 2006.

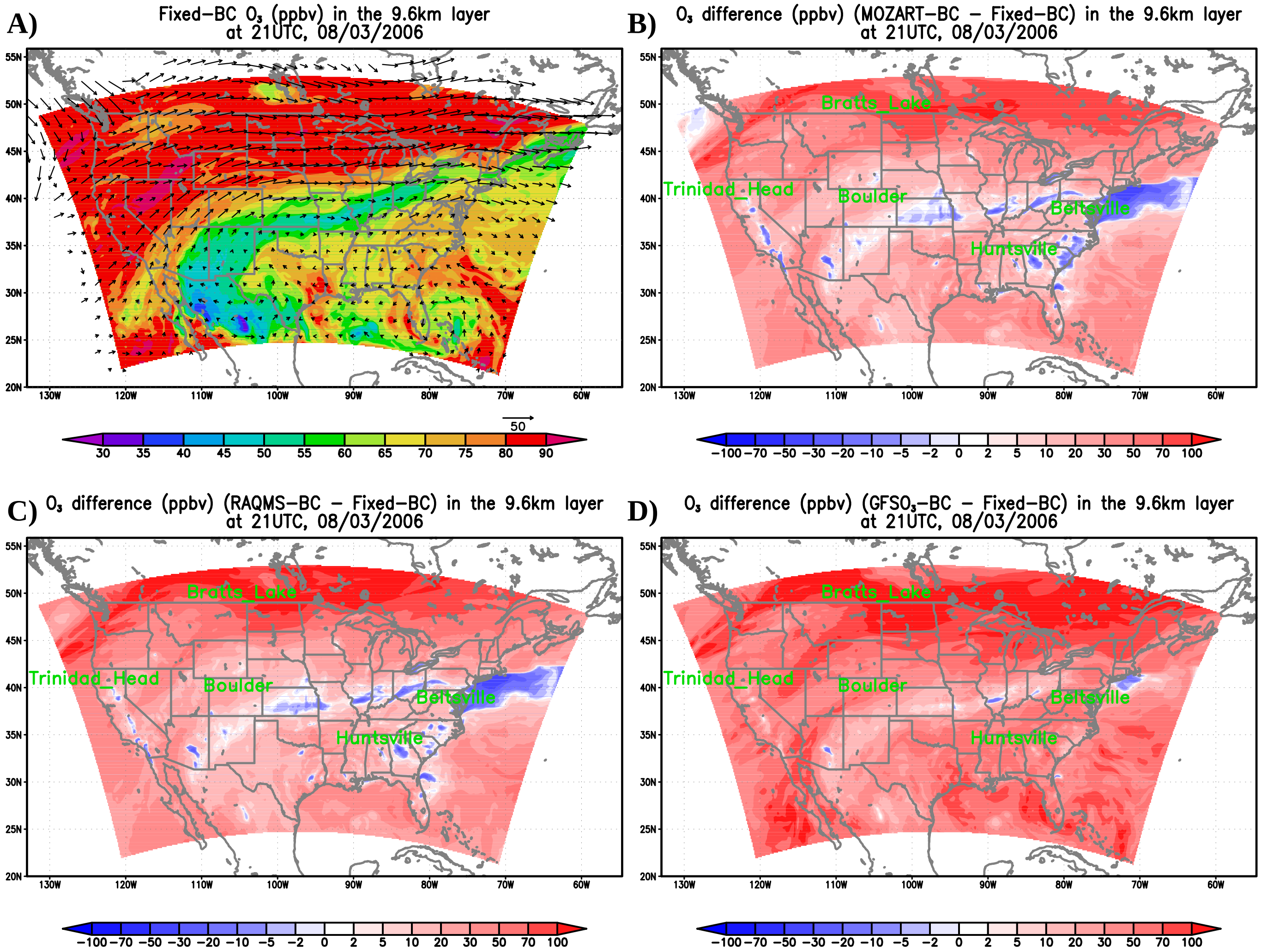


Figure 4, Model simulated ozone (A for Fixed-BC) and their differences (B, C, D) in the model's 9.6km layer at 21Z, Aug 3, 2006.

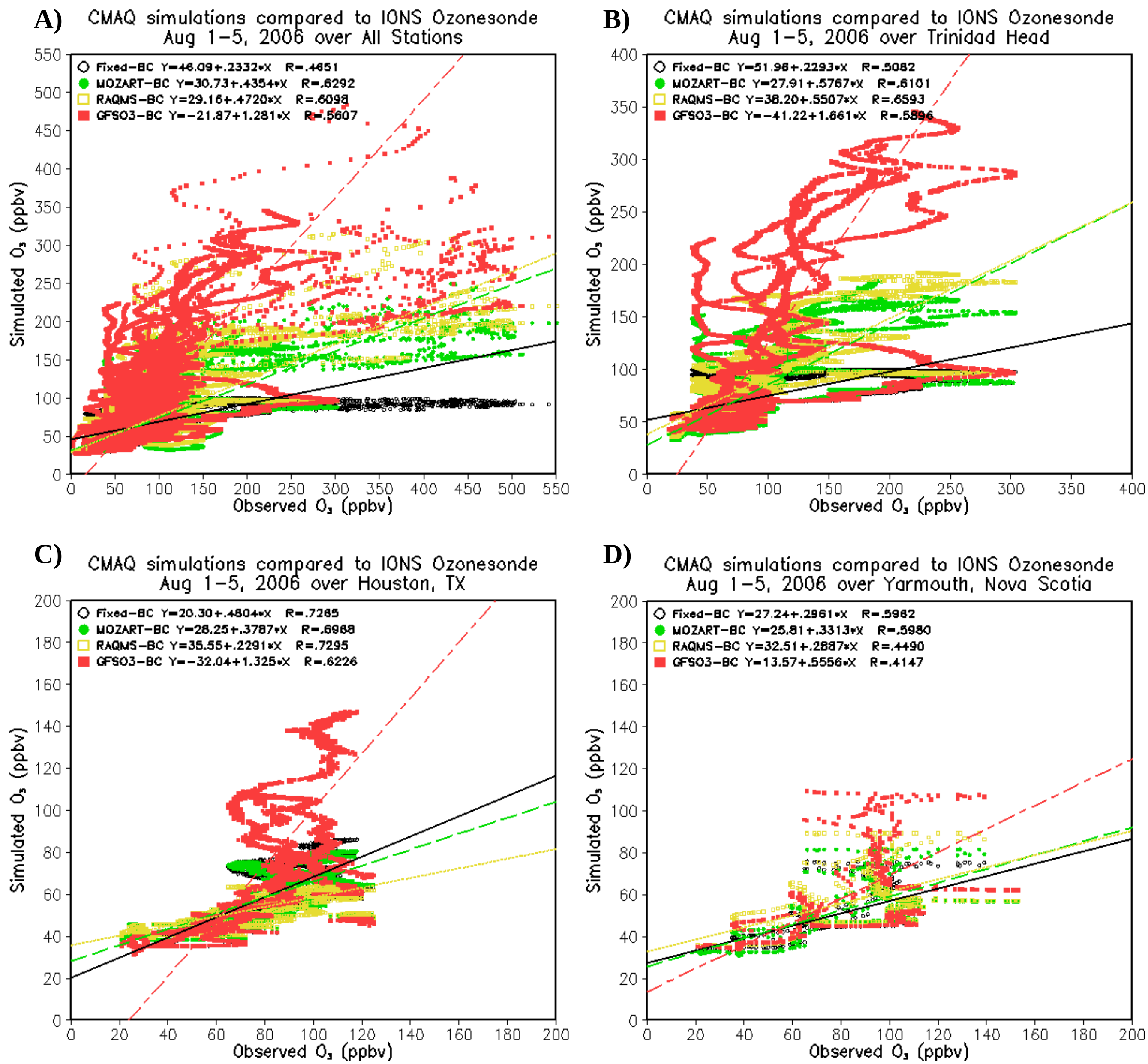


Figure 5, Statistical Correlations between the simulations and measurement over IONS stations from August 1 to August 5, 2006.

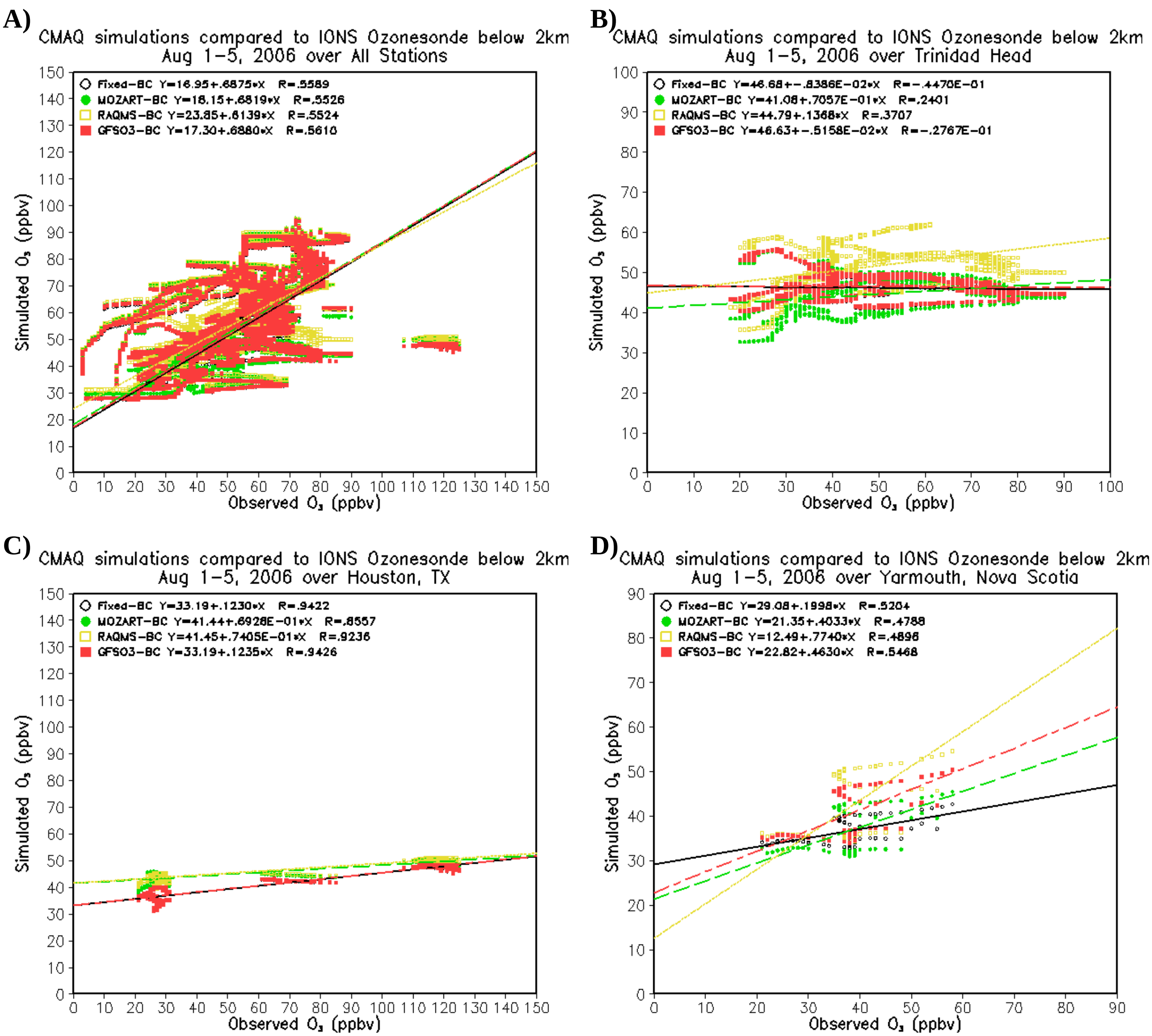


Figure 6, Same as Figure 5 but only for the altitudes below 2km.

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

	All AIRNOW Stations	**West of -115°W**	**North of 43°N**
Fixed BC	S=0.887 R=0.714 MB=8.0 ppbv	S=0.804 R=0.691 MB=4.7 ppbv	S=0.873 R=0.737 MB=7.5 ppbv
RAQMS BC	S=0.911 R=0.718 MB=10.0 ppbv	S=0.914 R=0.703 MB=7.1 ppbv	S=0.942 R=0.742 MB=10.0 ppbv
MOZART BC	S=0.941 R=0.716 MB=8.2 ppbv	S=0.872 R=0.730 MB=2.2 ppbv	S=0.985 R=0.743 MB=6.9 ppbv
GFS O_3 BC	S=0.935 R=0.714 MB=9.2 ppbv	S=0.820 R=0.697 MB=4.8 ppbv	S=0.922 R=0.724 MB=9.0 ppbv

*S is correlation slope, R is correlation coefficient, and MB is mean bias.

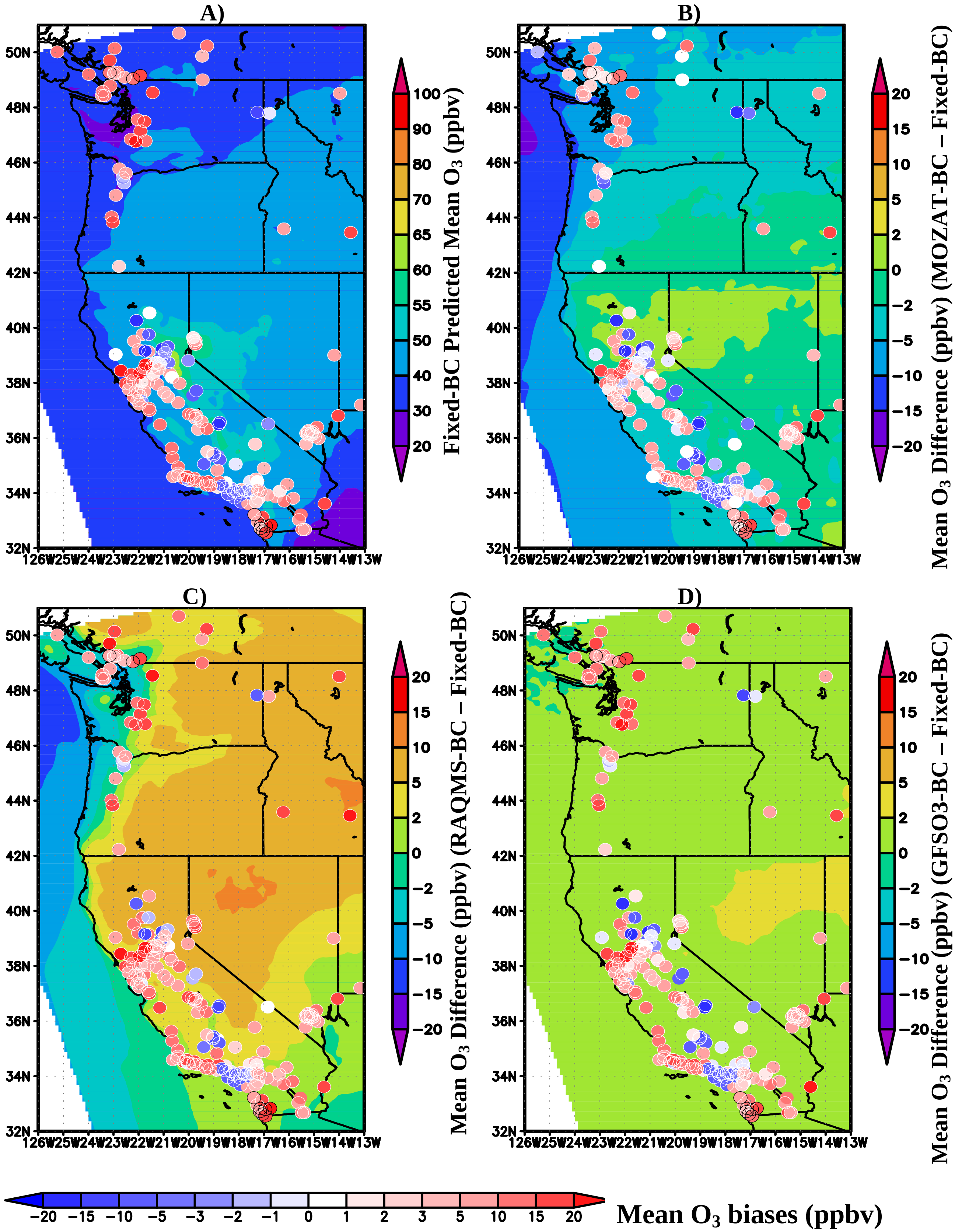


Figure 7, Mean surface O_3 prediction (plot A for Fixed-BC), mean O_3 prediction differences (B for MOZAR-BC, C for RAQMS-BC, D for GFS O_3 BC) and their mean biases from AIRNOW observations during Aug 1 to 5, 2006.