



Evaluation of the NAQFC Driven by the NOAA Global Forecast System Version 16: Comparison with the WRF-CMAQ Downscaling Method During the Summer 2019 FIREX-AO Campaign

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30 Abstract

- 31 The latest operational National Air Quality Forecasting Capability (NAQFC) has been advanced
- 32 to use the Community Multi-scale Air Quality (CMAQ) model version 5.3.1 with CB6 (carbon
- bond version 6)-Aero7 (version 7 of the aerosol module) chemical mechanism and is driven by
- 34 the Finite Volume Cubed-Sphere (FV3)-Global Forecast System, version 16 (GFSv16). This has
- 35 been accomplished by development of the meteorological preprocessor, NOAA-EPA
- 36 Atmosphere-Chemistry Coupler (NACC), which is adapted from the existing Meteorology-
- 37 Chemistry Interface Processor (MCIP). Differing from the typically used Weather Research and
- 38 Forecasting (WRF)/CMAQ system in the air quality research community, the interpolation-based
- 39 NACC can use various meteorological output to drive CMAQ (e.g., FV3-GFSv16) even though





- 1 they are in different grids. Here we compare and evaluate GFSv16-CMAQ vs. WRFv4.0.3-
- 2 CMAQ using observations over the contiguous United States (CONUS) in summer 2019. During
- 3 this period, the Fire Influence on Regional to Global Environments and Air Quality (FIREX-AQ)
- 4 field campaign was performed and we compare the two models with airborne measurements
- 5 mainly from the NASA DC-8 aircraft. The GFS-CMAQ and WRF-CMAQ systems have overall
- 6 similar performance with some differences for certain events, species and regions. The GFSv16
- 7 meteorology tends to have stronger planetary boundary layer height diurnal variability (higher
- 8 during daytime, and lower at night) than WRF over the U.S. Pacific coast, and it also predicted 9
- lower nighttime 10-m winds. In summer 2019, GFS-CMAQ system showed better surface O₃
- 10 than WRF-CMAO at night over the CONUS domain; however, their PM2.5 predictions showed 11 mixed verification results: GFS-CMAQ yielded better mean bias but poorer correlations over the
- 12 Pacific coast. These results indicate that using global GFSv16 meteorology with NACC to
- 13 directly drive CMAQ via the interpolation is feasible and yields reasonable results compared to
- the commonly-used WRF downscaling approach. 14
- 15

16 1. Introduction

17 Traditionally, mesoscale meteorological models such as the Weather Research and Forecasting Model (WRF) (Powers et al., 2017) are used as the meteorological drivers for air quality models 18 19 (AQMs) on the same ("native") model grid, such as Community Multiscale Air Quality Model 20 (CMAQ) (Byun & Schere, 2006). The NOAA National Weather Service's (NWS) National Air 21 Quality Forecasting Capability (NAQFC) has historically used a different approach, in which the 22 hourly meteorological outputs from prior operational models, such as North American Mesoscale 23 Model (NAM), need to be interpolated to the AQM grid to drive its air quality prediction. Prior 24 to this work, a "PREMAQ" coupler (Otte et al, 2004) combined both meteorological processing 25 and Sparse Matrix Operator Kernel Emissions (SMOKE) (Houyoux et al., 2000) processes, such 26 as point source plume rise effects. However, since the release of CMAQ version 5, the 27 meteorology-dependent plume rise, sea salt, and dust emission processes are included as inline 28 modules in CMAQ, and thus the corresponding emission processes are no longer needed in 29 PREMAQ. Furthermore, PREMAQ has no built-in interpolator, and thus relied on external 30 interpolators to remap the non-native-grid meteorological inputs, such as NAM, to the targeted 31 CMAQ domain, though it did perform vertical layer collapsing/interpolation to reduce layers. 32 The interpolation approach allows for more flexibility in using different meteorological drivers 33 (i.e., besides just WRF) for CMAQ; however, there is potential to raise mass-consistency issues 34 between models. It should be noted that the mass-consistency issues may also exist using native-35 grid couplers (Byun, 1999a, 1999b), which can stem from the mass-inconsistent meteorological 36 inputs or due to the temporal interpolation of the input data. The well-developed offline AQMs, 37 such as CMAQ, have already considered such mass-consistency treatments using different 38 meteorological inputs (Byun et al., 1999c).

- 39
- 40 To upgrade the NAQFC system with the latest CMAQ chemistry and NOAA operational
- 41 meteorology, we developed an updated interpolation-based meteorological coupler, the NOAA-
- 42 EPA Atmosphere-Chemistry Coupler (NACC) (Campbell et al., 2022) adapted from the U.S.





- 1 EPA's Meteorology-Chemistry Interface Processor (MCIP) version 5 (Otte and Pleim, 2010;
- 2 <u>https://github.com/USEPA/CMAQ</u>). The NACC system replaced PREMAQ, and effectively
- 3 couples the Finite-Volume Cubed-Sphere (FV3) Dynamical Core Global Forecast System
- 4 version 16 (GFSv16) (Yang et al., 2020; Harris et al, 2021) to CMAQ v5.3.1 (hereafter referred
- 5 to as GFS-CMAQ). Campbell et al. (2022) described the development and application of the
- 6 GFS-CMAQ system using NACC (in their work referred to as "NACC-CMAQ") and a
- 7 comprehensive comparison between the current (GFS-CMAQ since July 20, 2021) and previous
- 8 (NAM-CMAQv5.0.2) operational NAQFC model performances.
- 9
- 10 In this study, we analyze the impacts of the meteorological model drivers, and compare GFS-
- 11 CMAQ using NACC interpolation to the commonly-used downscaled, native-grid WRF-CMAQ
- 12 application and its impact on air quality predictions. Yu et al. (2012a, 2012b) had previously
- 13 compared the CMAQ performance driven by WRF-NMM and WRF-ARW during the 2006
- 14 TexAQS/GoMACCS field campaign, and found that the NMM-CMAQ and ARW-CMAQ
- 15 showed overall similar performance with some differences for certain events, chemical species,
- 16 and regions. Similarly, this study focuses on the comparison of GFS-CMAQ versus WRF-
- 17 CMAQ (see Section 2, Methodology), and verifies the model performance against the aircraft
- 18 observations from the Fire Influence on Regional to Global Environments and Air Quality
- 19 (FIREX-AQ) field experiment during summer 2019 (Section 3). Surface verification is also
- 20 performed using AIRNow data for August 2019 (Section 4), serving as a benchmark for the new
- 21 NAQFC versus the traditional WRF-CMAQ used in the air quality modeling community.
- 22

23 2. Methodology

24 Here we compare the two CMAQ (version 5.3.1) runs driven by the interpolated GFSv16

- 25 meteorology (GFS-CMAQ) and WRF downscaled meteorology (WRF-CMAQ). All other
- settings, such as emission and lateral boundary conditions are the same. The meteorology-
- 27 related physics is discussed in the following sections to address the models' performance
- 28 discrepancies. Both the GFS-CMAQ and WRF-CMAQ simulations are run from a period
- 29 covering 12 July 31 August, 2019, each using the last 10 days in July as the model spin-ups
- 30 that are not included in the analyses.
- 31

32 2.1 GFS Meteorological Inputs

33 The GFSv16 is the current operational global forecast system in NOAA/NCEP using FV3

34 dynamical core. Its detailed configuration can be found in Campbell et al. (2022) and Yang et al.

- 35 (2020). Compared to the previous version (v15), GFSv16 updated many physical schemes (Table
- 36 1) and added the parameterization for subgrid scale nonstationary gravity-wave drag. To use the
- 37 GFS's meteorology to drive CMAQ, a meteorological coupler, NACC, is developed (Campbell
- et al., 2022). Differing from the original MCIP which was developed to process WRF/ARW
- 39 meteorology for CMAQ, the NACC coupler interpolates non-native-grid meteorology to a user-





- 1 defined grid and has parallel processing capability, which drastically reduces its run time for
- 2 operational forecasts (Campbell et al., 2022). Currently, NACC's horizontal interpolation
- 3 employs two methods: bilinear and nearest-neighbor. In this study, we use the nearest-neighbor
- 4 method to categorical (discontinuous) variables that include land use types, vegetation fraction,
- 5 terrain elevation, Monin–Obukhov length, friction velocity, and soil temperatures, while the
- 6 bilinear interpolation is used for mainly smoothly varying (continuous) meteorological variables
- 7 that include wind fields, temperature, pressure, and specific humidity. The CMAQ model is
- 8 defined in the Arakawa C-grid (Arakawa and Lamb, 1977), and thus the GFSv16 horizontal wind
- 9 components (U, V) need to be interpolated to the perpendicular cell faces instead of the cell
- 10 center (Otte and Pleim, 2010) after rotation to the defined map projection. The scalar variables
- are defined in the target grid cell center, and thus their GFSv16 interpolations are more
- 12 straightforward. The NACC coupler can either use the native layers or collapse (i.e., interpolate)
- 13 to a set number of user-defined vertical layers for CMAQ use. The GFSv16 has 127 vertical
- 14 layers with global coverage in 13 km horizontal resolution, where the targeted domain is a 12×12
- 15 km Contiguous United States (CONUS) with 35 vertical layers (Campbell et al, 2022). Here we
- 16 use 24-hour GFSv16 forecasts starting at 12 UTC each day.
- 17

18 Most variables needed by CMAQ are directly interpolated from the GFSv16 outputs. The NACC

- 19 processor has options to calculate diagnostic variables, such as planetary boundary layer (PBL) 20 height, if they are needed. In this study, we use the interpolated GFSv16's PBL height instead of
- 20 neight, if they are needed. In this study, we use the interpolated OF SVIO'S FBL neight instead of
- the diagnostic one. It also has an option to import the externally provided land-surface variables.
- Here we import updated 2018–2020 climatological averaged leaf area index (LAI) and NOAA
- 23 near-real-time (NRT) greenness vegetation fraction (GVF) from satellite-based Visible Infrared
- Imaging Radiometer Suite (VIIRS) retrievals (Campbell et al., 2022). The updated satellite-based
- LAI and GVF impact CMAQ's biogenic emissions and dry deposition processes, which were
- 26 described in detail in Campbell et al. (2022).
- 27

28 2.2 WRF Meteorology

29 To compare with GFSv16 meteorology processed by NACC, a corresponding WRF version 4.0.3 30 (Skamarock et al, 2021) simulation is run covering the NAQFC's native grid, which is a 12 km 31 horizontal resolution, Lambert conformal map projection over CONUS. Table 1 shows the WRF 32 configuration, which is commonly employed in CONUS meteorological and air quality studies in the community, versus the current NOAA/NWS operational version of GFSv16. In contrast to 33 34 GFSv16, which is a global model that uses the NOAA/NCEP's Global Data Assimilation System 35 (GDAS) (https://www.emc.ncep.noaa.gov/data assimilation/data.html) for its initial conditions 36 and runs on its own global dynamics and physics without any other constraints, the regional 37 WRF simulation uses downscaled GFSv16 for its initial conditions. Furthermore, WRF also uses 38 downscaled lateral boundary conditions taken from GFSv16 every 6 hours. Here WRF runs 39 continuously after spin-up and we have enabled the four-dimensional data assimilation (FDDA)





1 for the u- and v-component winds, temperature, and humidity (Table 1) every 6 hours, thus

- 2 nudging towards GFSv16.
- 3

4 WRF and GFSv16 have similar settings for the land surface model, surface layer and radiation

5 schemes; however, their microphysics and PBL schemes are different (Table 1). Compared to the

6 35-layer WRF with a 100 hPa domain top, GFSv16 has a much higher domain top (0.2 hPa) and

7 127 vertical layers, which are collapsed by NACC to 35 sigma layers up to 14 km for CMAQ.

8 We use NACC (inherited from MCIP version 5.0) to process WRF hourly meteorology, while

9 maintaining the vertical layer structure. Thus, in contrast to GFS-CMAQ, the WRF-CMAQ

10 system uses the native grid without interpolation.

11

12 **2.3 CMAQ Configuration**

Here CMAQ version 5.3.1 (Appel et al., 2021) is used with the Carbon Bond 6 version r3

14 (CB6r3; Yarwood et al., 2010, 2014; Luecken, et al., 2019) chemical mechanism and Aero7

15 treatment of secondary organic aerosols (CB6r3_AE7_AQ). CMAQ 5.3.1 includes a series of

16 scientific updates from the previous version (Appel et al., 2021), including the updated air-

17 surface exchange and deposition modules, which showed significant impact on ozone prediction

18 compared to the previous NAQFC (Campbell et al., 2022). We also include the bi-directional

19 NH₃ (BIDI-NH₃) exchange model for NH₃ surface fluxes. An updated Biogenic Emissions

20 Landuse Dataset v5 (BELD5) is used in this study to drive the inline Biogenic Emissions

21 Inventory System (BEIS) version 3.61. The anthropogenic emissions are provided by the

22 National Emissions Inventory Collaborative (NEIC) with base year 2016 version 1 (NEIC 2019).

23 We replace the U.S. EPA default CMAQ dust emissions model with a novel inline windblown

24 dust model known as "FENGSHA" (Fu et al., 2014; Huang et al., 2015; Dong et al., 2016).

25 Campbell et al. (2022) include the details of the CMAQ 5.3.1 configuration for this study.

26

We have updated the wildfire emissions system in CMAQv5.3.1 based on the Blended Global Biometry Particles Product (CPREP) (71 and a decent to 2006, 71 and to 1

28 Biomass Burning Emissions Product (GBBEPx) (Zhang and Kondragunta, 2006; Zhang et al.,

29 2011). The GBBEPx uses satellite-detected fire radiative power (FRP) to estimate wildfire

30 smoke emissions for a number of species: CO (carbon monoxide), NOx (nitrogen oxides), SO₂

31 (sulfur dioxide), elemental carbon, total organic aerosols, and PM2.5. The satellite FRP is

32 estimated from satellite brightness temperature anomaly, and the GBBEPx processor assumes

that the wildfire emissions are proportional to the FRP over certain land use type in certain

34 regions. The GBBEPx emissions are based on polar orbiting satellites: MODIS (Aqua and Terra

35 satellites) and VIIRS (Suomi-NPP and NOAA-20 satellites) instruments, which are updated once

36 per day. A wildfire emission preprocessor converts the GBBEPx emissions to CMAQ-ready

37 input files using emission speciation and diurnal profiles (high during daytime and low at night)

38 (adopted from U.S. EPA-based profiles) (Baker et al., 2016), and a daily scaling factor. Here we

classify the wildfire into either a long-lasting fire (longer than 24 hours) or short-term fire

40 (shorter than 24 hours) based on land use types and regions. Only the fires west of 110° W that





- 1 have a model grid cell total forest fraction > 0.4 are assumed to be long-lasting fires, which incur
- 2 daily scaling factors of 1, 0.25, 0.25 for days 1, 2 and 3, respectively. All other short-term
- 3 GBBEPx fires are assumed to have smoke emissions for 24 hours (i.e., day 1 only). CMAQ
- 4 treats wildfire emissions as point sources that undergo inline plume rise to distribute the smoke
- 5 vertically. The default CMAQ plume rise used here is based on Briggs (1965), which is driven
- 6 by fire heat flux (converted from FRP with a ratio of 1) and fixed burning area (assumed to be
- 7 10% of the $0.1^{\circ} \times 0.1^{\circ}$ grid cell).
- 8

9 3. Model Evaluations over the U.S. for August 2019

- 10 To first gain a general picture and compare the overall GFS-CMAQ and WRF-CMAQ model
- 11 performances, in this section we evaluate near-surface meteorological and air quality predictions
- 12 during the FIREX-AQ August 2019 period against NOAA's METeorological Aerodrome Report
- 13 (METAR; <u>https://madis.ncep.noaa.gov/madis_metar.shtml</u>) and the U.S. EPA's AirNow
- 14 (<u>https://www.airnow.gov/</u>) observation networks.
- 15

16 **3.1 Domain-Wide Meteorology against the METAR Network**

- 17 Figure 1 shows the mean bias (MB) of GFS and WRF predicted surface meteorological variables
- 18 compared to METAR data during August, 2019. Both meteorological models have a cool bias
- 19 over the Western and Northeastern United States, and a warm bias over the western Rocky
- 20 Mountain region and Southeastern United States (Figure 1a, 1b). Similar temperature predictions
- 21 are expected since WRF uses the FDDA method nudging toward GFS data. However, GFS tends
- to be cooler than WRF over the Rocky Mountains and in the central and northeastern USA due
- 23 to their different dynamics and physics. The GFSv16 cold bias in the lower troposphere is
- 24 impacted by excessive evaporative cooling from rainfall (personal communication with
- 25 NOAA/NCEP). Campbell et al. (2022) had detailed discussions about GFSv16 biases.
- 26

27 Both GFSv16 and WRF models have similar and rather significant dry biases for specific

28 humidity (SH) predictions across CONUS (Figure 1c, 1d). Qian et al (2020) investigated this

- 29 common dry bias in many models, and found that neglecting an irrigation contribution could
- 30 cause this dry bias. GFS has widespread dry biases (Campbell et al. 2022) and WRF has similar

31 dry biases, too as it is nudged toward GFS. There are some noticeable differences for certain

32 regions. For instance, WRF has less dry bias over Southern Texas than GFS.

- 33
- 34 Both models underestimate the mean 10-m wind speeds compared to METAR stations over the
- 35 western U.S. WRF has stronger underpredictions over the Rocky Mountains and overpredictions
- 36 over northeastern U.S., while GFS has stronger underpredictions over the Appalachian
- 37 Mountains and overpredictions over Texas and Oklahoma. GFSv16's operational verification
- 38 also (https://www.emc.ncep.noaa.gov/gmb/emc.glopara/vsdb/v16rt2/g2o/g2o_00Z/index.html)
- 39 shows that it tends to underpredict the 10-m wind over the western U.S. during both daytime and





- 1 nighttime, but shows overprediction over the eastern U.S. Besides the difference of physical
- 2 schemes, etc. (Table 1), other possible reasons causing this surface wind difference could be
- 3 effect of the gravity-wave drag (GFSv16 includes it, but the WRF run here does not), and
- 4 vertical resolution (GFS's 127 layers versus WRF's 35 layers). Some studies (Skamarock et al,
- 5 2019) revealed the necessity of fine vertical resolution for atmospheric simulations, especially
- 6 within the PBL, near tropospheric top, and during convective events. Insufficient vertical
- 7 resolution could also cause plume dilution on chemical transport modeling (Zhuang et al., 2018).
- 8 The gravity-wave drag is also known to produce synoptic scale body forces on the atmospheric
- 9 flow over irregularities at the earth's surface such as mountains and valleys, and uneven
- 10 distribution of diabatic heat sources associated with convective systems (Kim et al., 2003). Its
- 11 parameterization is needed for large-scale models.
- 12
- 13 There is strong regional variability in the monthly mean PBL height differences between GFS
- 14 and WRF during daytime (represented by 18 UTC) and nighttime (represented by 06 UTC)
- 15 (Figure 2). During daytime, GFS has a higher PBL height compared to WRF over the U.S.
- 16 Pacific coast, northern Rocky Mountains, northeastern and southeastern U.S., but it becomes
- 17 lower over the central U.S. (e.g., Texas, Oklahoma, and Kansas). At night, however, most of
- 18 these regional differences between GFS and WRF are reversed. This diurnal difference is
- 19 mainly driven by the different PBL schemes employed in GFS (Han and Bretherton, 2019) and
- 20 WRF (i.e., YSU) and the associated other physical suites, including the land surface data. The
- 21 GFS's PBL height has a strong diurnal variation over these regions, including the western and
- 22 northeastern U.S. in the summer, including a sharp rise and collapse after sunrise and sunset,
- respectively (Campbell et al., 2022). The strong PBL diurnal variation has significant effects onthe air quality predictions in GFS-CMAQ.
- 25

26 **3.2 Evaluation of Regional Meteorology and Air Quality against the AirNow Network**

- 27 The U.S EPA AirNow network provides hourly observations of near-surface ozone, fine
- 28 particulate matter (PM2.5), and meteorology. Campbell et al. (2022) showed detailed verification
- 29 of GFS-CMAQ with the surface AIRNow data. Here we focus on the difference between the
- 30 interpolation-based GFSv16 versus WRF downscaling and the impacts on meteorological and
- 31 chemical model performances. Figure 3 shows a comparison of these two models over two
- 32 specific regions, the U.S. West (CA, OR and WA) and Northeast states (CT, DE, MA, MD, ME,
- 33 NH, NJ, NY, PA, RI, VT and District of Columbia) (Figure S1), where the two models had
- 34 relatively large differences for some meteorological variables. GFS and WRF had very similar 2-
- 35 m temperatures over the Pacific coast states: Washington, Oregon and California, and both of
- 36 them had similar cool bias (around 1K), R and RMSE (Figure 3a). However, these two models
- 37 had significant differences for 10-m wind speed prediction over the Pacific coast (Figure 3c),
- 38 where WRF overpredicted the wind speed, especially at night and in later August. Most AIRNow
- 39 stations are located near urban or suburban areas, which generally have weaker 10-m wind speed
- 40 than those at the METAR aviation weather stations near airports. For this reason, although





1 Figure 1e and 1f shows that GFS and WRF underpredict monthly-mean wind speed over the 2 METAR stations in the West, they still tend to overpredict AIRNow wind (Figure 3c), especially 3 for the WRF 10-m wind speed at night. Considering that the model grid cells represent 12×12 4 km² averages, the true model-observation comparisons likely fall somewhere between the 5 urban/suburban AIRNow stations and METAR stations, depending on the land use fractions of 6 each grid. Obviously the observation representation characteristics could affect the verification 7 results. Compared to AIRNow stations, GFSv16 has overall better scores for surface wind speed 8 predictions over the U.S. West, where the WRF's larger surface wind speed overprediction is 9 associated with its PBL height predictions (Figure 3e, 3f). During the nighttime, GFS has a lower PBL height (10-50% lower than WRF) and weaker vertical mixing, which brings less 10 11 momentum from the upper layers to the surface, which led to lower nighttime wind and better 12 agreements with the AIRNow wind-speed observation. 13 14 Over the northeast, the mean bias (MB) of GFS temperature was about -1K, while the WRF has 15 a smaller, slightly positive MB of about 0.22K (Figure 3b). However, the GFS's temperature 16 prediction has a better correlation coefficient, R, and RMSE, implying that it better captures 17 some events, such as the 28–29 of August. Both models overpredict 10-m wind speeds in the 18 northeast, but the GFS model yields better results due to a slightly lower PBLH at night (Figure 19 3f) than WRF that had significant ovepredictions, especially during 25–29, August (Figure 3d) 20 when the tropical storm Erin approached this region. Especially on 28 of August, when the storm 21 was centered near the east coast of North Carolina, the WRF run significantly underpredicts 2-m 22 temperature (Figure 3b) and overpredict 10-m wind speed (Figure 3d), implying that the some 23 WRF settings lead to relatively large surface prediction bias for the storm weather, such as its 24 relatively coarse vertical resolution compared to the 127-layer GFS model. 25 26 Figures 4a and 4b show the ozone predictions of the two models over these two regions, and 27 GFS-CMAQ yields predominantly lower O₃ than WRF-CMAQ, especially at night. Over the 28 northwest, the lower ozone in GFS-CMAQ is associated with their PBL height difference. First, 29 with a certain dry deposition velocity between the models, it is easier to deplete ozone given the smaller volume of a shallower PBL. Second, the thinner PBL results in higher NOx 30 31 concentrations and ozone titration rates near NOx source regions, and consequently lower ozone 32 there at night. Last, the lower PBL leads to weaker vertical mixing and downward transport of 33 ozone from the residual-layer at night (Caputi et al, 2019). All these factors contributed to the 34 lower nighttime ozone of GFS-CMAQ compared to WRF-CMAQ. Since GFS-CMAQ already 35 underpredicts ozone due to combined meteorological factors, such as the temperature underprediction (Figure 4a), the GFS-CMAQ's further ozone reduction (possibly due to its lower 36 37 PBLH at night) exacerbates its low bias. However, over the Northeast, the similar impacts help 38 the GFS-CMAQ yield much better MB due to its better agreement with the observed nighttime 39 low ozone over the Northeast. Over the entire CONUS domain, the situation is similar: for an 40 average August 2019, the GFS-CMAQ has a lower ozone MB (1.1 ppb) compared to WRF-





- 1 CMAQ (4.7 ppb). Figure 5 shows that both models have similar daytime ozone prediction over
- 2 CONUS. However, GFS-CMAQ better captures low nighttime ozone over the U.S. East than
- 3 WRF-CMAQ (Figure 5c, 5d).
- 4 5

6 7

- GFS-CMAQ has substantially higher PM2.5 mean concentrations over the U.S. West, but lower over the U.S. Northeast compared to WRF-CMAQ (Figures 4c, 4d). These model differences are also related to their interpolated GFSv16 versus downscaled WRF meteorological drivers.
- 8 Because both models use the same emissions under relatively clean background conditions in the
- 9 west (i.e., prevailing westerly flow from the Pacific Ocean), the PBL and wind speed differences
- 10 have significant impacts on their near-surface pollutant concentrations, especially at night. Both
- 11 models show strong PM2.5 diurnal variation (high at night and low during daytime), driven by
- 12 the meteorological diurnal variation (e.g. PBL), which overcomes the emission diurnal variation
- 13 (usually high during daytime and low at night). Compared to WRF-CMAQ, GFS-CMAQ has
- 14 lower nighttime PBL height and weaker wind speed at night, which leads to weaker vertical
- 15 mixing and venting, and increases the pollutant concentrations near the surface and yields higher
- 16 surface PM2.5 over the U.S. West (Figure 4c). Its higher surface PM2.5 could also result in
- 17 stronger local dry deposition. In contrast to local vertical mixing and venting effects on PM2.5
- discussed above, there are strong (and potentially counterbalancing) impacts of model PBL and horizontal wind speed differences on downstream PM2.5 concentrations at night. WRF-CMAQ's
- deeper PBL and stronger wind speeds at night (Figures 3c–3f) tends to transport aerosols and
- their precursors more efficiently downstream via the dominant advection pathway. Figure 6
- shows that these monthly mean background PM2.5 differences appear in East of Rocky
- 23 Mountain (WRF-CMAQ is about 2 μ g/m³ higher) during both daytime and nighttime. This effect
- 24 is very prominent in the Northeast region. Although both models predicted similar PM2.5
- 25 magnitude over the U.S. Northeast, GFS-CMAQ yields the overall PM2.5 underprediction, and
- 26 its monthly-mean PM2.5 is 2.6 μ g/m³ lower than the WRF-CMAQ prediction (Figure 4d).
- 27 Especially during 01–09 August, WRF-CMAQ had about 4 μ g/m³ higher surface PM2.5
- 28 background than that of GFS-CMAQ. In this case, the WRF-CMAQ model has a better
- agreement with observations (Figure 4d). It is possible that the GFS-CMAQ's nighttime PBL
- 30 heights (wind speeds) are too shallow (weak) in this case, which does not allow enough transport
- 31 of pollutants to the downstream (Eastern USA). Overall, GFS-CMAQ and WRF-CMAQ have
- 32 mixed performances for PM2.5 predictions during the August 2019 period: GFS-CMAQ has
- better PM2.5 prediction over the U.S West, and WRF-CMAQ yields better results over east of
- 34 Rocky mountain (Figure 6).

35 4 Model Comparisons against the FIREX-AQ Aircraft Data

36 From late July to early September, 2019, the joint NOAA-NASA FIREX-AQ field campaign

- 37 (https://csl.noaa.gov/projects/firex-aq/) employed a suite of satellites, aircraft, vehicles and
- 38 ground site platforms aimed to observe, analyze, and characterize air pollutants emitted from
- 39 wildfire sources over the CONUS (Ye et al., 2021). The FIREX-AQ airborne measurements





1 provide a three-dimensional dataset from various meteorological, gas, and aerosol instruments 2 that is used to verify the GFS-CMAQ and WRF-CMAQ model performance, while elucidating 3 reasons for any model differences. Here the focus of the FIREX-AQ model comparison and 4 verification is against observations taken primarily from the NASA DC-8 aircraft, which include 5 meteorological variables, gaseous and aerosol concentrations, and aerosol optical properties. The 6 majority of the FIREX-AQ flights were over the western United States, and sampled within 7 environments that both were and were not (see section 4.1) influenced by wildfire emissions 8 (https://daac.ornl.gov/MASTER/guides/MASTER FIREX AO JulySept 2019.html). During a 9 cluster of major wildfire events (see Section 4.2), the DC-8 sampled both near-source and aged 10 smoke plumes between 02-08 August, 2019 (i.e., the Williams Flats, Snow Creek, and Horsefly 11 Fires) across the states of Idaho, Washington, and Montana. 12 13 4.1 Comparison of the July 22 non-wildfire event over the central California Valley 14 On 22 July, the DC-8 aircraft flew from California to Boise, Idaho, while maintaining a 15 relatively low-altitude (<1 km) above sea level (ASL) over the California Central Valley (Figure 16 7). This flight was not impacted by any major wildfire event, and was mainly controlled by 17 anthropogenic emissions and local meteorological conditions. Figure 7 shows that the GFSv16 18 and WRF models had similar meteorological temperature and humidity predictions, and that both 19 models have dry and warm biases over the Central Valley at lower altitudes (Figures 7d–7e) 20 (Yun et al., 2020). GFS's horizontal wind speeds tended to have stronger variability than WRF 21 (Figure 7b), especially in high altitudes. For wind direction, WRF showed a better prediction 22 than GFS around 20 and 24 UTC (Figure 7c). 23 24 Both GFS-CMAO and WRF-CMAO underestimate the vertical wind (W) variability by at least 25 one order of magnitude, and WRF-CMAQ has weaker W variability than that of GFS-CMAQ, 26 especially in high altitudes (Figure 7f). The model vertical velocities are not from the GFS or 27 WRF model, but rather they are re-diagnosed in CMAQ to conserve mass (Otte and Pleim, 28 2010), and thus represent the whole layer's vertical movement across the 12 km by 12 km grid 29 cell. With its flight speed around 80 to 240 m/s, the DC-8 aircraft's one-minute average 30 sampling frequency results in an approximate 4.8 to 14 km horizontal scale, respectively, which 31 is comparable to the 12 km CMAQ model resolution. The aircraft observations, however, include 32 turbulence effects during its one-minute averages, which may not be temporally resolved by 33 CMAQ at this resolution. Thus, both the GFS-CMAQ and WRF-CMAQ model vertical 34 velocities are much lower and have almost no correlation with the aircraft observations. 35 36 Although both GFS-CMAQ and WRF-CMAQ have reasonable comparisons for most 37 meteorological variables, including the horizontal winds, it continues to be a challenge to 38 compare them with the observed vertical velocities. Thus to further elucidate the model vs. 39 observation differences in vertical motions, Figure 8 shows a curtain plot of vertical velocities 40 along the flight path from the two models. Since WRF-CMAQ remains in a native grid, its wind 10





- 1 fields tend to be more balanced and have lower variability compared to the interpolated GFS-
- 2 CMAQ wind fields. The stronger variability in W for GFS-CMAQ represents CMAQ's effort to
- 3 counteract mass inconsistency effects from the interpolated horizontal wind fields (Byun,
- 4 1999b).
- 5
- 6 GFS-CMAQ and WRF-CMAQ overall yield similar results for specific chemical species during 7 this DC-8 flight (Figure 9). Both models underestimate CO, O₃ and ethane (C_2H_6) concentrations 8 over the lower altitudes in the California Central Valley. Over the same flight segment, they had 9 better NOx (NO + NO₂) and ethene (C_2H_4) predictions, implying that the emissions of these two 10 species have better accuracy than those of CO and ethane. Figure 9f shows that the two models 11 also underestimate NOz (NOy–NOx), or the oxidized nitrogen species besides NOx, indicating
- that the photochemical ozone production may also be underestimated. NOz is a good indicator of
- 13 the ozone photochemical formation (Sillman et al., 1997), where the O_3/NOz ratio represents the
- 14 ozone photochemical efficiency per NOx oxidation products. Thus, NOz and O₃ are typically
- 15 highly correlated over regions with active photochemical production. The O₃ and NOz
- 16 underestimations are likely due to the underestimation of CO and some hydrocarbons, such as
- 17 ethane, as they are precursors of O₃.
- 18

19 The two models show slight differences in peak values of CO, ethene, and NOx around 23:30

20 UTC, where the GFS-CMAQ predicted concentrations are slightly higher and closer to

21 observations (Figure 9). These differences are due to their PBL predictions (both from the

- 22 corresponding meteorological model outputs), where GFS-CMAQ has a lower PBL height and
- 23 weaker emission vertical dilution compared to WRF-CMAQ (Figure 8). GFS-CMAQ tends to
- 24 underpredict O₃ more (Figure 9b), however, due to its higher NOx titration. This implies that the
- 25 effects of the transport and non-local transformation of O₃ could be stronger than that of local
- 26 precursor emissions. WRF-CMAQ has higher NOz (Figure 9f), but lower NOx compared to
- 27 GFS-CMAQ due to the time lag of O₃ and NOz photochemical formation. Consequently, the
- $28 \qquad \text{peak O}_3 \text{ values may not be well correlated with the emitted precursors, such as NOx and volatile}\\$
- $\label{eq:compounds} \ organic \ compounds \ (VOCs). \ Furthermore, the modeled \ peak \ C_2H_6 \ and \ C_2H_4 \ concentrations \ do$
- 30 not occur at the same time around 23:30 UTC, while observations indicate that these two species
- 31 should be highly correlated in this region. This model mismatch implies that the VOC speciation
- 32 factors for a certain area or emission sector need to be improved over Southern California.
- 33

34 **4.2** Comparison of the 6 August wildfire events over the U.S. Northwest

35 On 06 August, the DC-8 observed a cluster of three wildfires: the Williams Flats Fire (47.98 °N,

36 118.624 °W, 80 km to the northwest of Spokane, Washington), Snow Creek Fire (47.703 °N,

- 37 113.4°W, 32 km northeast of Condon, Montana), and Horsefly Fire (46.963 °N, 112.441°W, 24
- 38 km east of Lincoln, Montana). Figure 10a shows the flight path on that date, where the DC-8
- 39 aircraft departed from Boise, ID, flew over the Williams Flats Fire region, then flew to Montana
- 40 to sample the Snow Creek and Horsefly Fires (i.e., Montana Fires), and finally returned to the





- 1 Boise base. The aircraft flew below 8 km for most flight segments near the fire plumes. Figure
- 2 S1 shows the corresponding GOES-16 satellite true color image, where these 06 August fires and
- 3 associated smoke plumes are visible and can be distinguished from the cloud bands to the south
- 4 that move northward later that day (Figure S2). The Williams Flats Fire was ignited by
- 5 lightning, and was the largest fire event sampled during the FIREX-AQ campaign burning from
- 6 about 02–08, August, 2019.
- 7

8 Both models significantly underpredicted CO (Figure 10c), submicron organic aerosol (Figure

- 9 10e) and aerosol optical extinction coefficient (AOE) (Figure 10f), which suggests an issue with
- 10 the GBBEPx gas and aerosol emissions. The models performed well for NO₂ during the
- 11 Williams Flats and Montana Fires Fire below 6 km ASL, but there were prominent

12 underestimations for the high-altitude flight segments (Figure 10d). This indicates that the

- 13 background NO₂ was underestimated, or the models had insufficient inject height for fire plume
- 14 rise (both based on Briggs, 1965). WRF-CMAQ predicted higher O₃ values than the GFS-
- 15 CMAQ, which overall agreed better with observations for the Williams Flats Fire (Figure 10b).
- 16 However, for the Montana Fires (~ 23–24 UTC), WRF-CMAQ has higher O₃ biases and GFS-
- 17 CMAQ yields better results. The difference in O₃ is largely driven by the background
- 18 concentration difference between the two models, where WRF-CMAQ tends to have higher
- $19 \qquad \text{domain-wide O}_3 \text{ than GFS-CMAQ} \text{ due to the meteorological effects discussed in Sections 3}.$
- 20

21 Figure S3 shows the spatial overlay comparison of vertically averaged GFS-CMAQ predictions

22 at 21 UTC and the DC-8 flight observations for the altitude 1–3 km above ground level (AGL),

- 23 on 6 August, 2019. The peak NO₂ observation around 118.5°W, 48°N indicates the general
- 24 location of the Williams Flats fire. The GBBEPx emission and GFS-CMAQ prediction showed
- 25 shifted peak-value locations driven by the westerly modeled winds. For this flight, the GBBEPx
- 26 had stronger NOx fire emission over two Montana locations than that over Williams Flats. The
- 27 model overpredicts the column averaged NO_2 concentrations, especially over the Montana fires,
- 28 which can not be reflected by the point-by-point NO₂ comparison result in Figure 10d. For this
- 29 flight, the mean GFS-CMAQ NO₂ along the flight path for 1-3km AGL is about 0.125 ppbv
- 30 compared to the observed mean NO_2 of 0.169 ppbv, and the model indeed showed NO_2

31 underprediction along the flight path. However, in this case, the flight path missed some

- 32 locations where modeled peak NO₂ values existed or the modeled transport misplaced the
- 33 plumes, especially over the Montana fires leading to this inconsistency. For ozone comparison
- 34 (Figure S3b), this inconsistency could also exist, though not as significant as for the high-
- 35 gradient NO₂ concentrations. In the GFS-CMAQ prediction, the high ozone concentrations are
- 36 almost co-located with high NO₂ concentration (Figure S3b), but the observation did not show
- 37 this feature. Instead, some high-O₃ flight segments had relatively low NO₂, such as those circled
- 38 in the black rectangle box of Figure S3b. The observed NOx titration was not able to be
- 39 produced by the 12 km models. Wang et al (2021) used a 100m horizontal resolution large eddy
- 40 simulation and demonstrated the capability of using such techniques to capture some high-





- 1 resolution fire plume features and associated chemical behavior. While such high resolution
- 2 techniques are not currently feasible for the operational NAQFC, they demonstrate the limitation
- 3 of using regional scale (12×12 km) models to capture such fine scale features of plume behavior.
- 4
- 5 GFS-CMAQ has higher wildfire-related CO, NO₂, OA and AOE values that are closer to
- 6 observations than WRF-CMAQ for the Montana Fires between 23–24 UTC at flight altitudes of
- $7 \sim 4-5$ km (Figure 10c–10f). Since these two models use the same GBBEPx emissions and
- 8 wildfire plume rise algorithm (Briggs, 1965), the differences should be due to other reasons. To
- 9 help explain these model differences, Figure 11a and 11b show the Differential Absorption High
- 10 Spectral Resolution Lidar (DIAL-HSRL) retrieved aerosol backscatter coefficients (ABC)
- 11 aboard the DC-8 aircraft without and with cloud screen, respectively. It shows that the major fire
- 12 plumes of the William Flats Fire were below 4 km (~ 19–22 UTC), but the Montana Fires (~23–
- 13 24 UTC) extended from the surface up to 6 km, with some detached plumes reaching 10 km. The
- 14 model predicted AOEs have an overall similar pattern, with major plumes below 4 km for the
- 15 Williams Flats Fire (Figures 11c and 11d). Over the Montana Fires, the GFS-CMAQ predicts
- 16 slightly higher PBL, thus allowing for the fire plume to reach a higher height near the DC-8
- 17 cruising altitude. In contrast, the WRF-CMAQ wildfire plumes are slightly lower than the
- 18 aircraft flight path around 23–24 UTC, which leads to underprediction in the fire emitted species
- 19 (Figure 11d).
- 20

21 An interesting feature in the DIAL observations is the detached plume from 8 km to 10 km

22 altitude (Figure 11a), where some cirrus clouds existed, and the DIAL retrieval could not

23 distinguish whether they are pure clouds or clouds mixed with elevated aerosols above 8km. The

- cloud screen product (Figure 11b) mainly showed the enhanced aerosols below 7km and some
- 25 scattered signals near the high cloud edges. Cloud mixing with aerosols was usual for fire
- 26 induced clouds, or pyrocumulonimbus (Peterson et al., 2021). Although in this event, the middle-
- 27 size fires did not show evident of inducing the high-altitude clouds, the indicator of mixed clouds
- and aerosols in high altitudes still existed: both in-situ measured OA (Figures 10e) and AOE
- 29 (Figure 10f, 11c, 11d) showed the enhanced aerosols around 25 UTC above 8km. This elevated
- 30 plume was generally captured by the GFS-CMAQ simulation, while underestimating its strength
- 31 (Figure 11c); however, this feature was completely missed in WRF-CMAQ (Figure 11d).
- 32 Considering the altitude range of the detached plume, the major model disparities are likely due
- 33 to model convection differences in the free troposphere. To further investigate this impact,
- 34 Figures 11e and 11f show curtain plots of RH predicted by the two models. GFS-CMAQ yields
- 35 higher RH at such altitudes (10 km) compared to WRF-CMAQ around 23–24 UTC, indicating
- 36 that the GFS-CMAQ has stronger convection. The CMAQ model uses inputted meteorology to
- 37 diagnose convection activity and drive its ACM2 convection scheme. This convective activity is
- 38 apparent in GOES-16 satellite images (Figure S2), as more fractional clouds appeared ahead of
- 39 the northward moving frontal band. Both the GFSv16 and WRF models used here *do not*
- 40 consider the fire heat feedback effect, and thus their predicted convection and clouds are only





- 1 driven by the synoptic weather conditions. If such synoptic-to-mesoscale weather models
- 2 consider wildfire heat feedback effects, their predictions may result in stronger convection and
- 3 help correct underpredictions in PBL heights.
- 4

5 4.3 Statistical Results of Model Performances for FIREX-AQ

- 6 4.3.1 Meteorological Statistics
- 7

8 During the FIREX-AQ field campaign, the DC-8 aircraft performed more than 20 flights over 9 CONUS with detailed observations of various chemical compounds. Tables 2 and 3 show the 10 statistical results of mean bias (MB), normalized mean bias (NMB), root mean square error (RMSE), correlation coefficient (R), and linear regression/slopes for the two models' 11 12 performance over the western U.S. (west of 110°W) only at low altitudes (<3km ASL) for both 13 non-fire and fire flight segments. Most of these flights departed from Boise, ID, except the 22 14 July flight that flew from California to Idaho. As a result, they mainly flew over Idaho and its 15 surrounding regions. The GFS tends to have slightly higher wind speed with positive MB, while 16 WRF has a small negative wind speed bias. Most of the DC-8 flights are during the daytime, and 17 the GFS has a higher daytime wind speed than WRF at low altitudes. The GFS and WRF have 18 very similar temperature predictions. For the RH, the GFS predictions were slightly dryer than 19 those of WRF, especially for non-fire events. The meteorological models do not consider 20 wildfire heat effects, and thus may have (in part) led to slightly warm MB for the non-fire events 21 (Table 2) and slightly cool MB for the fire events (Table 3). Because both the GFSv16 and WRF 22 models have similar MB shifts from an average temperature overprediction (Table 2; non-fire 23 events) to an underprediction (Table 3; wildfire events), we can estimate that the fire effects 24 made roughly a 1–2 Kelvin temperature enhancement to the background along the DC-8 flight 25 paths below 3 km. This estimate assumes that the model temperature biases are generally 26 representative of the western U.S. (west of 110°W), and are independent of the averaged flight 27 segments that have different locations and periods in Table 2 and Table 3. Correspondingly, the 28 air masses are dryer in the sampled wildfire plumes, as shown by the large reduction in the RH 29 underpredictions (i.e., negative MBs) from Table 2 to Table 3. 30 31 4.3.2 Chemical Statistics During Non-Fire Events

32

For most chemical species, the two models also have similar performance, indicating that the
 emissions and chemistry are major driving forces. For non-fire events, both models overpredict

35 NOx, HNO₃, toluene, EC, and ammonium (NH₄⁺), but underestimate PAN, benzene, C₂H₂, SO₂,

and submicron sulfate and organic aerosols (OA) (Table 2). The SO₂ and submicron sulfate

37 underprediction may be impacted by underestimated NEIC2016v1 SO₂ emissions over the

38 western U.S. Since point sources, including power plant emissions, are the SO₂ sources, this

- 39 comparison implies that the point sources for 2019 events have large uncertainties.
- 40





- 1 Although the models agree well with NOy observations, they disproportionately underestimate 2 NOz as shown by the regression slopes and MBs. One of the important NOz species is PAN, and 3 both models underestimate PAN during the non-fire events (Table 2). PAN's carbonyl 4 precursors include acetaldehyde (CH₃CHO) (44% of the global source), methylglyoxal (30%), 5 acetone (7%), and a suite of other isoprene and terpene oxidation products (19%) (Fischer et al., 2014). CH₃CHO and acetone are also underestimated (Table 2), and help explain PAN's 6 7 underestimation. For the oxidized hydrocarbons, like aldehydes (HCHO, CH₃CHO), their main 8 atmospheric sources are the oxidation of highly reactive VOCs, including alkanes, alkenes, and 9 aromatics, instead of direct emissions (Parrish et al., 2012). So, the underestimation of HCHO 10 and CH3CHO are associated with the underestimation of their precursor hydrocarbons, including 11 anthropogenic and biogenic VOCs. Our other comparison indicated that BEIS tends to 12 underpredict biogenic emission over the U.S. West, e.g. isoprene in Table 2. In this comparison, 13 most anthropogenic hydrocarbons are disproportionately underestimated, except toluene, 14 implying the VOC speciation issue in the NEIC2016v1 anthropogenic emissions (Table 2). 15 Previous work had discovered that a model overprediction in toluene was also related to the 16 toluene speciation in the NEI emission inventory (Lu et al., 2020). 17 18 Submicron ammonium (NH4⁺) and the nitrate ion are also underestimated by both models during 19 non-fire events (Table 2), suggesting there are NH₃ underestimates due to either insufficient NH₃ 20 emissions or exaggerated NH₃ removal processes. There are, however, overpredictions in the 21 intermediate species nitric acid (HNO₃). It implies that the HNO₃ accumulates in the atmosphere 22 because the modeled NOz pathways toward the nitrate ion and organic nitrate aerosol products 23 are reduced due to their other precursor (NH₃ and VOCs) underestimation. 24 25 There are underestimations in the VOC and CO concentrations, which contributes to the ozone 26 underestimation during non-fire events (Table 2). These non-fire comparisons also highlight that 27 both models have similar biases due to similar meteorology (Section 4.3.1), and the use of the 28 same anthropogenic emissions (NEIC2016v1), BEIS biogenic emission and chemical 29 models/mechanisms (i.e CMAQv5.3.1). The differences in the two models' bias, error, and 30 correlation/slope are much smaller than their individual magnitudes. 31 32 4.3.3 Chemical Statistics During Fire Events 33 The WRF-CMAQ and GFS-CMAQ models significantly underestimate CO, VOC, HONO, and
- 34 OA for fire events at low altitudes (< 3 km) over the western U.S. (Table 3). In conjunction with
- 35 underestimated GBBEPx emissions during these wildfire events, other possible causes for the
- 36 average statistical underprediction are the CMAQ model's 12 km horizontal resolution and the
- 37 flight sampling coverage. Most of the fires that are averaged in the statistics, such as the Horsefly
- 38 (5.5 km² burning area) and Snow Creek Fires (7.3 km² burning area), are at a much finer scale
- than the model grid. Only the largest Williams Flats Fire, with a total burning area of 180 km²
- 40 (Ye et al., 2021), had a comparable horizontal scale to the model resolution.





1

2 The DC-8 aircraft had many flight segments near wildfire sources during the fire events in Table

- 3 3, and thus dilution of the emissions due to the relatively coarse model resolution may lead to
- 4 underestimations in the predicted slope for most wildfire emitted pollutants, such as CO and OA
- 5 (Table 3). The O_3 concentrations are also underestimated; however, the O_3 underpredictions are
- 6 reduced from the non-fire (Table 2) to fire events (Table 3). Abundant amounts of wildfire
- 7 emitted NOx can titrate ozone near the fire source region, and the models likely underestimate
- 8 these titration effects due to the 12 km model resolution. Thus, the models cannot capture the
- 9 strong spatial O₃ variability that is observed due to both reduction near source regions and
- 10 enhancement in downstream areas. Again, for this fire event comparison, both models showed
- similar behavior and their differences were relatively smaller compared to the overall model
- 12 biases.
- 13

14 **5. Summary and Discussion**

15 The operational NOAA/NWS National Air Quality Forecasting Capability (NAQFC) recently

- 16 underwent a major upgrade on July 20, 2021. The advanced NAQFC includes the recent
- 17 Community Multi-scale Air Quality (CMAQ) model version 5.3.1 with CB6 (carbon bond
- 18 version 6)-Aero7 (version 7 of the aerosol module) chemical mechanism, and is driven by the
- 19 latest operational Finite Volume Cubed-Sphere (FV3)-Global Forecast System, version 16
- 20 (GFSv16) (Campbell et al., 2022). Here we analyze the impacts of the driving meteorological
- 21 models on CMAQ model performance with the new GFSv16 interpolation-based meteorology
- 22 versus the commonly-used native-grid Weather Research and Forecasting (WRF) model version
- 23 4.0.3 meteorology. The meteorological and chemical analysis includes both 2D ground-based
- 24 and 3D aircraft measurements during the summer 2019, which encompasses the joint NOAA-
- 25 NASA Fire Influence on Regional to Global Environments and Air Quality (FIREX-AQ)
- 26 campaign. As CMAQ has existing mass conservation via adjustments of the contravariant
- 27 vertical velocity (Otte and Pleim, 2010), the NACC interpolated GFSv16 wind field can be well
- 28 handled in CMAQ (i.e., GFS-CMAQ).
- 29

30 The different NWS/NOAA operational GFS and commonly chosen WRF physics schemes

31 employed in this study (Table 1) clearly have impacts on temperature, wind fields (both

32 horizontal/advection and vertical/convection), PBL heights, and the corresponding CMAQ

- 33 model predictions. During this study period over the U.S. West, both models showed moisture
- 34 dry bias and temperature warm bias in low altitudes, which could be due to the issue mentioned
- 35 by Qian et al (2020) and impacts from soil moisture deficits on surface fluxes in both models.
- 36 Due to their different physics, GFS had stronger diurnal variation of PBL height, lower at night
- and higher during daytime over the U.S. West and Northeast. The differences in the GFS and
- 38 WRF physics have a larger impact than the meteorology driver methodologies (i.e., interpolation
- vs. native) on the models' meteorological and air quality predictions, even despite using FDDA
- 40 to nudge WRF simulation toward the GFSv16 data. While FDDA nudging was used here in





- 1 WRF to avoid growing errors across a continuous 1-month simulation, we note that if it is turned 2 off, the differences between GFSv16 and WRF predictions would have been even greater. This
- 3 would further substantiate the dominance of using different model physics compared to using
- 4 different meteorological-driver methodologies (i.e., native vs. interpolation) and their impacts on
- 5 CMAQ model predictions. Overall, the results of this study further corroborate the use of the
- 6 GFSv16 data and NACC interpolation-based methods (Campbell et al., 2022) for regional
- 7 CMAQ model applications in the scientific community.
- 8
- 9 Over CONUS, GFS-CMAQ demonstrated lower mean surface ozone (by about 3 ppb) and
- 10 PM2.5 (by about $1 \mu g/m^3$) than WRF-CMAQ in August 2019 (section 3). In the western U.S.,
- 11 the GFS has a stronger diurnal variability in PBL height and a better performance in nighttime
- 12 10-m wind speeds compared to WRF. The nighttime difference between these two models tends
- 13 to be more significant than the corresponding daytime difference. Their difference is also
- 14 impacted by both vertical/convective (mainly daytime) and upstream advective transport
- 15 differences in GFS-CMAQ and WRF-CMAQ, which somewhat confounds the impact of
- 16 different meteorological physics on chemical predictions from region to region. This transport
- 17 effect is more significant on PM2.5 than that on O₃, as O₃ has a shorter lifetime and is more
- 18 sensitive to local emissions in summer. In this study, neither GFS-CMAQ nor WRF-CMAQ
- 19 show overwhelming performance advantage over the other, similar to the NMM-CMAQ and
- 20 ARW-CMAQ comparison in Yu et al. (2012a, 2012b).
- 21
- 22 GFS-CMAQ and WRF-CMAQ demonstrated rather similar performance for major chemical
- 23 variables during both FIREX-AQ non-fire (Table 2) and fire events (Table 3). In most FIREX-
- AQ events, both GFS-CMAQ and WRF-CMAQ showed similar biases, indicating that other
- 25 factors, including emissions, model resolution and chemistry etc. could be more important for the
- 26 model predictions compared to the meteorological differences. The aircraft data comparison
- reveals many common issues in both model systems. One critical issue is whether the flight
- 28 sampling coverage is comparable to the 12 km model resolution, especially for high-gradient fire
- 29 emission, e.g. the case of 06 August flight (Figure S3). The observation representation issue also
- 30 exists in other places, such as near-surface meteorological comparison between AIRNow stations
- 31 and METAR stations. Emission is the driving force for atmospheric composition concentrations.
- 32 The comprehensive aircraft measurements help verify that the anthropogenic NEIC2016v1
- 33 inventory is overall reasonable, except for SO₂, NH₃ and certain hydrocarbons. The wildfire
- 34 emission has bigger uncertainties, including the emission intensities, specification and plume
- 35 rise, shown by the both models' results.
- 36

37 The NACC interpolation method is advantageous as it enables using the original meteorological

- 38 driver directly via interpolation, and avoids running another model such as WRF as a downscaler
- 39 for regional CMAQ applications. It is also faster, and more consistent with the original
- 40 meteorological driver. These aspects can simultaneously benefit real-time forecasting and



1



2 quickly use any regional domain globally, and may also use other global meteorological data 3 including reanalysis products. This helps mitigate the confounding factors of using different 4 model configurations across the myriad of WRF physics options, while alleviating the difficulty 5 in understanding their impacts on air quality predictions. The operational GFSv16 and associated 6 reanalysis products are well vetted and evaluated across different global agencies and 7 laboratories, and thus are well suited for regional CMAQ applications using NACC. In fact, there 8 is an ongoing project at NOAA to migrate both the GFSv16 data and NACC software to the 9 Amazon Web Services (AWS) Cloud platform to provide a streamlined product for the user to 10 generate the model-ready meteorological data for any regional CMAQ application globally. 11 12 Finally, we note that the current operational GFSv16 has enough meteorological variables to 13 drive CMAQ with other supplied data (fractional landuse, LAI etc), and its C768 grid has 14 horizontal resolution from 10.21 km to 14.44 km, which is close to the NAOFC's 12 km 15 horizontal resolution. However, some commonly available global meteorological data, such as 16 NCEP or ECMWF reanalysis data, may not have all meteorological variables needed by CMAQ, 17 and have relatively coarse model resolutions. In this case, the WRF downscaling may become the only available method to drive a finer scale CMAQ model application. WRF data generated 18 19 by different physics may be good for a finer scale CMAQ simulation; however NACC 20 developments are underway to also process/interpolate higher resolution FV3-based Limited 21 Area Models (LAMs) for direct application to CMAQ. All the physics schemes were developed 22 according to certain regions and meteorological conditions. We again stress, however, that the 23 downscaled WRF physics may significantly alter the original meteorological fields even with the 24 FDDA nudging. As shown in this study, GFS and WRF had mixed performance for driving 25 CMAO.

retrospective air quality applications in the scientific community. NACC can also adapt to

26

27 Code and Data Availability.

The FIREX-AQ field campaign data used in this study is in <u>https://www-air.larc.nasa.gov/cgi-bin/ArcView/firexaq</u> (last access, 16 May 2022). The NACC code used in this study is publicly available at https://doi.org/10.5281/zenodo.5507489 and via GitHub at https://github.com/noaa-oar-arl/NACC.git (last access: 5 April 2022). The modified CMAQv5.3.1 for GFS-CMAQ is available at https://doi.org/10.5281/zenodo.5507511 and via GitHub at https://github.com/noaa-oar-arl/NAQFC (last access: 5 April 2022).

34

35 Author contributions.

36 YT contributed to the project conceptualization, model run, software, data analysis, visualization,

37 investigation, and writing of the original draft. PCC contributed to software, the model run, data

- analysis, investigation and draft revision. DT and XZ contributed to wildfire emissions data. BB
- 39 contributed to software and funding acquisition. FY, JH and HH provided the GFS model data.
- 40 LP provided the global aerosol model for the lateral boundary condition. PL, RS, AS, JF, IS, JT-





- 1 D, YJ contributed to project supervision, project administration, and funding acquisition. MY,
- 2 IB, JF, TR, DB, JS, J-LJ, JC, GD, RM, JH, GH, AR and JD contributed to the FIREX-AQ
- 3 aircraft data.
- 4
- 5 Competing interests.
- 6 The contact author has declared that neither they nor their co-authors have any competing7 interests.
- 8

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- 12

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1 Table 1. The two meteorological datasets used in this stu-	o meteorological datasets used in this	study
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Model Settings	FV3-GFSv16/NACC	WRF-ARW/MCIP
Domain	Global C768L127 (~ 13 km horizontal resolution in 6 cubic spherical tiles , 127 vertical layers up to 80km), interpolated to the 12km CONUS domain with 35-layers up to about 14km (60hPa)	12km CONUS 35 vertical layers up to 100hPa
Dynamic core	Finite Volume 3, non-hydrostatic (Putman and Lin, 2007)	WRF-ARW dynamic in hybrid vertical coordinate (Skamarock et al., 2021)
Initial condition	FV3-GFSv16 analysis (GDAS) using the local ensemble Kalman filter (LETKF) (Ott et al., 2004) with 4- dimensional incremental analysis update (4D-IAU)	FV3-GFSv16 analysis (GDAS)
Lateral Boundary Condition	N/A	FV3-GFSv16 analysis (GDAS)
Cloud Microphysics	GFDL six-category cloud microphysics scheme (Lin et al., 1983; Lord et al., 1984; Krueger et al., 1995; Chen and Lin, 2011; Chen and Lin, 2013)	Morrison 2-moment scheme (Morrison et al., 2009)
PBL Physics Scheme	Scale-aware (sa) turbulent kinetic energy (TKE) -based moist eddy- diffusivity mass-flux (EDMF) (sa- TKE-EDMF) (Han and Bretherton,2019)	Yonsei University Scheme (Hong et al., 2006)
Shallow/Deep Cumulus Parameterization	SAS Scheme (Han et al. 2011; 2017)	Kain Fritsch multiscale (Kain, 2004)
Shortwave and Longwave Radiation	RRTMG (Mlawer et al. 1997; Clough et al. 2005; Iacono et al. 2008)	RRTMG (Iacono et al. 2008).
Land Surface Model	Noah Land Surface Model (Chen and Dudhia 2001; Ek et al. 2003;Tewari et al. 2004)	Noah (Tewari et al., 2004)
Surface Layer	Monin-Obukhov (Monin-Obukhov 1954; Grell et al. 1994; Jimenez et al. 2012)	Revised MM5 Scheme (Jimenez et al., 2012)
Other treatment		FDDA nudging is enabled for temperature and specific humidity whole domain, and for wind components (U, V) outside the PBL.





	Obs	GFS-CMAQ					WRF-CMAQ				
Variables	Mean	MB	NMB	RMSE	R	Slope	MB	NMB	RMSE	R	Slope
Temperature (K)	295	0.979	0.332	2.04	0.988	1.13	1.16	0.393	2.28	0.989	1.17
RH (%)	35.6	-7.3	-20.5	11.8	0.781	0.717	-6.05	-17	12.6	0.677	0.598
Wind Speed											
(m/s)	4.81	0.758	15.8	3.25	0.432	0.473	-1.11	-23.1	2.4	0.666	0.524
O ₃ (ppbv)	57.9	-10.7	-18.5	15	0.651	0.34	-10.4	-17.9	14.1	0.717	0.413
CO (ppbv)	134	-37.6	-28	53.2	0.654	0.573	-37.1	-27.7	52.9	0.652	0.572
NOx (ppbv)	1.11	0.507	45.6	2.9	0.704	1.15	0.345	31.1	2.86	0.695	1.12
NOy (ppbv)	2.56	-0.0418	-1.63	3.07	0.743	0.892	0.055	2.15	3.14	0.724	0.86
NOz (ppbv)	1.63	-0.465	-28.6	1.17	0.782	0.553	-0.125	-7.66	1.08	0.788	0.721
HONO (ppbv)	0.00432	0.012	279	0.0438	0.379	0.444	0.0134	311	0.0487	0.358	0.48
HNO ₃ (ppbv)	0.291	0.154	53.1	0.421	0.683	1.34	0.337	116	0.65	0.708	1.89
PAN (ppbv)	0.399	-0.251	-63	0.416	0.675	0.221	-0.222	-55.6	0.386	0.681	0.284
NH ₃ (ppbv)	3.55	-0.801	-22.6	5.26	0.0481	0.038	-1.58	-44.5	4.37	0.304	0.155
C ₂ H ₄ (ppbv)	0.121	0.0582	48.1	0.189	0.702	0.869	0.0385	31.9	0.187	0.682	0.836
C ₂ H ₂ (ppbv)	0.146	-0.0734	-50.3	0.137	0.784	0.496	-0.0696	-47.7	0.137	0.771	0.494
SO ₂ (ppbv)	0.342	-0.235	-68.8	0.567	0.0238	0.00835	-0.221	-64.5	0.568	-1.26×10 ⁻³	-0.00047
Acetone (ppbv)	2.74	-2.28	-83.1	2.45	0.686	0.192	-2.2	-80.4	2.38	0.668	0.199
HCHO (ppbv)	2.1	-0.972	-46.4	1.26	0.559	0.447	-0.909	-43.4	1.25	0.513	0.442
CH ₃ CHO (ppbv)	0.736	-0.326	-44.2	0.538	0.647	0.386	-0.349	-47.4	0.554	0.643	0.38
Benzene (ppbv)	0.0449	-0.0193	-43	0.057	0.398	0.385	-0.0191	-42.6	0.0564	0.397	0.375
Toluene (ppbv)	0.039	0.0409	105	0.153	0.759	1.74	0.0352	90.1	0.14	0.762	1.63
Isoprene (ppbv)	0.073	0.0361	49.4	0.174	0.6	0.838	0.00661	9.06	0.145	0.648	0.797
EC											
(µg/std m ³)	0.108	0.191	177	0.572	0.518	2.09	0.228	211	0.609	0.455	1.88
OA											
$(\mu g/\text{std m}^3)$	10.9	-7.15	-65.7	9.72	0.565	0.263	-6.48	-59.5	9.45	0.495	0.243
Sulfate											
$(\mu g/\text{std m}^3)$	1.31	-0.781	-59.7	1.11	0.0856	0.0188	-0.773	-59	1.11	0.0322	0.00677
\mathbf{NH}_4	0.745	0 6 1 5	00 E		0.416	0 102	0 506	70.0	0 770	0 500	0 1 4 5
(µg/sta m ⁺)	0.743	-0.015	-02.5	0.805	0.410	0.103	-0.590	-19.9	0.778	0.509	0.145
$(ug/std m^3)$	1 22	-1 08	-88 1	1 49	0 562	0 229	-1 04	-85 3	1 45	0.57	0 279
AOE (/Mm)	54.5	-29.3	-53.8	47	0.593	0.227	-27.4	-50.2	45.9	0.588	0.227

Table 2. Statistics of the two models compared to the observation for DC-8 flight segments with non-fire events below 3 km (ASL) over west of -100° W. All aerosols are in submicron. The normalized mean bias (NMB) is in unit %.





	Obs	GFS-CMAQ				WRF-CMAQ					
Variables	Mean	MB	NMB	RMSE	R	Slope	MB	NMB	RMSE	R	Slope
Temperature (K)	287	-0.389	-0.135	0.702	0.995	1.01	-0.688	-0.24	0.863	0.997	1.04
RH (%)	27.8	-0.761	-2.74	7.84	0.712	0.553	4.3	15.5	11.1	0.556	0.534
Wind Speed (m/s)	5.42	0.766	14.1	2.16	0.612	0.616	-0.811	-15	2.12	0.604	0.556
O ₃ (ppbv)	55.7	-6.61	-11.9	11.8	0.587	0.262	-7.01	-12.6	11.5	0.653	0.346
CO (ppbv)	486	-377	-77.6	873	0.596	0.0347	-383	-78.8	883	0.442	0.0242
NOx (ppbv)	2.63	0.06	2.28	6.41	0.465	0.231	-0.619	-23.5	7.02	0.31	0.153
NOy (ppbv)	7.32	-4.19	-57.3	13.3	0.507	0.123	-4.66	-63.7	14.2	0.31	0.073
NOz (ppbv)	5.7	-4.8	-84.3	10.2	-0.189	-0.0106	-4.68	-82	10.2	-0.204	-0.0121
HONO (ppbv)	0.283	-0.274	-96.8	1.18	0.355	0.0043	-0.274	-96.8	1.18	0.291	0.00457
HNO ₃ (ppbv)	0.148	0.148	99.7	0.256	0.532	1.07	0.179	121	0.28	0.402	0.768
PAN (ppbv)	0.971	-0.793	-81.7	1.63	0.27	0.0195	-0.765	-78.8	1.61	0.279	0.026
NH ₃ (ppbv)	17.7	-12.3	-69.3	28.3	0.379	0.0654	-13.7	-77.4	29.6	0.232	0.0386
C ₂ H ₄ (ppbv)	4.5	-4.34	-96.3	10.2	0.421	0.00498	-4.36	-96.8	10.2	0.14	0.0018
C ₂ H ₂ (ppbv)	1.04	-1.01	-96.9	2.08	0.534	0.00866	-1.01	-97	2.09	0.363	0.00623
SO ₂ (ppbv)	0.699	-0.322	-46.1	1.38	0.589	0.198	-0.392	-56.1	1.5	0.429	0.132
Acetone (ppbv)	3.54	-3.2	-90.3	4.56	0.13	0.00862	-3.18	-89.7	4.55	0.135	0.0112
HCHO (ppbv)	8.17	-7.13	-87.3	17.8	0.232	0.0062	-7.19	-88	17.8	0.119	0.00303
CH ₃ CHO (ppbv)	3.65	-3.18	-87.4	9.13	0.186	0.00547	-3.21	-88	9.2	-0.027	-0.00097
Benzene (ppbv)	0.683	-0.67	-98.1	1.84	0.54	0.00432	-0.672	-98.3	1.84	0.367	0.00275
Toluene (ppbv)	0.451	-0.436	-96.6	1.36	0.402	0.00491	-0.438	-97	1.36	0.195	0.00245
Isoprene (ppbv)	0.095	-7.9×10 ⁻³	-8.29	0.234	0.123	0.0579	-0.033	-34.7	0.242	-0.014	-0.00541
EC (ug/std m ³)	1.89	-0.53	-28	3.28	0.612	0.295	-0.787	-41.6	3.7	0.448	0.195
OA (µg/std m ³)	156	-146	-93.4	420	0.612	0.0174	-147	-94.2	423	0.472	0.0122
Sulfate (µg/std m ³)	0.791	-0.116	-14.7	0.676	0.415	0.184	-0.214	-27.1	0.728	0.322	0.13
NH_4^+ (µg/std m ³)	1	-0.591	-59.1	0.931	0.767	0.351	-0.615	-61.5	0.956	0.729	0.359
Nitrate (µg/std m ³)	1.7	-0.56	-32.9	1.47	0.805	0.613	-0.634	-37.2	1.59	0.774	0.599
AOE (/Mm)	391	-350	-89.3	994	0.688	0.027	-357	-91.1	1010.	0.532	0.0152

Table 3, same as Table 2 except for the wildfire affected flight segments.







c) GFS mean 2m specific humidity bias (g/kg), 08/2019 d) WRF mean 2m specific humidity bias (g/kg), 08/2019



e) GFS mean 10m wind speed bias (m/s), 08/2019

f) WRF mean 10m wind speed bias (m/s), 08/2019



Figure 1. GFS and WRF surface meteorological biases for METAR (METeorological Aerodrome Report) stations averaged over August, 2019





a) Monthly Mean PBL Difference (GFS-WRF) at 18UTC

Monthly Mean PBL Difference (GFS-WRF) at 06UTC

b)



Figure 2. Monthly mean PBL height difference (GFS-WRF) for daytime (a) and nighttime (b), August, 2019.







Figure 3. The WRF and GFS time-series comparison over AIRNow stations over the U.S. West and Northeast for 2m temperature (a, b), 10m wind speed (c, d), and PBL height (e,f).















Figure 5. Monthly mean surface ozone predictions by GFS-CMAQ (left plots) and WRF-CMAQ (right plots) for daytime (top plots) and nighttime (bottom plots) compared to the corresponding AIRNow observations, August, 2019







PM2.5 (µg/m³)

Figure 6, same as figure 5 but for surface PM2.5







Figure 7. Modeled meteorological variables compared with observations for the DC-8 flight on 22 July, 2019 (b to f). The plot a shows the flight path colored in altitudes above sea level with UTC time in red text. Base map credits: © OpenStreetMap contributors 2022. Distributed under the Open Data Commons Open Database License (ODbL) v1.0.







Figure 8, Curtain plots of the vertical velocity (W) predicted by GFS-CMAQ (a) and WRF-CMAQ (b) along the DC-8 flight on 22 July, 2019. The colored dots showed the DC-8 measured vertical velocities. The solid lines showed the predicted PBL heights of these two models.







Figure 9. Model predicted chemical concentrations compared with observations along with the DC-8 flight on 22 July, 2019







Figure 10. The DC-8 flight path (a), and model-observation comparisons for O_3 (b), CO (c), NO₂ (d), submicron organic aerosol (OA) (e) and aerosol optical extinction coefficient (AOE) at wavelength of 550nm (f) on 06 August, 2019. Base map credits: © OpenStreetMap contributors 2022. Distributed under the Open Data Commons Open Database License (ODbL) v1.0.







Figure 11, The Differential Absorption High Spectral Resolution Lidar (DIAL-HSRL) retrieved aerosol backscatter coefficient (ABC) at 532nm wavelength in unit /km/steradian (a) and cloud screened one (b); curtain plots of the AOE (b, c) and relative humidity (RH) (d, e) predicted by GFS-CMAQ (left) and WRF-CMAQ (right) along the DC-8 flight on 06 August, 2019. The colored dots showed the corresponding measured values. The solid lines showed the predicted PBL heights of these two models.