# The impact of observation nudging on simulated meteorology and ozone concentrations during DISCOVER-AQ 2013 Texas campaign

3

4 Xiangshang Li<sup>1</sup>, Yunsoo Choi<sup>1</sup>, Beata Czader<sup>1</sup>, Anirban Roy<sup>1</sup>, Hyuncheol Kim<sup>2,3</sup>, Barry Lefer
5 <sup>1</sup>, Shuai Pan<sup>1</sup>

6

<sup>1</sup>Department of Earth and Atmospheric Sciences, University of Houston, Houston, TX, 77204,
USA

<sup>9</sup> <sup>2</sup>NOAA Air Resources Laboratory, College Park, MD 20740, USA

<sup>3</sup>University of Maryland, Cooperative Institute for Climate and Satellite, College Park, MD,
USA

12 Corresponding author: Xiangshang Li: xli@central.uh.edu

## 13 Abstract

14 Accurate meteorological fields are imperative for correct chemical transport modeling. 15 Observation nudging, along with objective analysis, is generally considered as a low-cost and 16 effective technique to improve meteorological simulations. However, the meteorological impact 17 of observation nudging on chemistry has not been well characterized. This study involved two 18 simulations to analyze the impact of observation nudging on simulated meteorology and ozone 19 concentrations during the 2013 Deriving Information on Surface conditions from Column and 20 Vertically Resolved Observations Relevant to Air Quality (DISCOVER-AQ) Texas campaign 21 period, using the Weather Research and Forecasting (WRF) and Community Multiscale Air Quality (CMAQ) models. The results showed improved correlations between observed and 22 23 simulated parameters. For example, the index of agreement (IOA) improved by about 9% for 24 surface temperature and 6-11% for surface zonal (U-WIND) and meridional (V-WIND) winds 25 when observation nudging was employed. Analysis of a cold front event indicated that nudging 26 improved the timing of wind transition during the front passage. Observation nudging also

27 reduced the model biases for the planetary boundary layer (PBL) height predictions.

Additionally, the IOA for CMAQ simulated surface ozone improved by 6% during the

29 simulation period. The high ozone episode on September 25<sup>th</sup> was a post-front ozone event in

30 Houston. The small-scale morning wind shifts near the Houston Ship Channel combined with

31 higher aloft ozone early morning likely caused the day's ozone exceedance. While observation

32 nudging did not recreate the wind shifts on that day and failed to reproduce the observed high

33 ozone, analyses of surface and aircraft data found that observation nudging helped model yield

34 improved ozone predictions. In a two-hour period during the event, substantially better winds in

35 the sensitivity case noticeably improved the ozone. The average IOA for ozone in the period

36 increased from just over 0.4 to near 0.7. Further work on improving the capability of nudging to

37 reproduce local meteorological events such as stagnations and wind reversals could enhance a

38 chemical transport model's skill to predict high ozone events.

39 Keywords: WRF, CMAQ, air quality model, DISCOVER-AQ, observation nudging

40

## 41 **1. Introduction**

42 Meteorological variables such as cloud fraction, winds, planetary boundary layer (PBL) heights 43 and precipitation significantly impact air quality. They influence the production, transport, and 44 deposition of various chemical species (e.g., Banta et al. 2005; Pour-Biazar et al. 2007; Cuchiara 45 et al. 2014). Hence accurate meteorological inputs are imperative for air quality modeling. 46 Common approaches of improving meteorological simulations include the selection of updated 47 and high resolution terrain data (e.g., Cheng and Byun 2008), optimization of physics and 48 dynamics options (e.g., Zhong et al. 2007) and the implementation of Four Dimensional Data 49 Assimilation (FDDA) (e.g., Stauffer and Seaman 1994).

Four Dimensional Data Assimilation continuously merges new observational data into model simulation such that the model predictions do not drift away from observations. There are several FDDA methods including nudging (e.g., Stauffer and Seaman 1994) and Variational Methods (3D-VAR or 4D-VAR; e.g., Le Dimet and Talagrand 1986; Huang et al. 2009). 4D-VAR obtains optimal states of the atmosphere using multi-time-level observations by globally adjusting a model solution to all available observations over an interval of time. Nudging is a simple yet 56 flexible FDDA method originally developed by Stauffer and Seaman (1990, 1994), and 57 implemented in the Fifth-Generation PSU/NCAR Mesoscale Model (MM5). Not intended for 58 optimal adjustment, nudging is less computationally intensive but needs special care for the 59 nudging coefficients. Nudging involves adding an artificial tendency term to one or more model 60 prognostic equations that reflect the difference between the best estimate of the observed state and the model state at a given location and time. In short, the goal is to "nudge" the model state 61 62 towards the observed state. There are several types of nudging such as 3D analysis nudging, 63 surface analysis nudging, and observation nudging (obs-nudging). In the case of analysis 64 nudging, the model state is nudged toward gridded analysis. The difference between 3D and 65 surface analysis nudging is that 3D analysis (at all model levels except for surface) data are used 66 in 3D analysis nudging to improve 3D fields while in surface analysis nudging only surface 67 analysis are used to improve surface fields. In observation nudging, the model predictions are 68 nudged to match better with observations at individual locations both surface and aloft. The MM5 nudging codes were later improved and incorporated into the Weather Research and 69 70 Forecasting (WRF) model by Liu et al. (2005, 2006). The enhancements enable obs-nudging to 71 assimilate a large variety of direct or derived observations. In WRF, the inputs for obs-nudging 72 are generated by the WRF OBSGRID program. This program also performs Objective Analysis 73 (OA) to improve the quality of analysis nudging files. Objective Analysis updates first guess 74 meteorology analysis by incorporating observational data. Since obs-nudging is usually 75 performed along with OA (as in this study) to maximize the benefits of assimilating 76 observations, we also use OA to denote the combined Objective Analysis and obs-nudging 77 processes in case names.

78 The benefit of applying nudging to improve meteorological simulations has been demonstrated 79 by several studies (e.g., Deng 2009; Gilliam and Pleim 2010; Rogers et al. 2013). However, the 80 impact of the improved fields on air quality simulations has been investigated by relatively fewer 81 studies. Otte (2008) showed that the Community Multiscale Air Quality (CMAQ) model with 82 improved MM5 meteorology using analysis nudging was able to better simulate ozone chemistry 83 as reflected in model-measurement statistics. Their results indicated that better "model skill" 84 scores were achieved for daily maximum 1-hr ozone mixing ratios after analysis nudging over a 85 35-day simulation episode. Byun et al. (2008) performed over a dozen tests on obs-nudging (with 86 analysis nudging turned on) and showed obs-nudging improved both winds and temperature in

87 MM5 simulations. The study also gave an example in which improved wind fields on a given 88 day helped the CMAQ model better capture the ozone hotspot southwest of Houston. Ngan et al. 89 (2012) compared results from several CMAQ simulations coupled to the MM5 model which 90 included nudging. Their results indicated that fully nudged simulations (with both analysis 91 nudging and obs-nudging implemented) performed better than a forecast one with respect to both 92 meteorology and ozone chemistry. However, the statistics from their study cannot be used for 93 interpreting the sensitivity of obs-nudging since its base WRF case is a forecast run which used a 94 different analysis input. Previous studies by the current authors (e.g., Rappenglueck et al. 2011; 95 Czader et al. 2013) showed that obs-nudging helped correct errors in model wind fields, critical 96 to the transport of pollutants and production of secondary pollutants. To the best of the authors' 97 knowledge, there is no comprehensive study on the impact of obs-nudging on air quality 98 simulation using the WRF model.

99 This study intends to fill up the gap in the studies mentioned above by investigating the 100 sensitivity of WRF and subsequently, CMAQ simulations to observation nudging. In theory, 101 higher frequency of obs-nudging input should have a higher likelihood to capture small scale 102 events, such as local wind shifts. These events may only slightly impact local weather, yet have a 103 marked effect on chemistry. This is because local stagnation and wind convergence/reversals can 104 contribute to the pollutant build-up, as indicated by Banta et al. (1998), Cheung and Wang 105 (2001) and Tucker et al. (2010).

106 There is a significant presence of petrochemical facilities, power plants and motor vehicles in the 107 Houston-Galveston-Brazoria (HGB) region located in southeastern Texas (SETX). The major 108 pollutant in the region is ozone due to the abundant emissions of precursors like nitrogen oxides 109 (NO<sub>x</sub>) and Volatile Organic Compounds (VOCs). Summertime ozone concentrations often rise above the National Ambient Air Quality Standard (NAAQS). Consequently, HGB has been 110 111 designated as an ozone nonattainment region by the US Environmental Protection Agency 112 (USEPA) (http://www3.epa.gov/airquality/greenbook/hncs.html#TEXAS). The petrochemical 113 plants are largely concentrated in the Houston Ship Channel (HSC) area - just north of the 114 Galveston Bay. The VOCs emitted from the HSC area are highly reactive and have been shown 115 to contribute greatly to the high regional ozone episodes (e.g., Kleinman et al. 2002; Daum et al. 116 2004). Depending on the local meteorology, the plumes from HSC may be carried to different

117 locations in HGB and trigger high ozone events in its path. Metropolitan Houston has a high

118 level of NO<sub>x</sub> emissions partly due to heavy urban vehicular traffic. As a result, relatively

119 frequent high ozone events occur in the area.

120 Due to the reasons listed above, the Houston-Galveston-Brazoria region has been the focus of

several air quality studies in the recent past (e.g., Banta et al. 2005; Parrish et al. 2009; Lefer and

122 Rappengluck 2010; Olaguer et al. 2013; Czader et al. 2013, Choi et al. 2012; Choi 2014; Choi

and Souri, 2015; Pan et al. 2015). It is a good place for studying ozone production and transport

124 due to the existence of a dense surface monitoring network, as well as several intensive

125 measurement field campaigns which provide ample observational data. For example in

126 September 2013, the National Aeronautics and Space Administration (NASA), joined by a

127 number of agencies and universities, conducted a field measurement campaign in SETX as part

128 of its the Deriving Information on Surface conditions from Column and Vertically Resolved

129 Observations Relevant to Air Quality (DISCOVER-AQ) program (<u>http://www-</u>

130 <u>air.larc.nasa.gov/missions/discover-aq/discover-aq.html</u>). This program has conducted several

131 air quality and meteorology measurements at different locations in the U.S. The availability of

dense surface observations is important for obs-nudging to correct erroneous local winds in the

133 model. The performance of obs-nudging will be handicapped without a rich set of observations.

134 This study involved performing two sets of WRF and CMAQ model simulations for the 2013 135 DISCOVER-AQ Texas time period in order to understand the impact of obs-nudging. The data 136 for nudging included multiple sets of observation data from both surface and aloft measurements. 137 We evaluated model-measurement performance statistics for both WRF and CMAQ output. The 138 paper is structured as following: Section 1 is introduction; Section 2 describes the measurement 139 data and the modeling system; Section 3 covers the evaluation protocols; Section 4 discusses the 140 general meteorological conditions during the campaign period; Section 5 presents the modeling 141 results, and Section 6 provides discussions and conclusions.

## 142 **2. Observational Data and Model Configurations**

## 143 2.1 Observational Data

144 This study used regular measurements from the Continuous Ambient Monitoring Stations

145 (CAMS) operated by the Texas Commission on Environmental Quality (TCEQ). Additionally,

- 146 planetary boundary layer (PBL) and aloft ozone measurement data were obtained from the
- 147 DISCOVER-AQ campaign. For observation nudging, CAMS data and several data streams from
- 148 the Meteorological Assimilation Data Ingest System (MADIS) were used. The CAMS
- 149 measurement network collected real-time meteorology and pollutant data. The measured
- 150 parameters differ from station to station. The station density at South East Texas (SETX) is
- relatively high. There were 63 and 52 sites with meteorological and ozone measurements
- 152 respectively in the 4-km domain. The network is represented in Figure 1. The sites are
- 153 represented by dots, with the La Porte (C556) site labeled. All CAMS observations are accessible
- 154 online at the TCEQ website: <u>http://www.tceq.state.tx.us/cgi-</u>
- 155 <u>bin/compliance/monops/daily\_summary.pl</u>.



158 **Figure 1**. Locations of CAMS sites (dots) in CMAQ 4-km modeling domain during September

- 159 2013. Metro Houston, Houston Ship Channel, Galveston Bay and Gulf of Mexico are labeled.
- 160 Additionally, PBL height measurements for September were recorded at a site at the University
- 161 of Houston main campus. The PBL height was measured using the Light Detection and Ranging
- 162 (LIDAR) system. The PBL data is currently available only at this site. Primarily due to a
- 163 significant number of cloudy days, the PBL data coverage is incomplete (about 50%). For

analysis of aloft ozone, we also used measurements from aircraft P-3B, part of the rich datasets
 collected during DISCOVER-AQ campaign. The P-3B data had over 100 parameters which are
 accessible online.

## 167 2.2 Model Configurations

The modeling system consists of the WRF meteorological model (Skamarock et al., 2008), the Sparse Matrix Operator Kernel Emissions (SMOKE) model for emissions modeling (Houyoux et al., 2000) and the CMAQ model (Byun and Schere, 2006) for chemical transport modeling. The details about model configurations are presented in the following sections. Two sets of simulations were conducted, one set with obs-nudging and OA and the other without. The base case, referred as "No-OA", did not employ obs-nudging or OA. The second case, "1Hr-OA", performed obs-nudging and OA using hourly nudging input.

## 175 **2.2.1. WRF Setup**

176 Both WRF simulations used the same nested domain and NARR (North American Regional 177 Reanalysis) as input, with grid nudging turned on. Figure 2 depicts the horizontal domain setup. 178 Two nested domains were used, with 12-km and 4-km resolution respectively. The 4-km domain 179 covered SETX and a small portion of Louisiana. The 12-km domain (red box) encompassed 180 Texas and parts of a few neighboring states. The number of grid cells for the 12-km and 4-km 181 domains were 161×145 (E-W by N-S), and 95×77 respectively. The projection type was Lambert 182 conic conformal (LCC). Three projection parameters, first latitude (33 %), second latitude 183 (45 N) and standard longitude (97 W) are conformal to the projection parameters used by the 184 USEPA in developing emission inventories for air quality modeling (Mason et al. 2010). Both 185 domains had a vertical resolution of 27 eta layers based on dry hydrostatic pressures. The model 186 top is 100 hPa, corresponding to top layer pressure of the input NARR data.



Figure 2. Horizontal domains of WRF and CMAQ simulation at 4 km and 12 km grid resolution
(the bigger domains are for 12 km WRF and CMAQ and the smaller domains for 4 km WRF and
CMAQ).

## 191 **2.2.1.1. Input Data**

192 The NARR data used for WRF simulations are downloadable from

193 <u>http://rda.ucar.edu/datasets/ds608.0/</u>. The data were based on an Eta 221 grid at 29 pressure

194 levels. Its horizontal resolution was 32-km and the frequency was 3 hourly. The initial and

boundary conditions were generated from the NARR analysis by WRF Pre-Processing System

196 (WPS). An alternative to NARR was the Eta-NAM analysis data. However, the data temporal

197 frequency was lowered from 3-hourly to 6-hourly starting in 2013. Our tests showed that it was

198 not as good as the NARR dataset - likely because of lower temporal resolution.

199 **2.2.1.2. Physics and FDDA Options** 

Major physics options used in the model are listed in Table 1. Our past modeling experiences indicated that employing the Yonsei University (YSU) for the PBL scheme and the Kain-Fritsch (K-F) for the cumulous scheme gave the best results for the Houston area. The YSU scheme was also one of the two PBL schemes recommended by Cuchiara et al. (2014). The K-F scheme is "drier" than others and produces less number of "false" convectional thunderstorms. For grid nudging options, we generally followed the recommendations in the WRF User Guide. For example, the mass fields (temperature and moisture) were nudged only at layers above the PBL
while wind fields were adjusted at all layers including the surface layer.

## 208 2.2.1.3. Observation Nudging with MADIS and CAMS data in WRF

209 Additional observational data are required to implement obs-nudging and OA. To generate the 210 input files for the OBSGRID program, we processed the observation data using the approach of 211 Ngan et al. (2012) and Czader et al. (2013). Observational data came from the MADIS and 212 TCEQ CAMS. MADIS is a National Oceanic and Atmospheric Administration (NOAA) 213 program which collects, integrates, quality-controls and distributes observations from NOAA 214 and other organizations. Additional information is available online, https://madis.ncep.noaa.gov/. 215 The four MADIS datasets used for obs-nudging were NOAA Profiler Network (NPN), 216 Cooperative Agency Profilers (CAP), Meteorological Terminal Aviation Routine (METAR) 217 weather report and NOAA Radiosonde (RAOB). The METAR dataset was collected by mostly 218 first-order, METAR reporting, surface monitoring stations. NPN, RAOB and CAP were the most 219 commonly used upper air datasets.

The processed input observation data were fed into OBSGRID to update the domain analyses and generate additional surface analyses and text nudging files. Obs-nudging was performed by the main WRF program after obs-nudging namelist variables are properly set. The namelist for OBSGRID and relevant WRF section settings came largely from recommended values of the WRF User Guide and a previous study by Ngan et al. (2012).

225 Theoretically, obs-nudging updating at a higher frequency should enhance the model's 226 performance. A typical frequency of input analysis data is 3-hourly while the frequency for 227 observational data is hourly. The 3-hourly frequency of input analyses may be the reason for the 228 default 3-hour time interval in WRF's OBSGRID settings for generating the obs-nudging files. 229 Since there were few existing obs-nudging studies related to air quality and we were not aware of 230 any reference to the adoption of a 1-hour input frequency, we assumed that all previous studies 231 used the default 3-hour interval. As the WRF model allows the interval to be set to 1-hour or 232 smaller when corresponding observational data were available, we tested both 1-hour and 3-hour 233 scenarios. The results indicated that 1-hour obs-nudging had slightly better performance than the 234 3-hour one. As a result, this study adopted 1-hour temporal frequency for observation nudging.

235 The variables that were nudged were temperature, moisture, and the two wind components

- 236 (zonal U-WIND and meridional V-WIND). Obs-nudging for moisture was not performed in this
- study. This was based on our past experiences since performing moisture nudging sometimes
- 238 triggers excessive artificial thunderstorms which disrupted the model flow fields.

## 239 2.2.2. Emissions Processing

240 For anthropogenic sources we utilized the USEPA's National Emissions Inventory of 2008

- 241 (NEI2008). Motor vehicle emissions for this inventory were processed using the USEPA's
- 242 Motor Vehicle Emissions Simulator (MOVES) model (USEPA, 2015). The inventory was
- 243 processed using the Sparse Matrix Operator Kernel Emissions (SMOKE) model v3.1 (Houyoux
- et al., 2000) to obtain gridded emission rates and speciated for the Carbon Bond 05 (CB05)
- chemical mechanism for use in the CMAQ model. The biogenic emissions were modeled using
- the Biogenic Emissions Inventory System (BEIS) v3.14. Although NEI2008 might have
- 247 overestimated NO<sub>x</sub> emissions in Houston (e.g., Choi 2012; Czader et al. 2015), we used base
- 248 NEI2008 without adjustment because the adjustment of the NO<sub>x</sub> emission also has large
- uncertainty (Czader et al. 2015). Pan et al. (2015) showed that the CMAQ ozone performance
- using NEI2008 appears reasonable.

## 251 2.2.3. CMAQ Configurations

252 The USEPA's CMAQ (Byun and Schere 2006) version 5.0.1 was adopted for this study. Several 253 air quality studies focusing on the Houston area have used this model (e.g., Foley et al. 2010; 254 Czader et al. 2013, 2015; Choi 2014; Pan et al. 2015; Diao et al. 2016; Souri et al. 2016). The 255 model's horizontal domains were slightly smaller than its WRF counterpart in order to avoid the 256 discontinuity near the domain boundary. The domains are depicted in Figure 2 as green and 257 brown boxes, respectively. The chemical boundary conditions for all the species in the 4-km 258 domain were derived from 12-km domain air quality forecasting results, available online at 259 http://spock.geosc.uh.edu. The model used the same vertical structure as WRF. Additional model 260 configurations are listed in Table 2. Chemical processes were simulated with the CB05 chemical 261 mechanism with cloud/aqueous chemistry, active chlorine chemistry and an updated toluene 262 mechanism. For aerosol modeling, the fifth-generation CMAQ aerosol mechanism (AERO5)

263 which includes sea salt modeling was selected. The total number of included species is 132, with 264 70 reactive gas-phase, 49 aerosol and 13 non-reactive species.

#### 265 **3. Evaluation Metrics**

266 To assess model performance against observations, we computed a set of five statistics including 267 Pearson correlation, index of agreement (IOA, Willmott 1981), mean bias (MB), root mean 268 square error (RMSE), and Mean Absolute Error (MAE). This list is similar to one used by Li et 269 al (2008) for model performance evaluation. The goal is to have a comprehensive comparison 270 between model and observation time series. The set of five statistics was divided into three 271 groups:

272 1) Evaluation of the magnitude of model results vis-a-vis in-situ data, in measurement units

- 273 • Mean Bias (MB)
- 274 • Mean Absolute Error (MAE)
- 275 • Root Mean Square Error (RMSE)

276 2) Measuring how close the model values follow changes in the observations, unitless

277 Correlation

278 3) A composite performance index, Index of Agreement (IOA or d), unitless

279 IOA is considered a better performance index than correlation as it takes into account the

280 difference in the means and standard deviation. For example, when correlations are similar,

281 lower model biases would yield higher IOA values. Additionally for this study, the mean and the

282 standard deviation of model values and observations were included as a reference.

283

## 4. General Meteorological and Ozone Conditions in September 2013

284 The weather during September 2013 was relatively dry with mostly southerly, easterly or 285 southeasterly winds. For this study, date format is mm/dd unless stated otherwise. From 09/05 to 286 09/19, there was a lack of influence of strong synoptic weather systems. Shifting wind patterns 287 were observed during the period: light northeasterly in the early morning gradually turned 288 clockwise to southeasterly in the afternoon and evening hours. In this period, winds shifted from

southeast to near east and there were more clouds after 09/10. The only cold front arrived on the

- 290 early morning of 09/21. Figure 3 shows the regional average temperatures for the period and it
- can be seen that 09/21 had the lowest daily maximum temperature. The influences of the cold air
- intrusion lasted till early 09/21. Winds turned into southerly in the afternoon of 09/25 and
- warming continued in the next few days until 09/28.



294

Figure 3. Regional hourly temperature averaged over all available hourly CAMS observations,
two model cases also included for September of 2013.

297 Light rain events occurred on 09/02, 09/10, 09/16, 09/19 to 09/21 and 09/28 to 09/30. 09/20 and 298 09/21 events consisted of widespread light to medium showers. Besides the above-mentioned 299 dates, there were a few other days with sporadic drizzles. A majority of the days between 09/01 300 and 09/20 were either sunny or cloudy. The periods from 09/08 to 09/10 and 09/18 to 09/20 had 301 more clouds than other days. The period from 09/21 to 09/30 was influenced by a cold front 302 passage. The days between 09/22 and 09/24 were sunny and cold. The surface wind reversed 303 direction during midday of 09/25 and brought clouds back from 09/26 to 09/30. High ozone 304 events in SETX during fall are typically associated with the passage of a cold front (e.g., 305 Rappenglueck et al. 2008). The only ozone event with hourly surface ozone exceeding 120 ppb 306 (parts per billion) in September which occurred on 09/25 fell in this category.

307 Figure 4 plots the hourly regional averaged ozone. On most days, the in-situ averaged ozone

- 308 concentrations were below 70 ppb. Since the winds after dawn consistently pushed the
- 309 precursors from the industrial area to the southwest of the city, the wind pattern did not favor the

310 local ozone production. The daytime winds also contained a persistent easterly component which 311 moved the pollutants away from the Houston metropolitan area. In the first 10-day period, low 312 background ozone originating from the Gulf of Mexico contributed to the low-ozone days. With 313 overcast skies on 09/19 and 09/20, hourly high ozone values dipped below 30 ppb. The two 314 highest ozone days characterized by post-frontal ozone events were the 25<sup>th</sup> and the 26<sup>th</sup> of 315 September respectively.



Figure 4. The hourly regional averaged ozone for the two cases (No-OA and 1hr-OA) at the stations which include observation surface  $O_3$  over the 4km domain for September of 2013.

## **5. Results**

To evaluate the WRF simulation, we calculated statistics for surface temperature and winds in the 4-km domain. In addition, we plotted out the PBL height time-series for the one site we had observations. For CMAQ evaluation, we calculated the surface ozone statistics for the whole month. Also, we plotted vertical ozone profile and calculated biases for ozone aloft on 09/25.

324 **5.1.** Meteorology

325 **5.1.1. Temperature** 

326 The comparison of regional average hourly temperature for the simulation period is shown in

327 Figure 3. The regional observed averaged surface temperature was calculated by averaging the

328 hourly temperature from ~60 CAMS sites in the 4-km model domain. The base case temperature

was too high compared to the in-situ measurements. For example, the "No-OA" base case maximum temperature for the 21<sup>st</sup> was 30°C compared to 25°C for the in-situ data. The high biases in the base case are sharply reduced in the "1Hr-OA" sensitivity case and temperature matched better with the observations for several time periods, especially for 09/20 to 09/23. The statistics of hourly surface temperature are listed in Table 3. With higher IOA and lower mean biases (MB), the "1Hr-OA" case was clearly better than the base case "No-OA". The IOA of "1Hr-OA" was about 9% higher than the base case.

## 336 **5.1.2. Winds**

337 In ozone chemistry, winds affect the accumulation of precursors and hence the resulting ozone 338 production (e.g., Banta et al. 2005, 2011; Darby 2005). They are also responsible for dispersing 339 high ozone and bringing in background ozone. Prevailing summer time southerly to 340 southeasterly winds in the HGB region significantly lower the ozone concentrations in the 341 metropolitan area. Therefore, high ozone events usually occur when such wind patterns change. 342 Cold front intrusions coming as early as late August blow pollutants to the south. As a result, an 343 area of high ozone develops in the Gulf. Following cold fronts weakening and the weather 344 warming up, reversing winds can bring high ozone back to land. High ozone may also occur 345 during intra-day recirculation events when pollutants previously blown away from industrial 346 zone are brought back by reversing winds. Correctly simulating these recirculation events is 347 particularly important in predicting the high ozone event caused by post-front conditions. The 348 ozone event in the HSC area on 09/25 was likely due to a combination of local recirculation 349 caused by onset of the bay breeze and increased background ozone brought in by transport.

Due to the land-water thermal contrast and the different size of the Galveston Bay and the Gulf of Mexico, the western shore of the Galveston Bay often experiences a successive onset of bay breeze and sea breeze in the summer. The bay breeze is typically a weaker easterly while the sea breeze is a stronger southeasterly. The sea breeze usually arrives one to a few hours later after the bay breeze. The bay breeze and the subsequent sea breeze phenomena in Houston were described by Banta et al. (2005).

The statistics of zonal (U-WIND) and meridional (V-WIND) wind components are listed in Table 3. The purpose of choosing U and V over wind speed and direction is to avoid the

- anomalies in the wind direction statistics. For example, although wind direction of 5 and 355
  degrees are close, the statistics suggest that they are distinctively different.
- 360 For both U and V components of wind, "1Hr-OA" had higher correlation and IOA than "No-
- 361 OA". The model performance on U and V are similar, with the correlation in a range of 0.76 to
- 362 0.81 for all the cases. For comparison, the performance of the OA case ("M1") in Ngan et al.
- 363 (2012) is very close to that in this study, with a correlation of 0.75 for U and 0.82 for V. In terms
- of IOA, the OA case is higher by 5-6% in U and 10-11% in V over the base case. This can be
- 365 explained by the much reduced wind biases in the OA case.
- 366 The base case had consistently stronger winds, especially the southerly component, than the
- 367 observations. This was reflected in the mean bias "MB", as well as the model mean "M\_M".
- 368 Winds were reduced significantly after OA was performed. In fact, the high southerly bias in
- 369 "No-OA" turned slightly negative after OA. Winds originating from the Gulf were also stronger
- in the base case, which played a role in raising the ozone level comparing to the "1hr-OA"
- 371 sensitivity case. Figure 5 illustrated the slowing down of southerly winds after observation
- 372 nudging. As a result of nudging, the wind vectors matched the observations better.



Figure 5. Model and observed winds at 09/01\_00 CST: No-OA (top) and 1Hr-OA (bottom).

375 Model winds are blue arrows and the observations are orange arrows. Stronger southerly winds,

376 especially along coastal region, were reduced in the OA case.

## 377 **5.1.3. PBL height**

378 Atmospheric pollutants are largely confined in the PBL as most of the emissions sources are

379 close to the ground level. Hence the PBL height plays a critical role in mixing and spreading the

pollutants. Haman et al. (2014) studied the relationship between ozone level and PBL height at a

381 Houston CAMS site and found that nighttime and early morning PBL heights were consistently

lower on high ozone days than on low ozone days. Czader et al. (2013) pointed out that the

model underprediction of PBL during nighttime may have caused the CO overprediction at the same site. CO is a good proxy for understanding model transport since it has low reactivity and a relatively long lifetime in the troposphere (typically 2 months). Cuchiara el al. (2014) conducted four WRF/Chem sensitivity tests using different PBL schemes over southeast Texas. While no preferred PBL scheme was identified for WRF simulations, the YSU scheme performed better than others in terms of ozone prediction.

389 Haman et al. (2012) showed that the daily maximum PBL height at the University of Houston 390 site mentioned in section 2.1 reached its highest values of slightly over 2000 m in August. In 391 September, typical daily maximum PBL height was 1500 m at 15 CST while daily minimum was 392 just below 200 m between 00 CST and 06 CST. The comparison of observed and model PBL 393 height is shown at Figure 6. Our results indicated that the model tended to overpredict the daily 394 maximum PBL height; obs-nudging helped to reduce the overprediction. For the daily minimum 395 PBL height, "No-OA" case had slightly high biases while the OA case matched quite well with 396 in-situ height data. The observed minimum PBL height was lower than that reported by Haman 397 et al. (2012), likely due to the cloudy conditions prevailing in September 2013. There was no 398 apparent explanation on the reduced daytime PBL biases in the OA case than the base case, but it 399 is likely the results of improved winds and temperatures in PBL.



401 Figure 6. Planetary Boundary Layer (PBL) height time series at CAMS C695 for September
402 2013.

403 **5.1.4.** Cold Front Passage

- 404 The surface winds on 09/20 were overwhelmingly southerly in the region and reversed on 09/21
- 405 due to the arrival of a cold front. The hour-by-hour wind shifts for 11 sites in HGB on 21
- 406 September are plotted in Figure 7. The sites are sorted by latitude with the southernmost site,
- 407 Galveston C1034, located at the bottom row. There was only one site, Deer Park C35, showing
- 408 weak southerly at 00 CST while all the others had mostly weak northerly. Starting from 01 CST,
- 409 winds in the entire HGB area turned northerly to northeasterly and continued gaining strength in
- 410 the next few hours, indicating cold air had taken over the region.
- 411 Both cases performed reasonably well on 09/21 and the timing of wind shift was captured
- 412 accurately; although "No-OA" lagged behind by ~ 1 hr. The winds turned weak northerly at 00
- 413 CST for most sites but the "No-OA" case still showed the wind direction to be all southerly.
- 414 Besides the timing, the northeasterly winds in the "No-OA" case sometimes were too strong;
- 415 obs-nudging helped moderate the winds. The reduced V-wind bias in "1Hr-OA" was also evident
- 416 in the wind model-measurement statistics on 09/21.



417

Figure 7. Hourly model (blue) and CAMS (orange) winds at 11 sites on 09/21: No-OA (top) and
1hr-OA (bottom). The 1hr-OA case is better at 00 CST to 02 CST and 17 CST to 20 CST.



#### 421 **5.2.1. Regional Average Hourly Ozone**

422 Figure 4 plots the regional average hourly ozone, which was defined similarly to the average 423 temperature. Overall, observed ozone concentrations were low and the model did a reasonably 424 good job on capturing the timing of intra-day variations. However, both cases tended to 425 overpredict the daily highs and daily lows, especially in the first 8 days and between 09/15 and 09/21. An obvious departure is the  $25^{th}$  – both cases underpredicted the daily high. During the 426 427 model high bias period, the OA case usually did better in reaching the daily low although it 428 overpredicted the high a bit more than the base case. The night time biases were reduced likely 429 because the lower southerly winds in the OA case transported less ozone from the Gulf to the 430 land.

431 Our results suggested that the modeled ozone concentrations were likely higher in the Gulf than 432 those in the real world. During the periods 09/02 - 09/04 and 09/07 - 09/08, the incoming ozone 433 from the Gulf was markedly lower than "normal" values. Since the model ozone had fixed 434 boundary values, the model was unable to capture these daily ozone variations at the boundary. 435 The model showed the highest biases during the period of 09/19 - 09/20, which is likely due to 436 overcast skies and uncertainties in models cloud fields and high background ozone values. 437 Despite the overprediction, the biases in OA case are notably lower than the base "No-OA" case during the nights of 19<sup>th</sup> and 20<sup>th</sup>. A future study to upgrade the accuracy of cloud fraction using 438 439 remote sensing data (e.g., Moderate Resolution Imaging Spectroradiometer, or MODIS) should 440 be helpful in explaining the biases.

There were a few days with elevated ozone due to post-front meteorology conditions. The only exceedance happened on 09/25, which was likely caused by meteorological events in Houston and the Galveston Bay. The overall ozone on 09/26 was higher than normal after southerly winds transported the ozone back from the Gulf, raising ozone concentrations over the entire region. A more detailed analysis of model predictions on 09/25 will be presented in subsection of 5.2.3.

#### 446 **5.2.2. Performance Statistics**

The ozone statistics are listed in Table 4. Both cases had very similar correlation of 0.72 and
0.73. However, the mean biases in the OA case were lower than the base case by 3.2 ppb, which

helped raise the IOA from 0.78 to 0.83. The model standard deviation increased in the OA case

and matched better with that of the in-situ data. The IOA improvement of the "1Hr-OA" caseover the base case for ozone was slightly less as compared to that for temperature and winds.

## 452 **5.2.3.** High ozone episode after the passage of a front

In SETX, high ozone events during the fall season usually occurred after the passage of a cold
front (e.g., Rappenglück et al. 2008; Ngan and Byun 2011; Ngan et al. 2012; Haman et al. 2014).
Two factors may have contributed to the post-front ozone events: 1) Following a cold spell, light
winds and sunny skies create an ideal condition for ozone production and accumulation. 2) Wind
reversal may transport back the pollutants that were previously blown into the Gulf.

458 During the DISCOVER-AQ period, the two days with highest ozone concentrations were the 25<sup>th</sup> and 26<sup>th</sup> of September as indicated in Figure 4. The 1-hour maximum ozone on the 25<sup>th</sup> was 459 localized and higher by about 40 ppb than the 26<sup>th</sup>. In addition to heightened background ozone 460 on the 25<sup>th</sup>, the major contributor was the production resulting from favorable weather 461 462 conditions: sunny, overall light winds and shifting winds over the industrial area. The light 463 morning land breeze carried pollutants from the ship channel area to the Galveston Bay. As the 464 day warmed up, the bay breeze started to develop and carry pollutants back to the land. This 465 localized circulation was described by Banta et al. (2005). Ngan et al. (2012) reported the same 466 phenomenon in their Texas Air Quality Study-II 2006 study.

467 Figure 8 shows the ozone time series for the La Porte (C556 in Figure 1) site located in the HSC
468 area. The highest hourly ozone for September, 151 ppb occurred here at 13 CST on the 25<sup>th</sup>.
469 Ozone rose from 10 ppb to 150 ppb between 09-12 CST. Such a sharp increase in ozone was
470 likely the result of increased photochemical activity under favorable meteorological conditions in
471 an area with accumulated precursors. Figures 9 and 10 depict the wind and ozone concentrations
472 at 08 CST and 13 CST.

The wind plots of Figure 9 indicate that the observed winds in the HGB region at 08 CST were light northerly for sites located on the north side, while they were westerly in the middle and south. The base case winds were all northerly while the OA case had northwest winds for the north side and west winds for the middle and south. Hence, the model winds in OA case are more realistic than the winds in base case. The 09 CST winds were similar to those of 08 CST. As a result, the ozone statistics in Table 5 showed that the OA case had much better correlation and IOA than the base case during 08-09 CST. This example demonstrated the ability of obsnudging to correct erroneous winds. However, later events showed it may not always be able to
perform consistently.

482 The bay breeze started to develop at 10 CST near the C556 site. The early onset was likely to be 483 related to warming up on the previous afternoon on 09/24 as indicated in Figure 3. At 10 CST 484 most other sites to the west of HSC experienced light northwest winds while those at HSC 485 originated from the northeast. Combined with the easterly bay breeze, a convergence zone was 486 formed just below C556, where emissions from the HSC area stalled and accumulated. At 13 487 CST, the whole region had light winds and the bay breeze was well developed. The highest 488 ozone indeed appeared in C556 and its vicinity. The rapid increase of ozone concentration for 489 C556 between 09-13 CST is shown in Figure 8.



491 **Figure 8**. Ozone time series of La Porte (C556) between 09/24\_00 to 09/28\_00 CST of 2013.

492 It is important to note that both modeled cases missed the wind shifts in the HSC area, and the 493 resulting convergence zone near C556. This could explain the model's inability to recreate the 494 sharp ozone increase at C556. Figure 9 shows that the ozone concentrations around HSC area are 495 quite low (~10 ppb) at 08 CST. A further examination showed that while both model cases 496 missed the wind shift and convergence, the patterns were different. The base case winds differed 497 considerably from the observed ones for most of the morning: instead of a weak westerly, it had 498 stronger northwesterly to northerly. By 08 CST, winds were almost uniformly northerly in the 499 base case while they were weak west-northwesterly in the OA case (Figure 9). The oval in 500 Figure 9's top-left panel shows the mismatch of winds around C556 in the base case. As a result,

- 501 the NO<sub>x</sub> produced in the city was carried further to the southeast in the model in the base case.
- 502 Until 13 CST, base case winds did not shift directions by much. The OA case got the early hour
- 503 weak northwesterly right, but missed the bay breeze onset between 10 and 13 CST (oval in
- 504 Figure 10). The OA case could not reproduce the small-scale wind reversal near C556,
- 505 suggesting there is a limitation in the current WRF OA's capability in rectifying the winds when
- 506 poorly simulated. On the other hand, the OA case did improve the spatial ozone pattern, as the
- 507 high ozone area was closer to HSC after OA (Figure 10).



- 509 Figure 9. Zoom-in ozone concentrations (right) and wind plots (left) at 13 CST 25 September for
- 510 "No-OA" (top) and "1Hr-OA" (bottom). Ozone observation is in the small circle; wind
- 511 observation is indicated by an orange arrow. La Porte site C556 is labeled. The numerical range
- 512 of the right-side color scale is 0 to 200 ppb. Higher values than 200 ppb have the same color as
- 513 200 ppb.





516 Figure 10. Zoom-in ozone concentrations (right) and wind plots (left) at 13 CST 25 September 517 for "No-OA" (top) and "1Hr-OA" (bottom). Ozone observation is in the small circle; wind 518 observation is indicated by an orange arrow. Bay breeze is shown in the orange oval. The

numerical range of the right-side color scale is 0 to 200 ppb. Higher values than 200 ppb havethe same color as 200 ppb.

521 The ozone measurements from aircraft P3-B provided a more complete picture for the ozone 522 evolution on 09/25. During the day, the aircraft flew around the industrial area, Galveston Bay 523 and Galveston Island for about 9 hours. Figures 11 and 12 plot the ozone concentrations along 524 aircraft tracks at 08 and 13 CST. Surface layer ozone from the "No-OA" case is provided as 525 background for reference. At 08 CST, an ozone level of 60-80 ppb aloft was already observed at 526 three locations (three loops in Fig.11): Galveston Island, Smith Point and the inner city. Another 527 high of ~90 ppb could be seen above the HSC area. Ozonesonde observations over HGB showed 528 the aloft ozone concentrations were typically ~40-60 ppb (e. g., Li and Rappengluck 2014) at the 529 height level of a few hundred meters to 4 km. The higher-than normal ozone aloft suggested a 530 post-front ozone recirculation condition. Such high ozone aloft might raise surface ozone as a 531 growing PBL downwardly mixed the air aloft with near surface air. At 13 CST, high ozone over 532 100 ppb was observed at multiple locations. The highest aloft ozone of ~ 160 ppb occurred 533 southwest of Smith Point in the Galveston Bay. Such high increase in ozone concentrations was 534 likely the result of active photochemistry in the industrial zone and around Galveston Bay; 535 indicating a high level of precursor accumulation in the area.



537 Figure 11. Ozone along aircraft tracks at 08 CST of September 25th, overlaid upon model No-

- 538 OA surface ozone. The numerical range of the right-side color scale is 0 to 300 ppb. Higher
- 539 values than 300 ppb have the same color as 300 ppb.





Figure 12. Ozone along aircraft tracks at 09/25\_13 CST of September 25<sup>th</sup>, overlaid upon model
"No-OA" surface ozone. Plumes can be seen as dark purple circles in Galveston Bay. The
numerical range of the right-side color scale is 0 to 300 ppb. Higher values than 300 ppb have
the same color as 300 ppb.

Figure 13 shows hourly ozone vertical profiles from 08 CST to 16 CST on September 25<sup>th</sup>, with 545 546 ozone being displayed on the x-axis and height on the y-axis. The observed ozone was averaged 547 over multiple measurements in the same model cell, so that they could be compared to model 548 output. Next, both model and observed ozone values were averaged over all the grid cells in the 549 same model layer, such that one dot represents the average ozone of all the cells in the same 550 layer. The 08 and 09 CST profiles showed there was a high ozone layer with average ozone of 551  $\sim$ 65 ppb stretching from 450 m to 1200 m in height. In comparison, all model runs had lower 552 ozone in this layer. The model biases as shown in Figure 14 were about -10 ppb at 08 CST and 553 grew to -20 ppb at 09 CST. The large discrepancy between low surface ozone and ozone aloft 554 was unusual and may be explained by the arrival of a high ozone air mass from aloft. The 555 observed ozone rose continuously in the following hours; however simulated ozone stagnated 556 around 60 ppb from the surface up to 2000 m until 15 CST. At 16 CST, the ozone of the OA case

557 in the lowermost (0-1 km) layer rose 20 ppb over the previous hours yet the base case ozone 558 increased only a few ppb. Although different in magnitude, the aloft ozone had a few similar 559 features to the surface ozone. Firstly, the model missed the observed high ozone in the afternoon 560 by a large margin. For example, the base case underpredicted the 0-1 km level ozone by up to 50 561 ppb. The primary cause for the lower ozone production was likely the models wind fields as both 562 model and observations had a clear sky in the industrial area and the Galveston Bay. Secondly, 563 nudging clearly helped reducing the ozone biases aloft. In most plots of Figure 14, the OA case 564 had lower biases than the base case. The largest difference was at 16 CST when nudging reduced 565 biases from ~45 ppb to ~30 ppb in the 300 - 1000 m layer.

566 While it is easy to understand the improvements in temperature and winds after obs-nudging was 567 applied, it is more difficult to explain how other variables such as precipitation and clouds 568 reacted to obs-nudging. The indirect impact of these meteorological variables on ozone was 569 harder to assess. In our study, we did not evaluate clouds quantitatively as there were no 570 digitized cloud fraction data available for our modeling domains. A preliminary analysis on 571 convection showed that there were occasions in which the model missed the convection or 572 precipitation and there were other occasions in which the model created artificial convection. 573 The convection cells were usually visible as a "star-burst" from surface wind vector plots -574 arrows going out to different directions from a center. However, the mismatch in convection 575 appeared to be not a serious issue since only a few occurrences were observed in the month of 576 September.



578 **Figure 13**. Vertical ozone profiles from 09/25\_08 CST to 09/25\_16 CST of 2013 for two cases 579 of No-OA and 1Hr-OA compared with corresponding observations. There are 9 plots, each for 580 one hour, with date and hour as caption.





## **6.** Conclusions and Discussions

585 In this study, we performed two Weather Research and Forecasting (WRF) and Community 586 Multiscale Air Quality (CMAQ) model simulations to explore model sensitivity to observation 587 nudging. In evaluating meteorological and ozone conditions, we found that obs-nudging 588 improved the meteorology and ozone performance as shown in the index of agreement (IOA) of 589 temperature, winds, and ozone. While the base case winds were overall well simulated, obs-590 nudging significantly reduced the high wind biases (especially the meridional wind) shown in the 591 base case. For planetary boundary layer height, obs-nudging reduced high biases in both daily 592 maximum and daily minimum values. In the end, the combined changes in meteorology lowered 593 the ozone biases by about 3 ppb, a 35% reduction. There were short time periods (such as 594 between 07 and 09 CST on 09/25/2013) when the simulated base case model winds differed 595 significantly from observational data and obs-nudging significantly corrected the meteorological 596 simulation problems, leading to much better ozone simulation. However, model ozone biases are 597 also impacted by emissions and lateral boundary conditions.

The only high ozone episode in the simulation period was related to the cold front passage. The small-scale winds and high aloft ozone concentrations on 09/25 likely contributed to the ozone exceedance in the area. It is also possible that an unreported emission event played a role. Since the maximum surface ozone at La Porte was much higher than the morning-time ozone aloft, the active local ozone production was likely the dominant factor. Analyses of ozone aloft on 09/25 showed while there was high ozone aloft and large negative model biases, the OA case tended to have smaller biases, especially in late hours of the day.

605 Small-scale meteorological events such as wind transition and recirculation have been cited (e.g., 606 Banta et al. 2005; Darby 2005) for their contributions to high ozone events. The model capability 607 in reproducing these events is critical in simulating such high ozone episodes. The base case did 608 not recreate the 25 September small-scale events likely due to the complex winds and a lack of 609 local information which can be used to steer model state closer to reality. On the other hand, the 610 inability of the sensitivity case to replicate the local winds is likely a result of the shortcomings 611 of the nudging process pending further investigation. An ongoing study by the current authors 612 suggests that errors in the meteorological fields from the default grid nudging files are important 613 sources. Methods are being tested to improve the quality of grid nudging files. Early results 614 showed that the bay breeze which caused the wind reversal around La Porte was well captured

- 615 through improved grid nudging files. In addition, more observational data (e.g., more sites and
- 616 higher data frequency) and more testing on the combination of nudging settings should help
- 617 improve the obs-nudging performance. Also, the impact of obs-nudging on precipitation and
- 618 clouds should be further investigated to understand their effect on chemistry.

## 619 Acknowledgements

- 620 The authors thank Texas Air Research Center (TARC) for its support through grant number
- 621 413UHH0144A and Air Quality Research Program (AQRP) through 14-014, the DISCOVER-
- 622 AQ team for the aircraft data, Vanessa Caicedo for LIDAR data, and the TCEQ CAMS site team
- 623 for the in-situ ozone and meteorological data.

#### 624 **References**

- Banta, R. M., Senff, C. J., White, A. B., Trainer, M., McNider, R. T., Valente, R. J., Mayor, S.
- 626 D., Alvarez, R. J., Hardesty, R. M., and Parrish, D.: Daytime buildup and nighttime transport of
- urban ozone in the boundary layer during a stagnation episode, J. Geophys. Res.-Atmos., 103,
- 628 22519-22544, 1998.
- 629 Banta, R. M., Senff, C. J., Nielsen-Gammon, J., Darby, L. S., Ryerson, T. B., Alvarez, R. J.,
- 630 Sandberg, S. R., Williams, E. J., and Trainer, M.: A bad air day in Houston, B. Am. Meteorol.
- 631 Soc., 86, 657-669, doi:10.1175/bams-86-5-657, 2005.
- Banta, R. M., Senff, C. J., Alvarez, R. J., Langford, A. O., Parrish, D. D., Trainer, M. K., Darby,
- 633 L. S., Hardesty, R. M., Lambeth, B., Neuman, J. A., Angevine, W. M., Nielsen-Gammon, J.,
- 634 Sandberg, S. P., and White, A. B.: Dependence of daily peak O<sub>3</sub> concentrations near Houston,
- 635 Texas on environmental factors: Wind speed, temperature, and boundary-layer depth, Atmos.
- 636 Environ., 45, 162-173, doi:10.1016/j.atmosenv.2010.09.030, 2011.
- 637 Byun, D., and Schere, K. L.: Review of the governing equations, computational algorithms, and
- other components of the models-3 Community Multiscale Air Quality (CMAQ) modeling
- 639 system, Appl. Mech. Rev., 59, 51-77, doi:10.1115/1.2128636, 2006.
- 640 Byun, D., Ngan, F., Li, X., Lee, D., Kim, S..: "Analysis of Air Pollution Events in Summer 2006
- and Preparation of Model Input Data for the Assessment Study", Grant No. 582-5-64594- FY07-
- 642 02, Final Report: Evaluation of Retrospective MM5 and CMAQ Simulations of TexAQS-II
- 643 Period with CAMS Measurements, Texas Commission on Environmental Quality, February
- 644 2008, 25 pp
- 645 Cheng, F.Y., and Byun, D. : Application of high resolution land use and land cover data for
- atmospheric modeling in the Houston-Galveston metropolitan area, Part I: Meteorological
- 647 simulation results, Atmos. Env., 42, 7795-7811, doi:10.1016/j.atmosenv.2008.04.055, 2008.
- 648 Cheung, V. T., and Wang, T.: Observational study of ozone pollution at a rural site in the
- 649 Yangtze Delta of China, Atmos. Env., 35, 4947-4958, 2001.

- 650 Choi, Y.: The impact of satellite-adjusted NO<sub>x</sub> emissions on simulated NO<sub>x</sub> and O<sub>3</sub> discrepancies
- 651 in the urban and outflow areas of the Pacific and Lower Middle US, Atmos. Chem. Phys., 14,
- 652 675-690, doi:10.5194/acp-14-675-2014, 2014.
- 653 Choi, Y. and Souri, A.: Chemical condition and surface ozone in large cities of Texas during the
- last decade: observational evidence from OMI, CAMS, and Model Analysis, Remote Sensing of
- 655 Environ., 168:90-101, doi:10.1016/j.rse.2015.06.026, 2015
- 656 Choi, Y., Kim, H., Tong, D., and Lee, P.: Summertime weekly cycles of observed and modeled
- NO<sub>x</sub> and O<sub>3</sub> concentrations as a function of satellite-derived ozone production sensitivity and
- land use types over the Continental United States, Atmos. Chem. Phys., 12, 6291-6307,
- 659 doi:10.5194/acp-12-6291-2012, 2012
- 660 Cuchiara, G. C., Li, X., Carvalho, J., and Rappenglück, B.: Intercomparison of planetary
- boundary layer parameterization and its impacts on surface ozone concentration in the
- 662 WRF/chem model for a case study in Houston, Texas, Atmos. Environ., 175-185,
- 663 doi:10.1016/j.atmosenv.2014.07.013, 2014.
- 664 Czader, B. H., Li, X. S., and Rappenglueck, B.: CMAQ modeling and analysis of radicals,
- radical precursors, and chemical transformations, J. Geophys. Res.-Atmos., 118, 11376-11387,
  doi:10.1002/Jgrd.50807, 2013.
- 667 Czader, B.H., Choi, Y., Li, X., Alvarez, S., Lefer, B.: Impact of updated traffic emissions on
- 668 HONO mixing ratios simulated for urban site in Houston, Texas. Atmos. Chem. Phys., 15(3),
- 669 1253-1263, doi:10.5194/acp-15-1253-2015, 2015.
- 670 Darby, L. S.: Cluster analysis of surface winds in Houston, Texas, and the impact of wind
- 671 patterns on ozone, J. Appl. Meteorol., 44, 1788-1806, doi:10.1175/jam2320.1, 2005.
- Daum, P. H., Kleinman, L. I., Springston, S. R., Nunnermacker, L. J., Lee, Y. N., Weinstein-
- 673 Lloyd, J., Zheng, J., and Berkowitz, C. M.: Origin and properties of plumes of high ozone
- observed during the Texas 2000 Air Quality Study (TexAQS 2000), J. Geophys. Res.-Atmos.,
- 675 109, D17306, doi:10.1029/2003jd004311, 2004.

- 676 Deng, A., Stauffer, D., Gaudet, B., Dudhia, J., Hacker, J., Bruyere, C., Wu, W., Vandenberghe,
- 677 F., Liu, Y., Bourgeois, A.: Update on WRF-ARW End-to-end Multi-scale FDDA System. 10th
- 678 WRF Users' Workshop, Boulder, CO, NCAR, 2009
- Diao, L., Roy, A., Czader, B., Pan, S., Jeon, W., Souri, A. H., and Choi, Y.: Modeling the effect
- of relative humidity on nitrous acid formation in the Houston area, Atmospheric Environment,
- 681 131, 78-82, 2016
- 682 Foley, K. M., Roselle, S. J., Appel, K. W., Bhave, P. V., Pleim, J. E., Otte, T. L., Mathur, R.,
- 683 Sarwar, G., Young, J. O., Gilliam, R. C., Nolte, C. G., Kelly, J. T., Gilliland, A. B., and Bash, J.
- 684 O.: Incremental testing of the Community Multiscale Air Quality (CMAQ) modeling system
- 685 version 4.7, Geosc. Model Development, 3, 205-226, doi:10.5194/gmd-3-205-2010, 2010.
- 686 Gilliam, R. C., and Pleim, J. E.: Performance Assessment of New Land Surface and Planetary
- Boundary Layer Physics in the WRF-ARW, J. Appl. Meteorol. Climatol., 49, 760-774,
- 688 doi:10.1175/2009jamc2126.1, 2010.
- Haman, C.L., Lefer, B., Morris, G.A.: Seasonal Variability in the Diurnal Evolution of the
- Boundary Layer in a Near-Coastal Urban Environment. J. Atmos. Ocean Tech., 29, 697-710,2012.
- Haman, C.L., Couzo, E., Flynn, J.H., Vizuete, W., Heffron, B., Lefer, B.L.: Relationship
- 693 between boundary layer heights and growth rates with ground-level ozone in Houston, Texas. J
- 694 Geophys Res-Atmos., 119, 6230-6245, 2014.
- 695 Houyoux, M., Vukovich, J., Brandmeyer, J., 2000. Sparse Matrix Kernel Emissions Modeling
- 696 System: SMOKE User Manual. MCNC-North Carolina Supercomputing Center. Available at:
- 697 <u>https://cmascenter.org/smoke/.</u>
- Huang, X.-Y., Xiao, Q., Barker, D. M., Zhang, X., Michalakes, J., Huang, W., Henderson, T.,
- Bray, J., Chen, Y., and Ma, Z.: Four-dimensional variational data assimilation for WRF:
- Formulation and preliminary results, Mon. Weather Rev., 137, 299-314, 2009.

- 701 Kleinman, L. I., Daum, P. H., Lee, Y. N., Nunnermacker, L. J., Springston, S. R., Weinstein-
- 702 Lloyd, J., and Rudolph, J.: Ozone production efficiency in an urban area, J Geophys Res.-Atmos,
- 703 107, 4733, doi:10.1029/2002jd002529, 2002.
- 704 Le Dimet, F.X. and Talagrand, O.: Variational algorithms for analysis and assimilation of
- meteorological observations: theoretical aspects, Tellus, 38A, 97–110, 1986.
- Li, X., and Rappenglück, B.: A WRF–CMAQ study on spring time vertical ozone structure in
- 707 Southeast Texas, Atmos. Environ., 97, 363-385, doi:10.1016/j.atmosenv.2014.08.036, 2014.
- Li, X., Lee, D., Kim, S.-T., Kim, H., Ngan, F., Cheng, F., and Byun, D.: Performance
- 709 Evaluation of a Year-long Run of an Air Quality Forecasting System for Southeast Texas,
- 710 10th Conference on Atmospheric Chemistry, New Orleans, January 2008, 2008.
- 711 Liu, Y., Bourgeois, A., Warner, T., Swerdlin, S., and Hacker, J.: An implementation of
- observation nudging-based FDDA into WRF for supporting ATEC test operations, 2005 WRF
- view of the second seco
- 714 Liu, Y., Bourgeois, A., Warner, T., Swerdlin, S., and Yu, W.: An update on "observation
- 715 nudging"-based FDDA for WRF-ARW: Verification using OSSE and performance of real-time
- 716 forecasts, 2006 WRF user workshop, Boulder, CO, 2006.
- 717 Ngan, F., Byun, D., Kim, H., Lee, D., Rappengluck, B., and Pour-Biazar, A.: Performance
- assessment of retrospective meteorological inputs for use in air quality modeling during TexAQS
- 719 2006, Atmos. Environ., 54, 86-96, doi:10.1016/j.atmosenv.2012.01.035, 2012.
- 720 Mason, R.; Strum, M.; Houyoux, M. Technical Support Document (TSD) Preparation of
- 721 Emissions Inventories for the Version 4, 2005-based Platform; U.S. Environmental Protection
- Agency, Office of Air and Radiation, Office of Air Quality Planning and Standards, Air Quality
- 723 Assessment Division, 2010
- 724 Olaguer, E. P., Rappengluck, B., Lefer, B., Stutz, J., Dibb, J., Griffin, R., Brune, W. H., Shauck,
- 725 M., Buhr, M., Jeffries, H., Vizuete, W., and Pinto, J. P.: Deciphering the Role of Radical
- 726 Precursors during the Second Texas Air Quality Study, J. Air & Waste Manag. Assoc., 59, 1258-
- 727 1277, doi:10.3155/1047-3289.59.11.1258, 2009.

- 728 Otte, T. L.: The impact of nudging in the meteorological model for retrospective air quality
- simulations. Part I: Evaluation against national observation networks, J. Applied Meteorol.
- 730 Climatol., 47, 1853-1867, doi:10.1175/2007jamc1790.1, 2008.
- Pan, S., Choi, Y., Roy, A., Li, X., Jeon, W., and Souri, A.: Modeling the uncertainty of several
- VOC and ints impact on simulated VOC and ozone in Houston, Texas, Atmos. Environ., 120,
- 733 404-416, 2015
- 734 Parrish, D. D., Allen, D. T., Bates, T. S., Estes, M., Fehsenfeld, F. C., Feingold, G., Ferrare, R.,
- 735 Hardesty, R. M., Meagher, J. F., Nielsen-Gammon, J. W., Pierce, R. B., Ryerson, T. B., Seinfeld,
- J. H., and Williams, E. J.: Overview of the Second Texas Air Quality Study (TexAQS II) and the
- 737 Gulf of Mexico Atmospheric Composition and Climate Study (GoMACCS), J. Geophys. Res.-
- 738 Atmos., 114, doi:10.1029/2009jd011842, 2009.
- 739 Pour-Biazar, A., McNider, R. T., Roselle, S. J., Suggs, R., Jedlovec, G., Byun, D. W., Kim, S.,
- Lin, C. J., Ho, T. C., Haines, S., Dornblaser, B., and Cameron, R.: Correcting photolysis rates on
- the basis of satellite observed clouds, J. Geophys. Res.-Atmos., 112, D10302,
- 742 doi:10.1029/2006jd007422, 2007.
- 743 Rappengluck, B., Perna, R., Zhong, S. Y., and Morris, G. A.: An analysis of the vertical structure
- of the atmosphere and the upper-level meteorology and their impact on surface ozone levels in
- 745 Houston, Texas, J. Geophys. Res.-Atmos., 113, doi:10.1029/2007jd009745, 2008.
- 746 Rappenglück, B., Lefer, B., Mellqvist, J., Czader, B., Golovko, J., Li, X., Alvarez, S., Haman,
- 747 C., and Johansson, J., 2011: University of Houston Study of Houston Atmospheric Radical
- Precursors (SHARP), Report to the Texas Commission on Environmental Quality, August 2011,
  145 pp
- 750 Souri, A. H., Choi, Y., Li, X., Kotsakis, A., and Jiang, X.: A 15-year climatology of wind pattern
- 751 impacts on surface ozone in Houston, Texas, Atmospheric Research,
- 752 doi:10.1016/j.atmosres.2016.02.007, 2016.
- 753 Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, M., Duda, K. G., Huang, Y.,
- Wang, W., and Powers, J. G.: A description of the Advanced Research WRF Version 3, 1-113,
- 755 2008.

- 756 Stauffer, D. R., and Seaman, N. L.: Use of 4-dimensional data assimilation in a limited-area
- mesoscale model .1. Experiments with synoptic-scale data, Mon. Weather Rev., 118, 1250-1277,
  1990.
- 759 Stauffer, D. R., and Seaman, N. L.: Multiscale 4-dimensional data assimilation, J. Appl.
- 760 Meteorol., 33, 416-434, 1994.
- Tucker, S. C., R. M. Banta, A. O. Langford, C. J. Senff, W. A. Brewer, EJ. Williams, B. M.
- 762 Lerner, H. D. Osthoff, and R. M. Hardesty. : Relationships of coastal nocturnal boundary layer
- vinds and turbulence to Houston ozone concentrations during TexAQS 2006. Journal of
- 764 Geophysical Research: Atmospheres, 115, no. D10 (2010).
- 765 Willmott, C. J.: On the Validation of Models, Physical Geography, 2, 184-194,
- 766 doi:10.1080/02723646.1981.10642213, 1981.
- 767 Zhong, S. Y., In, H. J., and Clements, C.: Impact of turbulence, land surface, and radiation
- parameterizations on simulated boundary layer properties in a coastal environment, J. Geophys.
- 769 Res.-Atmos., 112, doi:10.1029/2006jd008274, 2007.
- 770

- Table 1. Major WRF physics and FDDA options, the numbers in the parentheses are the related
- 773 settings in WRF namelist file.

WRF Version	V3.5.1
Microphysics	Lin et al Scheme (1983)
Long-wave Radiation	RRTMG (Rapid Radiative Transfer Model for General Circulation
	Models), Iacono et al. (2008)
Short-wave Radiation	New Goddard scheme, Chou and Suarez (1999)
Surface Layer Option	Monin-Obukhov with Carlson-Boland viscous sublayer scheme
	(1978)
Land-Surface Option	Unified Noah LSM, Chen and Dudhia (2001)
Urban Physics	None
Boundary Layer Scheme	YSU, Hong et al. (2006)
Cumulus Cloud Option	Kain-Fritsch (2004)
FDDA	Grid nudging on for all; Observation-nudging on for the OA case

- Table 2. Major CMAQ options, the text in the parentheses are the related settings in CMAQ
- build script.

CMAQ version	V5.0.1
Chemical Mechanism	CB05 gas-phase mechanism with active chlorine chemistry,
	updated toluene mechanism, fifth-generation CMAQ aerosol
	mechanism with sea salt, aqueous/cloud chemistry
Lightning NOx emission	Included inline
Horizontal advection	YAMO (Yamartino)
Vertical advection	WRF omega formula
Horizontal	Multiscale
mixing/diffusion	
Vertical mixing/diffusion	Asymmetric Convective Model (ACM) version 2
Chemistry solver	EBI (Euler Backward Iterative)
Aerosol	AERO5 for sea salt and thermodynamics
Cloud Option	ACM cloud processor for AERO5
Boundary conditions	Default static profiles

777

778 Table 3 Statistics of surface T, U-wind and V-wind for three WRF simulations: N – data points;

779 Corr – Correlation; IOA – Index of Agreement; RMSE – Root Mean Square Error; MAE – Mean

780 Absolute Error; MB – Mean Bias; O – Observation; M - Model; O\_M – Observed Mean; M\_M –

781 Model Mean; SD – Standard Deviation; Units for RMSE/MAE/MB/O\_M/M\_M/O\_SD/M\_SD:

782 temperature, degree C; winds (U and V), m/s

783

784

Surface temperature T										
Case	Ν	Corr	IOA	RMSE	MAE	MB	O_M	M_M	O_SD	M_SD
No-OA	41058	0.83	0.89	2.0	1.5	0.9	27.4	28.3	3.1	2.8
1Hr-OA	41058	0.94	0.97	1.0	0.8	0.0	27.4	27.4	3.1	3.1
Surface U w	Surface U wind									
Case	Ν	Corr	IOA	RMSE	MAE	MB	O_M	M_M	O_SD	M_SD
No-OA	43246	0.76	0.84	1.4	1.1	-0.6	-1.3	-1.9	1.6	1.9
1Hr-OA	43246	0.81	0.89	1.0	0.8	-0.3	-1.3	-1.6	1.6	1.6
Surface V wind										
Case	N	Corr	IOA	RMSE	MAE	MB	O_M	M_M	O_SD	M_SD
No-OA	43246	0.76	0.8	2.1	1.7	1.2	0.4	1.7	2.0	2.6
1Hr-OA	43246	0.80	0.89	1.2	0.9	-0.1	0.4	0.4	2.0	2.0

786 Table 4 Statistics of ozone for CMAQ simulations, see table 3 for column header information

Case	Ν	Corr	IOA	RMSE	MAE	MB	O_M	M_M	O_SD	M_SD
No-OA	33308	0.72	0.78	14.9	12.3	9.3	24.4	33.7	16.5	14.1
1Hr-OA	33308	0.73	0.83	13.8	11.0	6.1	24.4	30.6	16.5	17.4

- Table 5 Statistics of ozone on 09/25/2013, all day and hour 0 to 13. Both correlation and index of
- agreement are unitless. The red numbers indicate the three hours (07 CST to 09 CST) when the
- 790 ozone in 1Hr-OA case is significantly better than the No-OA case due to much improved winds.

		No-	ЮA	1Hr	-0A
	N	Corr	IOA	Corr	IOA
Hr All	1150	0.79	0.86	0.81	0.88
0	48	0.04	0.30	0.40	0.46
1	43	0.20	0.24	0.36	0.30
2	48	0.14	0.25	0.35	0.35
3	48	0.19	0.30	0.32	0.35
4	48	0.27	0.36	0.31	0.35
5	47	0.24	0.36	0.28	0.37
6	47	0.33	0.38	0.35	0.37
7	48	0.06	0.39	0.29	0.47
8	48	0.09	0.43	0.53	0.63
9	47	0.05	0.41	0.55	0.74
10	47	-0.10	0.29	0.30	0.51
11	47	0.13	0.39	-0.07	0.36
12	49	0.09	0.38	0.25	0.40
13	49	-0.09	0.37	0.36	0.46