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Sensitivity of the WRF-Chem v4.4 ozone, formaldehyde, and precursor simulations to multiple bottom-up emission inventories over East Asia during the KORUS-AQ 2016 field campaign

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1 **Abstract**

2 In this study, the WRF-Chem v4.4 model was utilized to evaluate three bottom-up

3 emission inventories (EDGAR-HTAP v2, v3, and KORUS v5) using surface and

4 aircraft data in East Asia during the Korea-United States Air Quality (KORUS-AQ)

5 campaign period in 2016. All emission inventories were found to reproduce the diurnal

6 variations of O_3 and NO_2 as compared to the surface monitor data. However, the spatial

7 distributions of the daily maximum 8-hour average (MDA8) O_3 in the model do not

8 completely align with the observations. The model MDA8 O_3 had a negative (positive)

9 bias north (south) of 30°N over China. All simulations underestimated the observed CO

10 by 50-60% over China and South Korea. In the Seoul Metropolitan Area (SMA),

11 EDGAR-HTAP v2, v3, and KORUS v5 simulated the vertical shapes and diurnal

12 patterns of O_3 and other precursors effectively, but the model underestimated the

13 observed O_3, CO and HCHO concentrations. Notably, the model aromatic VOCs were

14 significantly underestimated with the three bottom-up emission inventories, although

15 the KORUS v5 shows improvements. The model isoprene estimations had a positive

16 bias relative to the observations, suggesting that the Model of Emissions of Gases and

17 Aerosols from Nature (MEGAN) version 2.04 overestimated isoprene emissions.

18 Additional model simulations were conducted by doubling CO and VOC emissions

19 over China and South Korea to investigate the causes of the model O_3 biases and the

20 effects of the long-range transport on the O_3 over South Korea. The doubled CO and

21 VOC emission simulations improved the model O_3 simulations for the local emission

22 dominant case, but led to the model O_3 overestimations for the transport dominant case,

23 which emphasizes the need for accurate representations of the local VOC emissions

24 over South Korea.

1. Introduction

Air pollutants not only harm human health but also affect radiative balance, resulting in climate change (Anenberg et al., 2018; Franklin et al., 2015; Lee et al., 2014; Manning and von Tiedemann, 1995; Rosenzweig et al., 2008; Wild et al., 2001). Anthropogenic activities are the primary source of air pollutant emissions, which have significant temporal and spatial variability. Chemical transport models (CTMs) use bottom-up emission data to simulate ambient concentrations of air pollutants. CTMs then process these emissions, tracking their impact through chemistry, transport, and loss through deposition (Zhong et al., 2016). Therefore, sensitivity evaluations of CTMs to anthropogenic emission data are an essential part of atmospheric modeling research.

Several bottom-up emission inventories are available for chemical modeling of Asia, including the Multi-resolution Emission Inventory for China (MEIC), Regional Emission inventory in Asia (REAS), and Emissions Database for Global Atmospheric Research-Hemispheric Transport of Air Pollution (EDGAR-HTAP). Since 2010, Tsinghua University has developed the high-resolution MEIC emission inventory for China and updated the data to the v1.3, providing anthropogenic emissions by sector and species from 2008 to 2017 (Zheng et al., 2018). REAS provides emission data in Asia from 1950 to 2015 (Kurokawa and Ohara, 2020). In Europe, EDGAR-HTAP has been developed and widely used for CTM simulations from global to regional scale (Kim et al., 2021; Sharma et al., 2017; Sicard et al., 2021). Recently, EDGAR-HTAP v3 has been published, covering 19 years from 2000 to 2018 compared to only two years (2008 and 2010) in the version 2 data (Crippa et al., 2023). Zhong et al. (2016) compared REAS with EDGAR in July, 2007 over China, while Saikawa et al. (2017) compared 5 emission inventories including REAS, EDGAR, MEIC in China, without

1 validation. As bottom-up emission inventories are continuously updated for recent years,

2 there is an ongoing need to evaluate new emissions data.

3 The Ministry of Environment (MOE) in South Korea and National Aeronautics and

4 Space Administration (NASA) in the U.S. conducted the Korea-United States Air

5 Quality (KORUS-AQ) campaign in May-June 2016. The campaign provided a variety

6 of data sets, including ground-based and airborne observations, useful for the validation

7 of model simulations. The KORUS emissions, developed by Konkuk University, were

8 used by many modeling teams to simulate the air pollutant concentrations during the

9 campaign period. In this study, we selected the EDGAR-HTAP versions 2, v3, and

10 KORUS version 5 emission data and used the Weather Research and Forecasting model

11 coupled with Chemistry (WRF-Chem) version 4.4 for intercomparison of the three

12 emissions data sets. O_3 and its major precursors were selected for model evaluation and

13 the model results were validated with surface observation data in China and South

14 Korea and aircraft data acquired over the South Korean peninsula and surrounding

15 waters.

16

17 **2. Data and Methods**

18 **2.1. WRF-Chem model configurations**

19 In this study, we utilized the WRF-Chem v4.4, which was developed by the National

20 Oceanic and Atmospheric Administration (NOAA) and National Center for

21 Atmospheric Research (NCAR), to simulate meteorological variables and chemical

22 species in the atmosphere (Grell et al., 2005). The WRF-Chem v4.4 includes N_2O_5

23 heterogeneous chemistry that consists of several chemical reactions related with $ClNO_2$

1 and N_2O_5 reactions, resulting in nitrate aerosol. The reactions are incorporated in

2 Secondary Organic Aerosol-Volatility Basis Set (SOA-VBS) with Regional

3 Atmospheric Chemistry Mechanism (RACM) chemistry option (chem = 108) in WRF-

4 Chem (Li et al., 2016).

5 We set 59 vertically customized eta (η) levels as vertical layers. The model's first

6 layer height is approximately 40 m above ground level for the entire domain. The

7 model's vertical layers are designed to include about 17 layers under 1.5 km to simulate

8 planetary boundary layer chemistry and near surface vertical distribution in detail. The

9 horizontal resolution is 28 x 28 km^2. The simulations in this study start at 12 UTC on

10 April 24 and end at 12 UTC on June 11. The model meteorology restarts every 12 UTC

11 (9 PM local time in South Korea) to minimize numerical errors. After the first 7 days

12 of model initiation (spin-up), we analyzed the model results from May 1 to June 10. We

13 used China standard time (+8 UTC) and Korea standard time (+9 UTC) for evaluations

14 with observations. The model physics, chemistry, and aerosol schemes are summarized

15 in Table S1 with corresponding references. The Global Forecast System (GFS) Final

16 (FNL) analysis data are used for meteorological input and boundary conditions. The

17 Community Atmosphere Model with Chemistry (CAM-Chem) output is used for

18 chemical boundary conditions (https://www.acom.ucar.edu/cam-chem/cam-chem.html)

19 (Buchholz et al., 2019; Emmons et al., 2020). We used the Model of Emissions of Gases

20 and Aerosols from Nature (MEGAN) v2.04 to calculate biogenic emissions (Guenther

21 et al., 2006). We did not account for fire emissions because of small impact on air

22 quality simulations during the KORUS-AQ campaign period (Park et al., 2021).

1 **2.2. The model simulations using different anthropogenic emissions**

2 **2.2.1. Bottom-up emission data**

3 EDGAR-HTAP v2, v3, and KORUS v5 emissions are compared with respect to their

4 spatial distribution and total amount in Figure 1 and Table S3. We applied the same

5 diurnal factor for all three emissions data by species, following the diurnal patterns for

6 the Los Angeles Basin as in Kim et al. (2016) (also see Figure S1).

7 EDGAR-HTAP v2 provides 2-dimensional emissions of CH_4, CO, SO_2, NOx (NO

8 + NO_2), non-methane volatile organic compound (NMVOC), NH_3, PM_{10}, $PM_{2.5}$, BC,

9 and OC in 2008 and 2010 with a horizontal resolution of 0.1˚ x 0.1˚. We used 2010 data

10 since it is the most recent data available. The data are partitioned by each sector and its

11 sources such as air, ships, energy, industry, transport, residential, and agriculture

12 (https://edgar.jrc.ec.europa.eu/dataset_htap_v2). For East Asia, it included data from

13 the Model Inter-Comparison Study for Asia (MICS-Asia) and REAS v2.1. In South

14 Korea, it adopted data from the Clean Air Policy Support System (CAPSS) (Janssens-

15 Maenhout et al., 2015), and the underlying emission data had an original horizontal

16 resolution of 0.25˚ x 0.25˚ over East Asia, which is resampled to 0.1˚ x 0.1˚ resolution

17 by raster resampling and aggregation. The specifically mapped EDGAR-HTAP v2 data

18 were obtained through the WRF-Chem site (https://www.acom.ucar.edu/wrf-

19 chem/download.shtml) in the *anthro_emiss* program with the Model for Ozone and

20 Related chemical Tracers (MOZART) species. The *anthro_emiss* program converts the

21 EDGAR-HTAP v2 data into 28 x 28 km^2 grid by the RACM chemical species. It

22 mapped the MOZART volatile organic compounds (VOC) species into the RACM

23 VOC species (See the detailed equations in Supporting Information, Table S4) (Li et al.,

24 2014; Emmons et al., 2010).

1   The EDGAR-HTAP v3 is extended to much longer time scale than the previous

2   version EDGAR-HTAP (v2). The EDGAR-HTAP v3 covers 2000 to 2018 with a more

3   detailed horizontal resolution (https://edgar.jrc.ec.europa.eu/dataset_htap_v3) (Crippa

4   et al. 2023). While EDGAR-HTAP v2 uses MICS-Asia, only the REAS data are used

5   in China and India in the EDGAR-HTAP v3. It adopts the CAPSS-Konkuk University

6   (CAPSS-KU) data for South Korea and emission data provided by the Japanese

7   government for Japan. We chose the data for 2016, according to the KORUS-AQ

8   campaign period. Because the original EDGAR-HTAP v3 data provide VOC as total

9   NMVOC with the unit of ton/month, we distributed the total NMVOC to MOZART

10   VOC species with the ratio of each VOC species to total NMVOC from EDGAR-HTAP

11   v2 in *anthro_emiss* program. Then, the assigned EDGAR-HTAP v3 data were again

12   converted to the RACM.

13   The KORUS v5 emission data represent 2016 in China and 2015 in other regions.

14   The Comprehensive Regional Emissions Inventory for Atmospheric Transport

15   Experiment (CREATE) v2.3 data from 2015 were used and the ship emissions from

16   CAPSS were added near the coastal region in South Korea (Jang et al., 2020; Woo et

17   al., 2012). The CREATE is originally developed by combining REAS, MEIC, Japan

18   Auto-Oil Program emission inventory (JATOP), and Korean Clear Air Policy Support

19   System (CAPSS). The NMVOC species from KORUS v5 were mapped following the

20   Statewide Air Pollution Research Center (SAPRC-99) mechanism, and we also

21   assigned the SAPRC-99 species to RACM (Carter, 2000) (Supporting information,

22   Table S4).

23   Figure 1 shows the spatial distribution of NO, CO, and toluene emissions in May

24   for each inventory. The NO_X emissions were assumed to be emitted as NO. The major

1   cities in China and South Korea had relatively high NO, CO, and TOL (toluene and less

2   aromatics defined in RACM, see Table S2) emissions, which are major precursors

3   affecting O_3 formation. We define three boxes representing Eastern China, South Korea,

4   and the Seoul metropolitan area (SMA) and calculated the emissions (see Table S3). In

5   South Korea including SMA, EDGAR-HTAP v3 had the largest NO_X emission among

6   the emission inventories. The KORUS v5 has lower NO_X emissions in Eastern China

7   by 46% and 39% compared to EDGAR-HTAP v2 and v3, respectively. The CO

8   emission was the lowest in EDGAR-HTAP v2 in South Korea, being 56% (69%) lower

9   than that in KORUS v5 (EDGAR-HTAP v3). KORUS v5 showed the highest CO

10   emissions in SMA though EDGAR-HTAP v3 showed more CO emissions in South

11   Korea. However, KORUS v5 had the smallest CO emissions in China, being 7% (9%)

12   lower than that in EDGAR-HTAP v2 (v3). The TOL emission from KORUS v5 is higher

13   than those from EDGAR-HTAP v2 (EDGAR-HTAP v3) by 176% (98%) in China. The

14   relative difference between KORUS v5 and EDGAR-HTAP v2 (EDGAR-HTAP v3) is

15   larger in South Korea by 263%. These discrepancies of VOC emissions may lead to a

16   change in the NOx/VOC-sensitive regime and O_3 production efficiency. The sensitivity

17   of O_3 formation to NOx emission has discrepancies by its regime, which will be further

18   discussed in section 3.2.

19

20   **2.2.2. The model experiments**

21   The model experiments are summarized in Table 1. The simulations using EDGAR-

22   HTAP v2, v3, and KORUS v5 emissions are named as EDV2, EDV3, and KOV5,

23   respectively. In this study, we found consistent underestimation of CO and VOC for all

1 emissions by -40% (± 2%) and -25% (± 1%) (HCHO) compared to DC-8 in South

2 Korea. This is in line with the results reported by Park et al. (2021), who found that

3 almost every model underestimated CO. Underestimation of CO in East Asia is a well-

4 known feature revealed by many studies. For example, Gaubert et al. (2020) mentioned

5 that CAM-Chem underestimates CO during the KORUS-AQ campaign period and

6 presented a CO compensation method utilizing data assimilation with CO observations.

7 Wada et al. (2012) pointed out that EDGAR v4.1 underestimates anthropogenic CO

8 emissions in China by 45% compared to observation-based estimations of CO

9 emissions. Moreover, underestimation of VOC is also found for all anthropogenic

10 emission inventories. Kwon et al. (2021) estimated top-down emissions of

11 anthropogenic VOCs utilizing Geostationary Trace gas and Aerosol Sensor

12 Optimization spectrometer (GeoTASO). They found that top-down VOC emissions

13 were up to 6.9 times higher than bottom-up emissions (KORUS v5). With all emission,

14 O_3 is underestimated at most ground-based observation sites in South Korea. Therefore,

15 we conducted two additional model simulations using EDGAR-HTAP v3 that shows

16 lowest bias of O_3 concentrations compared to DC-8 (-14.2 ppb) than EDGAR-HTAP

17 v2 (-16.9 ppb) and KORUS v5 (-18.1 ppb) over the SMA: one is with twice the

18 anthropogenic CO and VOC emissions in China (EDV3_Ch2) and the other simulation

19 uses double CO and VOC emissions in both China and South Korea (EDV3_ChKo2)

20 to investigate possible improvements in the simulated O_3 and CO from these emission

21 changes.

22

2.3. Observations

2.3.1. Meteorological data

The meteorological field that WRF-Chem reproduced is validated with the surface synoptic observation (SYNOP) data operated by the World Meteorological Organization (WMO) (http://www.meteomanz.com). Surface temperature, relative humidity, and surface wind speed are adopted for model validation. As the SYNOP data are provided every 3 or 6-hourly, we selected model data when the observation data are available. There were 271 sites in China-Taiwan-Hongkong and 48 sites in South Korea.

2.3.2. Ground-based observations

The surface observation network used in this study was obtained from Airkorea in South Korea and the China Ministry of Ecology and Environment (MEE) in China. The Airkorea observation network provides 1-hourly measurements of NO_2, SO_2, CO, O_3, PM_{10}, and $PM_{2.5}$ at suburban, background, roadside, city, and port sites (www.airkorea.or.kr). The concentrations of NO_2, CO, and O_3 are measured using the chemiluminescent, non-dispersive infrared, and ultraviolet photometric methods, respectively. The model data with 28 x 28 km^2 horizontal resolution were linearly interpolated to the 365 sites in South Korea, and we selected NO_2, O_3, and CO for model validation. The Chinese observations were provided by MEE through the website (beijingair.sinaapp.com). Surface NO_2 over China was measured using a molybdenum converter, which has the potential for positive biases due to other NO_2-related oxidation products (Dunlea et al., 2007). In South Korea, the positive biases exist regarding NO_2 surface observations, which could to overestimations of 28.9% at suburban sites in

1   spring (Jung et al., 2017). CO was measured using infrared absorption (Zhang and Cao.,

2   2015), and there were 1454 stations in China during the campaign period.

3   For validation of NO_2 and HCHO vertical column density, data from the Pandora

4   spectrometer were used, which the model reproduced with emission inventories at the

5   Olympic Park site (37.5232˚N, 127.126˚E). The HCHO data from Pandora is corrected

6   because of internal off-gasing to avoid positive biases (Spinei et al., 2021). At the same

7   observation site, surface NO_2 was also measured by a KENTEK NOx analyzer with

8   photolytic method, and surface O_3 was measured using the same instrument. Ground-

9   based HCHO was measured using Aerodyne QCL. We compared the observed diurnal

10   cycle of vertical column and surface concentrations of NO_2 and HCHO with the model

11   simulations utilizing EDV2, EDV3, and KOV5. We also used ground-based VOC data

12   from gas chromatography flame ionization detector (GC-FID) operated by the Seoul

13   Research Institute of Public Health and Environment (SIHE).

14

15   **2.3.3. Aircraft data**

16   The DC-8 research aircraft, operated by NASA, performed multiple flight

17   measurements with a variety of measuring instruments. We utilized 1 minute interval

18   merged data of O_3, NO_2, CO, HCHO, and VOC along the 20 flight paths (Figure 7).

19   The nearest WRF-Chem grid is selected and then temporally and vertically interpolated

20   to the aircraft data to fully utilize the observations. Atmospheric NO_2 and O_3

21   concentrations were measured using a 4-channel chemiluminescence instrument, with

22   an uncertainty of 100 pptv + 30% and 5 ppbv + 10%, respectively. CO concentrations

23   were observed using a diode laser spectrometer, with an uncertainty of 2% or 2 ppbv.

1 The Compact Atmospheric Multi-species Spectrometer (CAMS) was used to measure

2 HCHO concentration, with a possible 3% systematic error (Richter et al., 2015). We

3 also utilized data from the Whole Air Sampler (WAS) to analyze VOC species from

4 different emission inventories (Colman et al., 2001). In this study, we focused on DC-

5 8 observations below a height of 2 km to concentrate on planetary boundary layer (PBL)

6 chemistry. The observation height was determined by GPS altitude above ground level.

7

3. Results

9 **3.1. The model meteorology simulations**

10 The model temperature and relative humidity were compared with surface observations

11 in China and South Korea. The model-simulated temperature had a slight negative mean

12 bias of -0.91 ˚C (correlation coefficient R = 0.90) in China, with the largest negative

13 bias in southwestern China. In South Korea, the mean bias was -1.71 ˚C (R = 0.88). The

14 simulated relative humidity showed a negative bias of -20 to -10% in the North China

15 Plain (NCP) area and a positive bias of 10 to 20% in southwestern China. There was a

16 negative bias of relative humidity over the west coastal area and a positive bias of 10 to

17 20% at most observation stations in South Korea. The correlation coefficients between

18 the model relative humidity and observations were 0.85 and 0.76 for China and South

19 Korea, respectively. Overall, the comparisons showed decent model simulations of

20 meteorology. During the KORUS-AQ campaign period, WRF-Chem accurately

21 simulated the daytime PBL height from a laser ceilometer (CL-31, Vaisla Inc., Finland)

22 observed at Yonsei University in Seoul, South Korea (Lee et al., 2019). But, Travis et

23 al. (2022) has indicated the possibility of PBL height underestimations by CTM.

24 Furthermore, due to limitations of the instrument, the ceilometer has potential to

1   inadequately estimate nighttime PBL height. It is primarily attributed to the method

2   based on aerosol gradients (Jordan et al., 2020). Therefore, the interpretation of

3   simulated nighttime concentrations of air pollutants should be approached with caution.

4   More analysis of meteorological fields, including PBL height, can be found in the

5   Supporting Information (Table S5 and Figure S2-S3).

6

7   **3.2. Evaluations with routine surface chemical observational data**

8   The study compared simulated concentrations of O_3, NO_2, and CO with data from

9   routine surface observational networks (Table 2 and Figure 2-6). First, the diurnal

10   variations of the model O_3 using different emissions inventories were compared with

11   observations for each subregion (Table 2 and Figure 2). Overall, all emissions

12   successfully reproduced diurnal variations and absolute values of O_3 for most regions,

13   but there were notable discrepancies in several regions. In the North China Plain (NCP)

14   region, EDV2 led to a negative model O_3 bias (-12 ppb) with R=0.65, while EDV3 and

15   KOV5 simulated O_3 better with reduced biases and increased correlations (R=0.68-

16   0.71). Similarly, EDV2 had a negative O_3 bias (-17 ppb) with R=0.62 in the Yangtze

17   River Delta (YRD) area, but EDV3 and KOV5 much improved the simulations, which

18   was also observed in the Northeastern China (NEC) area. However, the model O_3

19   concentrations based on the three emission inventories were overestimated in the

20   Sichuan-Chongqing-Guizhou (SCG) and Southeastern China (SEC) area. In Pearl River

21   Delta (PRD), EDV2 showed the lowest bias (-0.3 ppb) compared to EDV3 and KOV5.

22   In the suburban area of Northern China (NOC), all emission inventories reasonably

23   simulated hourly O_3 concentrations. Averaged O_3 was well simulated in South Korea

24   (KOR) with low biases (-1 to 0.7 ppb), but a negative bias appears over the Seoul

1   metropolitan area (SMA) with all emissions (-5.5 to -3.5 ppb) (Table 2).

2   The study also analyzed the mean values of daily maximum 8-hour average (MDA8)

3   O_3 concentration at each site and their spatial distributions for the entire campaign

4   period (Figure 3). The spatial distributions of the model MDA8 O_3 were not well

5   correlated with those of the observations. But, notable disparities were observed in

6   simulating MDA8 O_3 when the different emissions were used. For the north and eastern

7   part of China including Beijing and Shanghai, large negative biases disappear when

8   using EDV3 and KOV5. KOV5 only shows a significant correlation with the surface

9   MDA8 O_3 observations (including 929 sites) than EDV2 and EDV3 in China (0.43

10   versus 0.01, 0.20). The correlations between the time series of the model MDA8 O_3 and

11   observations varied at each site, with about 40-60% of sites (depending on the emission

12   inventories) showing a correlation coefficient greater than 0.6 (see Supporting

13   Information, Figure S4), and the locations of these sites were scattered. The correlation

14   slightly improved with hourly O_3 concentrations instead of MDA8 O_3, with about 50-

15   60% of sites having a correlation coefficient greater than 0.6 (Supporting Information,

16   Figure S4). For this metric, high correlations occurred in pollution hot spots north of

17   30°N and the South Coast of China, in which the ratio of HCHO to NO_2 (FNR) was

18   much less than 1, suggesting VOC-limited/NOx-saturated chemical regime (Supporting

19   Information, Figure S5). The model MDA8 O_3 were underestimated for the pollution

20   hot spots with a low HCHO to NO_2 ratio located north of 30°N, suggesting a possibility

21   of model underestimations of anthropogenic VOC emissions causing model MDA8 O_3

22   biases at these sites. In contrast, the simulated MDA8 O_3 was generally overestimated

23   for sites south of 30°N in which HCHO concentrations were high (Supporting

24   Information, Figure S5). Zhang et al. (2020) reported that simulated biogenic isoprene

1   from MEGAN was overestimated compared to observation sites under 35˚N in China.

2   The EDV2 and EDV3 showed a positive NO_2 bias over the YRD, NCP, and PRD

3   regions, which include large cities in China (Table 2 and Figure 4-5). On the other hand,

4   EDV2 and EDV3 had low negative NO_2 biases in the NEC and NOC regions (Figure

5   4). All models demonstrated reasonable NO_2 model performance in the SCG region,

6   where MDA8 O_3 was overestimated (Figure 2 and 4). In the YRD region, there were

7   large positive NO_2 biases with EDV2, EDV3, and KOV5 (ranging from 6.4 to 22.7 ppb).

8   Liu et al. (2021) reported that YRD is in a VOC-limited regime when using EDV2 The

9   findings indicated that a reduction in NOx emissions led to an increase in O_3

10   concentrations, while a reduction in VOC emissions resulted in lower O3

11   concentrations. The lower bias of O_3 in YRD can be attributed to the combined

12   influence of higher anthropogenic NOx emissions and VOC originated from both

13   anthropogenic and biogenic sources (Figure S5). In contrast, KOV5 underestimated

14   NO_2 in the NCP region, while EDV2 and EDV3 did not. All emissions showed

15   significant discrepancies compared to NO_2 observations in the SEC area, with a low

16   correlation coefficient (0.19 to 0.26). EDV3 showed the lowest bias of -1.9 ppb (-0.8

17   ppb) compared to EDV2 and KOV5 in South Korea (SMA). The daily averaged NO_2

18   exhibited spatial distributions similar to MDA8 O_3 and CO (Figure 5). The slopes of

19   regression between the three model simulations and observations were 1.31, 1.03, and

20   0.8 for EDV2, EDV3, and KOV5, respectively, in China. The correlation coefficients

21   between the simulated NO_2 utilizing EDV2, EDV3, and KOV5 and surface data were

22   around 0.6 in China. EDV2, EDV3, and KOV5 demonstrated good correlations with

23   observations in South Korea (R = 0.69-0.74). Correlation coefficient (R) was the

24   highest with KOV5 in South Korea (R=0.74).

1   The simulated CO was averaged at each site and compared with observations

2   during the KORUS-AQ campaign period (Figure 6). The three model results showed

3   similar spatial distributions to observations, indicating higher CO concentrations in the

4   NCP, YRD, and PRD regions than their surrounding areas. However, all simulations

5   failed to reproduce the abundance of CO, indicating large negative biases throughout

6   the country. The bias was larger in South Korea than in China.

7

8   **3.3. Evaluations with the airborne and special surface chemical observations**
9   **during KORUS-AQ**

10

11   **3.3.1. The aircraft observations**

12   Figure 7 shows the flight paths flown by the DC-8 during the KORUS-AQ campaign

13   period. In Table 3, we compare the model results for O_3, NO_2, CO, HCHO, TOL, XYL,

14   ETE (Ethene or OL2), and ISO with the corresponding observed values for all flight

15   tracks under 2 km height in South Korea (Table 3). On average, the model

16   underestimated O_3 by 15-18 ppb, with EDV3 exhibiting the lowest O_3 bias (-15.1 ppb)

17   compared to EDV2 and KOV5 (-16.8 and -17.5 ppb, respectively). All emissions

18   showed positive biases for NO_2 (0.64 to 1.72 ppb), ETE (0.08 to 0.14 ppb), and ISO

19   (0.1 to 0.11 ppb). However, the model significantly underestimated CO, HCHO, TOL,

20   and XYL for all three emissions. Given the large spatial variability of air pollutants in

21   South Korea, we also sampled aircraft data from six regions (see Figure 7) and

22   compared the three model results with the aircraft observations under 2 km height

23   (Figure 8).

24   The flight tracks that surveyed large power plants and factories in the Chungnam

25   region on a daily basis are shown in Figure S6 in the Supporting Information. The

1 largest negative model O_3 bias was observed over the Chungnam region, with a

2 difference of 38-41 ppb. Emission estimation uncertainties can be significant over this

3 region, where there are large point sources such as coal-burning power plants and

4 petrochemical industries. The model NO_2 agreed with the aircraft observations in SMA,

5 but it tends to overestimate the measurements in the other areas. There were substantial

6 model overestimations of NO_2 with EDV3 over the Chungnam and Busan areas, while

7 KOV5 showed the most reasonable model NO_2 simulations. The model CO near the

8 surface was underestimated in the entire domain, resulting in high negative model CO

9 biases relative to the aircraft observations across the six regions (Figure 8). Additionally,

10 the model HCHO was underestimated by all emission inventories for all subregions,

11 with negative biases being evident in the SMA, Yellow Sea, and Chungnam regions.

12 Other model VOC species, such as TOL, XYL, ETE, and ISO, were also analyzed.

13 These VOC species are classified by their chemical structures and reactivities in the

14 RACM (Stockwell et al., 1997) (Table S2). For example, TOL includes toluene and

15 relatively less reactive aromatics, while XYL includes xylene and more reactive

16 aromatics. The WAS data from DC-8 were lumped into RACM (Supporting

17 Information Table S6, Lu et al., 2013) and were compared with aircraft observations.

18 When the model TOL or XYL was compared with the observed toluene and xylene, the

19 model using KOV5 reasonably reproduced the observed concentrations (light gray bars

20 in Figure 8). However, the model TOL (even using KOV5) underestimated the observed

21 lumped TOL for most of the regions except for Busan (bars including the dark gray part

22 in Figure 8). The model using KOV5 reasonably reproduced the observed xylene or

23 XYL, except for the Chungnam and Busan regions. The observed ethene (or ETE)

24 concentrations were low (< 0.5 ppb), except for the Chungnam region, where the

1   average of measurements was 2.1 ppb. The model ethene concentration was higher than

2   the observations for the SMA, Kyungbuk, and Busan regions, while it had a large

3   negative bias (-1.6 ~ -1.3 ppb) for the Chungnam region. Regarding isoprene (ISO), one

4   of the most important biogenic VOCs, the model values were larger than the

5   observations by a factor of 2. In summary, underestimated CO and aromatic VOCs are

6   the main features, along with underestimated ozone and HCHO. The largest

7   discrepancies occur over the Chungnam region, where large point sources are located

8   on the west coast of South Korea. The detailed statistics over the SMA and Chungnam

9   area can be obtained from the Supporting Information (Table S7-S8).

10   Figure 9 displays the vertical distributions of observed and simulated O_3 and related

11   species over SMA. The shapes of the simulated profile were in agreement with the

12   observations. Particularly, the model accurately reproduced the observed NO_2 profiles

13   though the surface NO_2 is underestimated by -4.2 to -0.8 ppb in SMA (Table 2 and

14   Figure 9b). The underestimation of simulated surface NO_2 is explained by the

15   overestimation of molybdenum converter method; surface concentrations of NO_2 from

16   molybdenum converter is larger than photolytic converter by 13.6% on average and 64%

17   at 4 pm (Figure 10). However, the simulated O_3 and HCHO had negative biases of 16.4

18   ppb and 0.73 ppb, respectively, persisting from the surface to 2 km. Additionally, the

19   simulated CO underestimated the observations by 40% throughout the vertical layer.

20   While the model TOL and XYL, utilizing KOV5, agreed well with the observations

21   below 1 km, the results using EDV2 and EDV3 substantially underestimated the

22   observations throughout the layer. On the other hand, the model simulated ETE and

23   ISO overestimated the observations below 1 km over SMA.

24

3.3.2. The ground-based observations

During the KORUS-AQ campaign, Pandora and surface measurements were co-located at the Olympic Park. Figure 10 compares the observed diurnal cycle of Pandora vertical columns and surface concentrations of NO_2 and HCHO with the model simulations. The photolytic converter was used to measure surface NO_2 to minimize positive bias from the molybdenum converter. All emissions reasonably simulated the diurnal patterns of vertical column and surface NO_2 and HCHO concentrations. The surface NO_2 peak appeared at 07 LT in the model and 08 LT in the observations, associated with the increase of traffic and the under-developed convective boundary layer. On the other hand, the Pandora NO_2 column amount increased from 06 LT to 12 LT and stayed at that value throughout the afternoon, indicating the increase of NO_x emissions from morning to afternoon. The model-simulated NO_2 columns agreed with those from Pandora in terms of absolute values and diurnal variations. The opposite patterns between surface and column NO_2 were also shown in Crawford et al. (2020). The simulated and observed HCHO show similar diurnal variations, but all three emissions underestimated both column and surface HCHO values by up to -8.5×10^{15} molecules·cm^{-2} (-46%) at 7 LT and -0.9 ppbv (-26%) at the surface on average. The underestimations of the model HCHO relative to the Pandora and surface observations are similar to findings from comparisons of the model results with the aircraft data (Figure 9). Therefore, the model VOC performance needs to be investigated at the Olympic Park.

The diurnal variations of the model O_3, CO, TOL, and XYL were evaluated against the surface observations at the Olympic Park acquired during the KORUS-AQ campaign (Figure 11). The diurnal pattern and hourly averaged mixing ratio of O_3 were

1   well simulated with the three emission inventories with slight model negative biases.

2   The observed CO was 2.7 times higher than the model on average. Considering the

3   diurnal profile of observed TOL and XYL, KOV5 reduced the model negative biases

4   from EDV2 and EDV3, but it still showed negative biases. The model TOL and XYL

5   showed peak concentrations at 08 LT, but the observation had a maximum value at 06

6   LT. The model biases of XYL (-3.7 to -0.6 ppb, -89 to -20%) were much larger than

7   those in TOL at the surface. Our study demonstrates that the improvement of VOC

8   emission/chemistry representations in the model is necessary for better simulations of

9   air quality over SMA and South Korea.

10

11 **3.4. The model performances for the Local and Transport Cases**

12 Previous studies have used meteorological conditions to classify synoptic patterns that

13 affect air pollutant concentrations (Park et al. 2021; Peterson et al. 2019). In contrast,

14 we defined the Transport and Local cases by comparing model results that used the

15 EDV3 base emission and the EDV3 zero-out-Chinese emission (see Figure 12). The

16 Local case comprises May 4, May 20, June 2, and June 3 (Supporting Information,

17 Figure S7), while the Transport case includes May 25, May 26, and May 31 (Supporting

18 Information, Figure S8). The Local (Transport) case in this study generally aligns with

19 the Stagnant and Blocking (Transport) cases in Peterson et al. (2019). The Local case

20 has a Chinese contribution to O_3 of under 11%, whereas the Transport case has a

21 Chinese contribution to O_3 of over 46%. EDV3 performed better in simulating O_3 for

22 the Transport case compared to EDV2 and KOV5, with a bias of only 2.7 ppb in

23 comparison with the DC-8 airborne observations. In contrast, for the Local case, all

24 emissions had a negative bias ranging from 15.5-18.2 ppb. See the Table S9 in

1   Supporting Information to obtain detailed information of model performances against

2   DC-8 measurements for different cases. Surface concentrations of O_3 at Olympic Park

3   also exhibited enhanced contributions from Chinese anthropogenic emissions for

4   Transport case (Figure S9). This section focuses on the model simulations using EDV3

5   and its modified versions, EDV3_Ch2 and EDV3_ChKo2 (doubling Chinese and South

6   Korean CO and VOC emissions).

7   Figure 13 illustrates the biases in the model O_3, CO, and HCHO using EDV3 and

8   its variants relative to DC-8 observations over SMA. The plot highlights differences in

9   biases for the Local and Transport cases. The model O_3 biases were negative, and the

10   absolute values of biases were larger in the Local case than in the Transport case (-20%

11   versus -6%). The model CO biases were also negative, and the absolute values of biases

12   were larger in the Transport case than in the Local case. The model HCHO biases were

13   negative and similar for the two cases, except for a larger discrepancy between model

14   and observation in the Local case than in the Transport case. Doubling Chinese CO and

15   VOC emissions (EDV3_Ch2) only slightly reduced biases in the Local case, whereas

16   doubling South Korean CO and VOC emissions, as well as Chinese CO and VOC

17   emissions (EDV3 ChKo2), were necessary to substantially reduce the model biases for

18   the Local case. For the Transport case, doubling Chinese CO and VOC emissions

19   reduced biases to almost zero for CO and HCHO, but the model O_3 was much

20   overestimated, with 14% positive biases (from an original bias of -6%). Further

21   increasing South Korean CO and VOC emissions led to overestimations of O_3 (20%)

22   and HCHO (33%). These sensitivity tests modifying EDV3 indicate that increases in

23   CO and VOC emissions over South Korea improve the model O_3, CO, and VOC

24   simulations. However, increasing Chinese VOC (and CO) emissions may overestimate

1   the model O_3 for the studied period.

2

3   **4. Summary and conclusions**

4   We conducted sensitivity tests using WRF-Chem with three different bottom-up

5   emission inventories (EDGAR-HTAP v2, v3, and KORUS v5) to investigate the

6   impacts of different emissions on the simulation of O_3 and precursors in East Asia. This

7   study is the first to use EDGAR-HTAP v3 with WRF-Chem v4.4 and extends the

8   validation domain to the whole of China during the KORUS-AQ campaign period. We

9   extensively validated these emission inventories using both ground and aircraft

10   observations in East Asia.

11   The three emission inventories accurately reproduced the diurnal profiles and

12   absolute values of surface O_3 for most subregions in China, except for the SCG and

13   SEC areas. However, discrepancies were observed in the model performance for the

14   MDA8 O_3 concentrations, with poor correlations observed over regions with high

15   HCHO concentrations (south of 30°N) and relatively low ratios of FNR (north of 30°N).

16   The emission inventories reasonably reproduced the spatial distribution of daily surface

17   NO_2 concentrations. However, we found that CO was considerably underestimated by

18   the emission inventories over both China and South Korea.

19   We evaluated the model simulations against vertical profile measurements of O_3,

20   NO_2, CO, HCHO, TOL, XYL, ETE, and ISO from the DC-8 aircraft, as well as surface

21   observations over South Korea. The simulated vertical shapes of O_3, NO_2, CO, HCHO,

22   TOL, XYL, ETE, and ISO agreed well with the DC-8 measurements in the SMA,

23   although negative biases were observed for O_3, CO, TOL, XYL, and HCHO, with the

1 largest discrepancy between the model results and observations in the Chungnam area.

2 When we compared the simulations with the surface in-situ measurements and

3 PANDORA observations at the Olympic Park in Seoul, the model accurately

4 reproduced the diurnal patterns of surface and vertical columns of NO_2 and HCHO.

5 However, we found that the model underestimated TOL and XYL. This underestimation

6 of TOL and XYL is one of the reasons why the model underestimates O_3 concentrations,

7 as VOCs contribute to NO to NO_2 conversions resulting in O_3 production via

8 photochemistry.

9 We also classified the flight tracks into two categories: Local and Transport cases.

10 We found that the negative bias of O_3 was much larger under the Local case than the

11 Transport case. When the increment of CO and VOC emissions in South Korea is taken

12 into account, the biases of O_3 are significantly reduced, indicating the need for local

13 emission adjustments to decrease O_3 bias in South Korea.

14 Our study revealed a consistent overestimation of isoprene over SMA. The

15 uncertainty of biogenic VOC emissions from MEGAN can affect the model O_3

16 performance. Therefore, to achieve more accurate simulations of O_3 in East Asia, it is

17 essential to explore precise representations of both anthropogenic and biogenic VOC

18 emissions.

19

20 **Code and data availability**

21 WRF-Chem source codes are distributed by NCAR (https://doi:10.5065/D6MK6B4K).

22 WRF-Chem v4.4 is available in the GitHub (wrf-model, 2022). The exact version of

23 WRF-Chem codes and configuration files are archived at

1   https://doi.org/10.5281/zenodo.8260026 (Kim et al., 2023). National Centers for

2   Environmental Prediction (NCEP) FNL data can be accessed from Research Data

3   Archive (RDA) (NCEP, 2019). The CAM-Chem data for boundary conditions is also

4   obtained from RDA (ACOM, 2019). The EDGAR-HTAP v2 data can be downloaded

5   in the website (https://edgar.jrc.ec.europa.eu/dataset_htap_v2). The EDGAR-HTAP v3

6   is archived on Zenodo (Crippa, 2023). The KORUS-AQ data are available in the

7   website   (https://www-air.larc.nasa.gov/cgi-bin/ArcView/korusaq)

8   (doi:10.5067/Suborbital/KORUSAQ/DATA01). The EDGAR-HTAP v2, v3, and

9   KORUS v5 data including emission processing programs are available at

10 https://doi.org/10.5281/zenodo.8260026 (Kim et al., 2023).

11

12 **Author contribution**

13 KMK conducted simulations, analysis and wrote the paper. SWK designed this study,

14 secured funding, performed analysis and wrote the paper. SS supported model set-up

15 and contributed to refining the paper. DRB measured VOC data from DC-8. SC

16 acquired ground-based in-situ VOC data at Olympic Park. JHC performed analysis and

17 wrote the paper. LE and GF assisted in setting up the model emissions and discussed

18 about the model performance. AF measured HCHO data from DC-8. JRH measured

19 Pandora data (NO_2, HCHO). JH retrieved PBL height and discussed about the results.

20 JJ acquired NO_2 data at Olympic Park with different methods. AJW acquired NO_2 and

21 O_3 data from DC-8. JHW and QZ provided emissions inventories and related

22 information. All authors reviewed the manuscript.

23

1 **Competing interests**

2 At least one of the (co-)authors is a member of the editorial board of Geoscientific

3 Model Development.

4

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16

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1 **Table List**

2

3 Table 1. The model experiments with different emissions.

4

5 Table 2. Comparison of the ground-based hourly O_3, NO_2, and CO observations with
6 the simulations utilizing EDGAR-HTAP v2 (EDV2) and v3 (EDV3) and KORUS v5
7 (KOV5) in each regional box (unit = ppb). N is the number of samples. R is correlation
8 coefficient.

9

10 Table 3. Comparison of aircraft-based 1-minuite-interval O_3, NO_2, CO, HCHO, TOL,
11 XYL, ETE, and ISO observations with EDV2, EDV3, and KOV5 for all flight cases
12 under 2 km height (unit = ppb). N is the number of samples. R is correlation coefficient.

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1 **Figure List**

2

3 Figure 1. The averaged spatial distribution map of the NO, CO, and TOL emissions
4 from EDGAR-HTAP v2, v3, and KORUS v5 in May.

5

6 Figure 2. Averaged O_3 concentrations from ground-based observations and model
7 simulations over the areas that distinguish urban (red box) and non-urban (green box)
8 region (central plot). Box-averaged diurnal cycle (solid lines) of O_3 and 1/4 of standard
9 deviations (filled area) from observations (black), EDV2 (green), EDV3 (blue), and
10 KOV5 (red) by local time are shown. The results are shown for Northern China (NOC,
11 38-42˚N/106-110˚E), Sichuan-Chongqing-Guizhou (SCG, 27-33˚N/103-109˚E), Pearl
12 River Delta (PRD, 21.5-24˚N/112-115.5˚E), Southeastern China (SEC, 24-28˚N/116-
13 120˚E), Yangtze River Delta (YRD, 30-33˚N/119-122˚E), South Korea (KOR, 34.5-
14 38˚N/126-130˚E), North China Plain (NCP, 34-41˚N/113-119˚E), and Northeastern
15 China (NEC, 43-47˚N/124-130˚E).

16

17 Figure 3. Comparison of (a) the campaign averaged ground-based maximum daily
18 average of 8-hour O_3 (MDA8 O_3) (unit: ppb) observations and WRF-Chem simulations
19 with (d) EDGAR-HTAP v2 (EDV2), (e) v3 (EDV3), (f) KORUS v5 (KOV5) and (g, h,
20 i) the differences between the observations and model results. The sub-regions are
21 presented with red (urban) and green (non-urban) boxes. The scatter plots comparing
22 averaged observations and the three-emission-based WRF-Chem simulations (green;
23 EDV2, blue; EDV3, red; KOV5) are shown in (b) and (c) for Eastern China and South
24 Korea, respectively. (a, d-e) Color-filled circles in (a), (d), (e), and (f) represent the
25 averaged MDA8 O_3 for the whole campaign period (1st May to 10th June).

26

27 Figure 4. The same as Figure 2 except NO_2.

28

29 Figure 5. The same as Figure 3 except daily NO_2 (unit: ppb).

30

31 Figure 6. The same as Figure 3 except daily CO (unit: ppm).

32

33 Figure 7. The DC-8 flight paths during the KORUS-AQ campaign period (black) and 6
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35 Kyungbuk; 5: Gwangju; 6: Busan) (red).

36

1   Figure 8. The mean (bars) and 1/4 of standard deviations (whiskers) of (a) O_3, (b) NO_2,
2   (c) CO, (d) HCHO, (e) TOL, (f) XYL, (g) ethene (ETE), and (h) isoprene (ISO) (unit =
3   ppb) from EDV2 (green), EDV3 (blue), and KOV5 (red) for each box are shown,
4   respectively. TOL and XYL are calculated based on Table S6 (Supporting Information).
5   The contribution of toluene to TOL and m/p-Xylene + o-Xylene to XYL is represented
6   with light grey bars (e, f). The sampling numbers are represented with magenta color
7   above the plots.

8

9   Figure 9. Vertically averaged (a) O_3, (b) NO_2, (c) CO, (d) HCHO, (e) TOL, (f) XYL, (g)
10   ETE, and (h) ISO from DC-8 (black), EDV2 (green), EDV3 (blue), and KOV5 (red) in
11   SMA under 2 km height above ground level. The 1/2 of standard deviations are
12   represented with black whiskers in each 200m layer. The sample number is presented
13   with magenta color on the right side of the plots.

14

15   Figure 10. The diurnal cycles of vertical columns and surface concentrations of (a) NO_2
16   and (b) HCHO from Pandora spectrometer (column), and ground-based instruments
17   (TEI 42i NOx analyzer and Aerodyne QCL) at the Olympic Park site (37.5232˚N,
18   127.126˚E). EDV2 (green), EDV3 (blue), and KOV5 (red) are compared with
19   observations. The WRF-Chem vertical column concentrations are produced by
20   summing all vertical layers.

21

22   Figure 11. Diurnal cycles of surface (a) O_3, (b) CO, (c) TOL, and (d) XYL at the
23   Olympic Park site. EDV2 (green), EDV3 (blue), and KOV5 (red) are compared with
24   the observations. 1/4 of standard deviations are represented with grey shades. The
25   average period is from the 11th May to the 10th June.

26

27   Figure 12. Averaged O_3 (bars) and 1/4 of standard deviations (whiskers) (unit: ppbv)
28   for the 20 DC8 flights (under 2 km height). The observations (grey) are compared with
29   the model results utilizing EDV2 (green), EDV3 (blue), and KOV5 (red). White hatch-
30   filled bars over blue bars are the contribution of Chinese emissions to O_3 concentrations
31   obtained from the default and sensitivity model runs with/without Chinese
32   anthropogenic emissions. The Local (5/4,20 and 6/2,3) and Transport (5/25,26,31) cases
33   are shaded with light blue and orange, respectively.

34

35   Figure 13. The biases in (a) the model O_3, (b) CO, and (c) HCHO concentrations (bars)
36   relative to the DC-8 observations under 2 km height over SMA (dark gray: EDV3, red:
37   EDV3 Ch2, blue: EDV3 ChKo2): (left panel) Local and (right panel) Transport case.
38   Fractional differences (%) are shown in the white boxes.

1   **Table 1.** The model experiments with different emissions

Experiments	Emissions
EDV2	EDGAR-HTAP v2
EDV3	EDGAR-HTAP v3
KOV5	KORUS v5
EDV3_Ch2	EDGAR-HTAP v3 with double CO, VOC emission in China
EDV3_ChKo2	EDGAR-HTAP v3 with double CO, VOC emission in China & South Korea

2

1 **Table 2.** Comparison of the ground-based hourly O_3, NO_2, and CO observations with
2 the simulations utilizing EDGAR-HTAP v2 (EDV2) and v3 (EDV3) and KORUS v5
3 (KOV5) in each regional box (unit = ppb). N is the number of samples. R is correlation
4 coefficient.

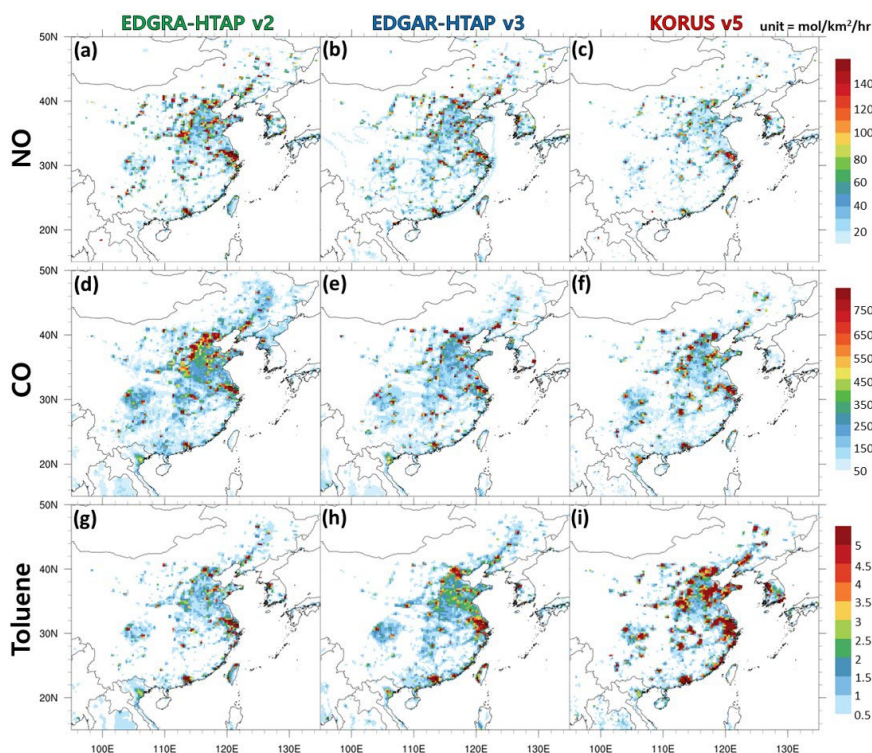|	Region		[1]NCP	[1,a]SCG	[1]YRD	[1]PRD	[1,b]KOR (SMA)	[2,c]NEC	[2,d]NOC	[2,e]SEC
	N		190	104	93	68	358 (125)	45	28	43
O_3	**OBS**	Mean	44.5	34.6	38.2	27.9	41.5 (36.6)	40.9	44.3	26.1
	EDV2	Mean	32.2	53.5	21.6	27.6	40.5 (31.1)	28.6	39.4	40.8
		Bias	-12.3	18.9	-16.6	-0.3	-1.0 (-5.5)	-12.3	-4.9	14.7
		R	0.65	0.53	0.62	0.61	0.59 (0.60)	0.48	0.63	0.52
	EDV3	Mean	43.4	57.5	35.7	34.7	41.0 (32.6)	35.2	43.7	45.5
		Bias	-1.1	23.0	-2.5	6.8	-0.5 (-4.0)	-5.7	-0.6	19.4
		R	0.68	0.55	0.66	0.65	0.56 (0.57)	0.63	0.67	0.55
	KOV5	Mean	49.0	55.3	41.1	35.7	42.2 (33.1)	37.1	43.8	42.4
		Bias	4.5	20.7	2.8	7.8	0.7 (-3.5)	-3.8	-0.5	16.3
		R	0.71	0.53	0.65	0.70	0.62 (0.64)	0.62	0.67	0.54
NO_2	**OBS**	Mean	17.5	13.8	17.1	12.9	23.2 (32.5)	13.5	11.9	9.6
	EDV2	Mean	25.8	12.7	39.8	22.0	18.8 (29.6)	13.7	12.9	11.0
		Bias	8.3	-1.0	22.7	9.1	-4.3 (-3.0)	0.2	1.0	1.5
		R	0.45	0.37	0.38	0.54	0.51 (0.34)	0.49	0.47	0.19
	EDV3	Mean	21.8	12.2	30.4	21.0	21.3 (31.8)	11.2	10.3	11.3
		Bias	4.3	-1.6	13.3	8.1	-1.9 (-0.8)	-2.3	-1.6	1.7
		R	0.44	0.34	0.36	0.52	0.49 (0.31)	0.49	0.52	0.22
	KOV5	Mean	13.9	7.5	23.5	13.3	17.7 (28.3)	7.0	7.7	7.7
		Bias	-3.6	-6.3	6.4	0.3	-5.5 (-4.2)	-6.5	-4.2	-1.9
		R	0.44	0.37	0.41	0.52	0.51 (0.39)	0.49	0.51	0.26
CO	**OBS**	Mean	835	597	694	636	443 (493)	527	579	655
	EDV2	Mean	373	389	455	282	175 (210)	206	162	258
		Bias	-462	-208	-239	-354	-267 (-283)	-321	-417	-397
		R	0.24	0.20	0.42	0.30	0.31 (0.30)	0.21	0.09	0.18
	EDV3	Mean	374	359	535	282	196 (208)	221	162	256
		Bias	-461	-238	-159	-354	-247 (-285)	-306	-417	-398
		R	0.22	0.19	0.35	0.31	0.26 (0.33)	0.24	0.10	0.25
	KOV5	Mean	355	358	475	305	190 (217)	231	176	266
		Bias	-480	-239	-219	-331	-253 (-276)	-296	-404	-388
		R	0.27	0.21	0.48	0.29	0.35 (0.36)	0.15	0.10	0.13

5 1) Urban area, 2) Non-urban area

6 a) Sichuan-Chongqing-Guizhou, b) South Korea, c) Northeastern China, d) Northern China, e) Southeastern China

1 **Table 3.** Comparison of aircraft-based 1-minuite-interval O_3, NO_2, CO, HCHO, TOL,
2 XYL, ETE, and ISO observations with EDV2, EDV3, and KOV5 for all flight cases
3 under 2 km height (unit = ppb). N is the number of samples. R is correlation coefficient.

Species	Type	N	Mean	Bias	σ	R
O_3	**OBS**		84.4		19.9	
	EDV2	5191	67.5	-16.8	16.7	0.44
	EDV3		69.3	-15.1	17.8	0.43
	KOV5		66.9	-17.5	15.8	0.50
NO_2	**OBS**		2.19		4.49	
	EDV2	5047	3.06	0.87	4.60	0.71
	EDV3		3.91	1.72	5.34	0.67
	KOV5		2.83	0.64	4.73	0.73
CO	**OBS**		253		100	
	EDV2	5575	148	-105	48	0.60
	EDV3		156	-97	47	0.59
	KOV5		146	-107	43	0.62
HCHO	**OBS**		2.37		1.64	
	EDV2	5365	1.75	-0.62	1.01	0.69
	EDV3		1.78	-0.59	1.02	0.67
	KOV5		1.80	-0.57	1.10	0.71
TOL	**OBS**		2.60		2.02	
	EDV2	730	0.47	-2.13	0.38	0.39
	EDV3		0.55	-2.05	0.48	0.38
	KOV5		1.58	-1.01	1.30	0.37
XYL	**OBS**		0.73		0.65	
	EDV2	289	0.23	-0.50	0.23	0.30
	EDV3		0.30	-0.43	0.31	0.30
	KOV5		0.49	-0.24	0.47	0.27
ETE	**OBS**		0.42		1.59	
	EDV2	2573	0.51	0.09	0.65	0.14
	EDV3		0.56	0.14	0.76	0.15
	KOV5		0.51	0.08	0.58	0.20
ISO	**OBS**		0.08		0.09	
	EDV2	1294	0.18	0.10	0.21	0.41
	EDV3		0.19	0.11	0.20	0.41
	KOV5		0.17	0.10	0.20	0.42

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Figure 1. The averaged spatial distribution map of the NO, CO, and TOL emissions from EDGAR-HTAP v2, v3, and KORUS v5 in May.

Figure 2. Averaged O_3 concentrations from ground-based observations and model simulations over the areas that distinguish urban (red box) and non-urban (green box) region (central plot). Box-averaged diurnal cycle (solid lines) of O_3 and 1/4 of standard deviations (filled area) from observations (black), EDV2 (green), EDV3 (blue), and KOV5 (red) by local time are shown. The results are shown for Northern China (NOC, 38-42˚N/106-110˚E), Sichuan-Chongqing-Guizhou (SCG, 27-33˚N/103-109˚E), Pearl River Delta (PRD, 21.5-24˚N/112-115.5˚E), Southeastern China (SEC, 24-28˚N/116-120˚E), Yangtze River Delta (YRD, 30-33˚N/119-122˚E), South Korea (KOR, 34.5-38˚N/126-130˚E), North China Plain (NCP, 34-41˚N/113-119˚E), and Northeastern China (NEC, 43-47˚N/124-130˚E).

Figure 3. Comparison of (a) the campaign averaged ground-based maximum daily average of 8-hour O_3 (MDA8 O_3) (unit: ppb) observations and WRF-Chem simulations with (d) EDGAR-HTAP v2 (EDV2), (e) v3 (EDV3), (f) KORUS v5 (KOV5) and (g, h, i) the differences between the observations and model results. The sub-regions are presented with red (urban) and green (non-urban) boxes. The scatter plots comparing averaged observations and the three-emission-based WRF-Chem simulations (green; EDV2, blue; EDV3, red; KOV5) are shown in (b) and (c) for Eastern China and South Korea, respectively. (a, d-e) Color-filled circles in (a), (d), (e), and (f) represent the averaged MDA8 O_3 for the whole campaign period (1st May to 10th June).

Figure 4. The same as **Figure 2** except NO2.

Figure 5. The same as **Figure 3** except daily NO₂ (unit: ppb).

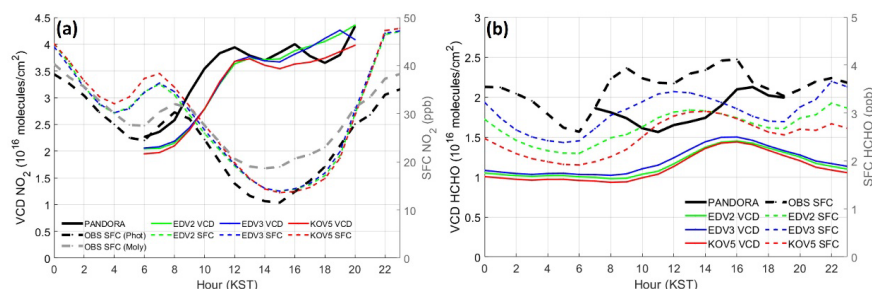**Figure 6.** The same as **Figure 3** except daily CO (unit: ppm).

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Figure 7. The DC-8 flight paths during the KORUS-AQ campaign period (black) and
6 regional boxes (1: Seoul Metropolitan Area (SMA); 2: Yellow Sea; 3: Chungnam; 4:
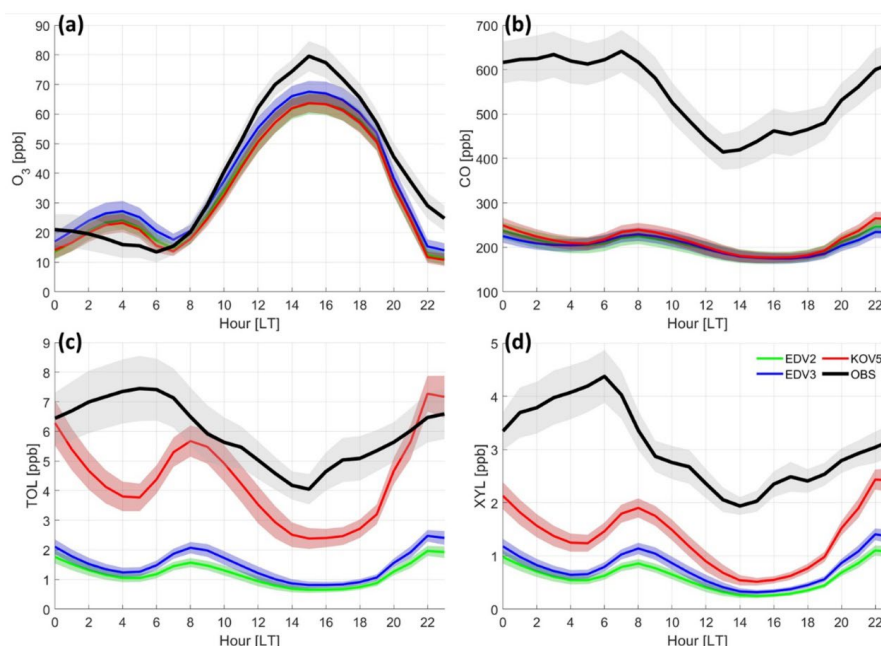Kyungbuk; 5: Gwangju; 6: Busan) (red).

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Figure 8. The mean (bars) and 1/4 of standard deviations (whiskers) of (a) O_3, (b) NO_2, (c) CO, (d) HCHO, (e) TOL, (f) XYL, (g) ethene (ETE), and (h) isoprene (ISO) (unit = ppb) from EDV2 (green), EDV3 (blue), and KOV5 (red) for each box are shown, respectively. TOL and XYL are calculated based on Table S6 (Supporting Information). The contribution of toluene to TOL and m/p-Xylene + o-Xylene to XYL is represented with light grey bars (e, f). The sampling numbers are represented with magenta color above the plots.
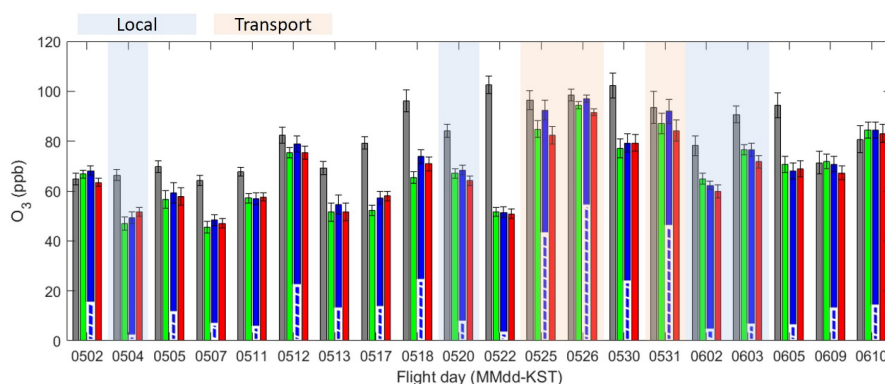
Figure 9. Vertically averaged (a) O$_3$, (b) NO$_2$, (c) CO, (d) HCHO, (e) TOL, (f) XYL, (g) ETE, and (h) ISO from DC-8 (black), EDV2 (green), EDV3 (blue), and KOV5 (red) in SMA under 2 km height above ground level. The 1/2 of standard deviations are represented with black whiskers in each 200m layer. The sample number is presented with magenta color on the right side of the plots.
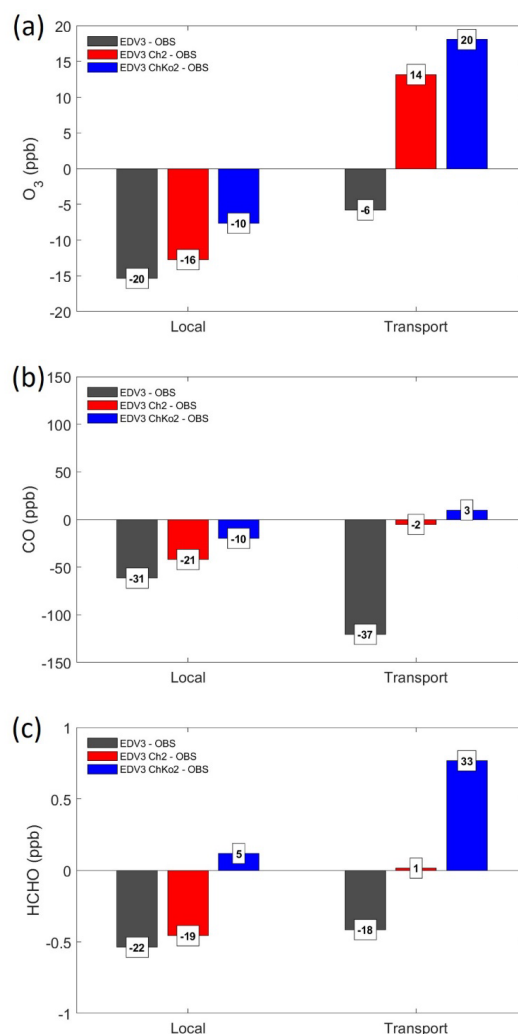
Figure 10. The diurnal cycles of vertical columns and surface concentrations of (a) NO_2 and (b) HCHO from Pandora spectrometer (column), and ground-based instruments (TEI 42i NO_x analyzer and Aerodyne QCL) at the Olympic Park site (37.5232˚N, 127.126˚E). Surface concentrations of NO_2 are obtained by the two methods: molybdenum converter and photolytic method. EDV2 (green), EDV3 (blue), and KOV5 (red) are compared with observations. The WRF-Chem vertical column concentrations are produced by summing all vertical layers.

Figure 11. Diurnal cycles of surface (a) O_3, (b) CO, (c) TOL, and (d) XYL at the Olympic Park site. EDV2 (green), EDV3 (blue), and KOV5 (red) are compared with the observations. 1/4 of standard deviations are represented with grey shades. The average period is from the 11th May to the 10th June.

Figure 12. Averaged O_3 (bars) and 1/4 of standard deviations (whiskers) (unit: ppbv) for the 20 DC8 flights (under 2 km height). The observations (grey) are compared with the model results utilizing EDV2 (green), EDV3 (blue), and KOV5 (red). White hatch-filled bars over blue bars are the contribution of Chinese emissions to O_3 concentrations obtained from the default and sensitivity model runs with/without Chinese anthropogenic emissions. The Local (5/4,20 and 6/2,3) and Transport (5/25,26,31) cases are shaded with light blue and orange, respectively.

Figure 13. The biases in (a) the model O_3, (b) CO, and (c) HCHO concentrations (bars) relative to the DC-8 observations under 2 km height over SMA (dark gray: EDV3, red: EDV3 Ch2, blue: EDV3 ChKo2): (left panel) Local and (right panel) Transport case. Fractional differences (%) are shown in the white boxes.