

Response to Reviewer 1's Comments

The reviewer's comments are written in blue and our responses are in black.

The authors ran WRF-Chem simulations over East Asia using three different emission inventories (EDGAR-HTAP v2, EDGAR-HTAP v3, and KORUS v5) and compared the model output to three sets of observations (routine monitoring data, airborne KORUS-AQ data, and ground-based KORUS-AQ data). They also ran sensitivity tests to doubling CO and VOC emissions and probed how the chemistry changed. Such comparisons are useful for model development, but I think there is opportunity for the paper to be strengthened in the following ways:

1. Currently, O₃ and NO₂ are treated separately in the comparison. I suggest adding a comparison of odd-oxygen (O_x = O₃ + NO₂) to probe whether the model issue is too much O₃ titration by NO or problems with the O₃ production regime.

→ We added analysis of O_x with surface observations in China and South Korea after P14 L24 (also see **Figure R1 and R2**). The diurnal patterns of O_x are well simulated with all emission inventories (**Figure R1**), showing similar issues that are previously discussed in section 3.2.

As the reviewer expected, underestimations in the model O₃ in YRD and NCP using EDV2 (light blue lines) disappeared when it was replaced by O_x, suggesting that the O₃ biases using EDV2 in the regions are caused by too much NO_x titration or inefficient O₃ formation in a NO_x-saturated regime. For other regions and cases using EDV3 and KOV5, O_x plots highlight biases in the model NO₂ levels. In YRD, O_x overestimations correspond to NO₂ overestimations in Figure 4. Meanwhile, in SCG and SEC, there are O_x overestimations caused by O₃ overestimations, suggesting a potential VOC emission overestimation.

Detailed descriptions, along with Figure R1 and R2, are included in the revised manuscript and Supporting Information.

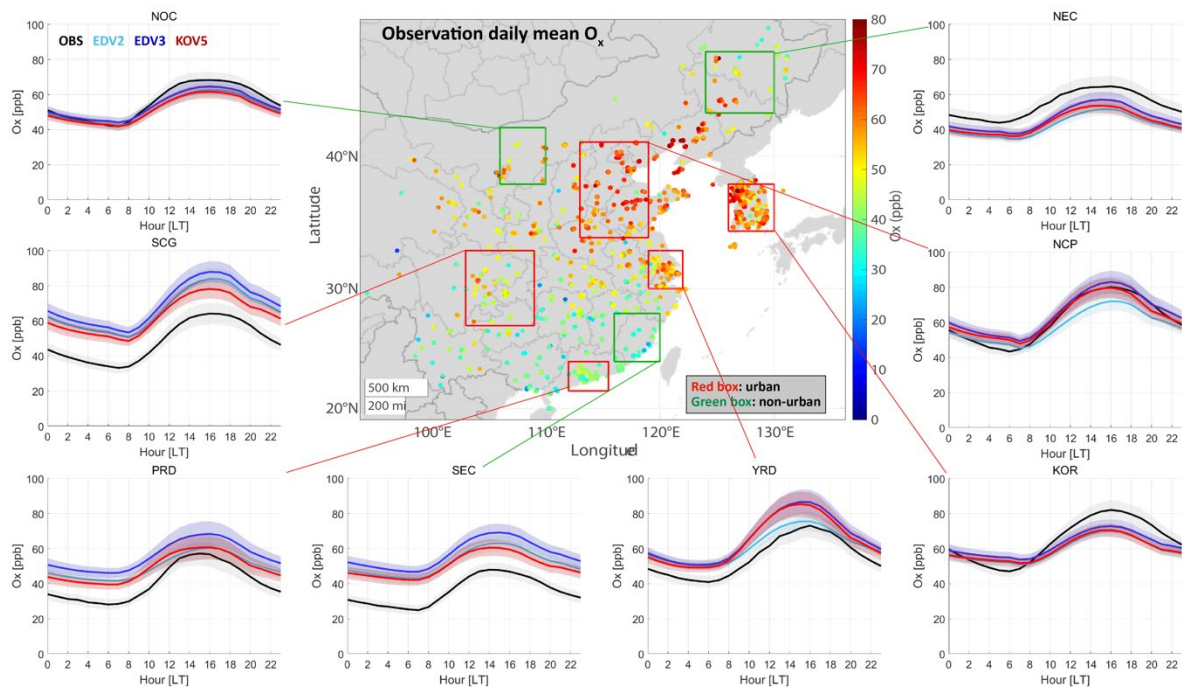


Figure R1. Averaged O_x concentrations from ground-based observations and model simulations over the areas that distinguish urban (red box) and non-urban (green box) region (central plot). Box-averaged diurnal cycle (solid lines) of O_x and 1/4 of standard deviations (filled area) from observations (black), EDV2 (sky blue), EDV3 (blue), and KOV5 (red) by local time are shown.

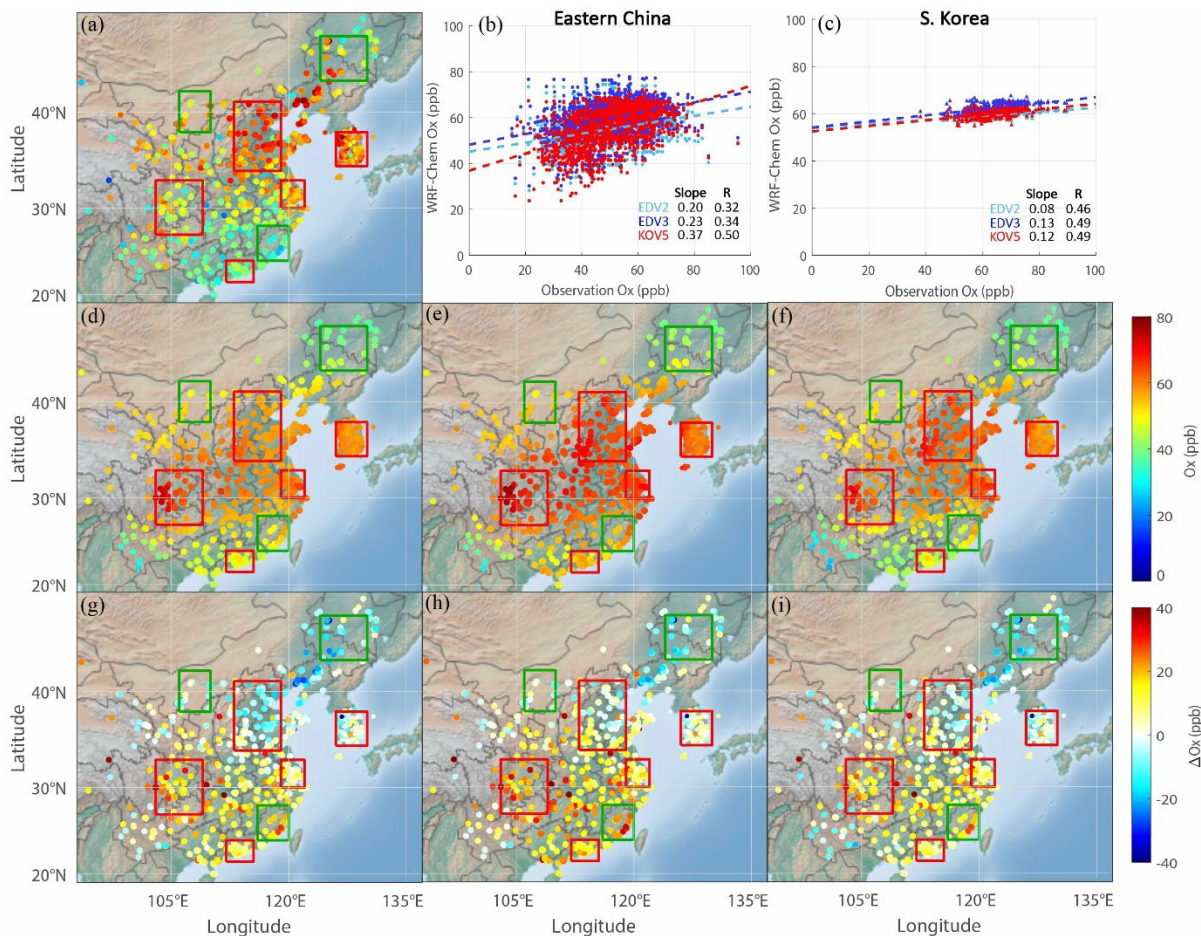


Figure R2. Comparison of (a) the campaign averaged ground-based maximum daily average of 8-hour Ox (MDA8 Ox) (unit: ppb) observations and WRF-Chem simulations with (d) EDGAR-HTAP v2 (EDV2), (e) v3 (EDV3), (f) KORUS v5 (KOV5) and (g, h, i) the differences between the observations and model results. The scatter plots comparing averaged observations and the three-emission-based WRF-Chem simulations (sky blue; EDV2, blue; EDV3, red; KOV5) are shown in (b) and (c) for Eastern China and South Korea, respectively.

2. On a related note, can you use the individual comparisons you've done of VOCs, NO_x, and O₃ in different regions with each inventory to draw some conclusions about how biases in either or both NO_x or VOC emissions affect O₃ predictions / chemical regimes in the different regions? There is a little bit of this on pages 13-14, but more organized conclusions about this (especially with your sensitivity tests to doubling the CO and VOC emissions) would be very useful for future model interpretation and emissions inventory development. For example, on page 12, can you discuss why the differences in VOCs and NO_x in each of the inventories cause them to simulate O₃ differently?

→ We provided **Table R1**, detailing NO_x, TOL, XYL, biogenic isoprene emissions, and formaldehyde-to-NO₂ ratio (FNR) for each region and emission inventory to enhance the understanding of regional differences. We included some discussions in section 3.2, such as the descriptions of VOC-limited regime in NCP with low FNR (< 1). The higher emissions of TOL and XYL in EDV3 and KOV5 resulted in higher O₃ concentrations with the smaller biases than EDV2 (**Table R1**). In SCG and SEC, biogenic emissions exceeded TOL and XYL by up to the factor of 10 with all emission inventories.

Table R1. Comparison of total NO_x, TOL, XYL, biogenic isoprene emissions in May, and formaldehyde-to-NO₂ ratio (FNR) for the KORUS-AQ campaign period for different emission datasets in each regional box. The MEGAN biogenic isoprene emissions are equally applied to all simulations using different emission data. (unit = mol/s for emissions)

Type	emissions	NCP	SCG	YRD	PRD	KOR(SMA)	NEC	NOC	SEC
NO _x emission	EDV2	5967	1500	2366	1178	990(196)	987	688	590
	EDV3	5202	1654	1642	1091	1191(214)	876	597	662
	KOV5	3237	902	1166	607	886(191)	513	373	410
TOL emission	EDV2	140	56	84	47	27(6)	26	8	20
	EDV3	220	77	99	68	27(8)	40	9	36
	KOV5	403	106	234	155	98(26)	68	21	79
XYL emission	EDV2	84	34	51	28	15(4)	15	4	12
	EDV3	132	46	60	41	16(4)	24	6	22
	KOV5	133	35	79	52	41(9)	21	7	26
Biogenic isoprene emission		132	364	43	127	135(6)	106	23	310
FNR (14-16LT)	EDV2	0.25	1.31	0.19	0.52	0.53(0.19)	0.68	0.76	1.18
	EDV3	0.44	1.30	0.32	0.52	0.43(0.18)	0.93	0.94	1.33
	KOV5	0.72	2.33	0.48	1.00	0.71(0.22)	1.44	1.49	1.91

Interpreting O₃ biases using FNR is cautioned due to the complex interplay of VOC and NO_x emissions and chemistry. Therefore, we added section 4 (discussion) with 4 additional sensitivity simulations (C5-C8 in **Figure R3**) as discussed in Kim et al. (2023), providing insights for O₃ bias correction in each region and city.

In SCG and SEC, the C5 case (50% anthropogenic VOC emission reduction only) exhibited the lowest O₃ biases, with a slight decrease in O₃ concentrations in the C4 case (50% NO_x reduction only), implying the need to reduce VOC emissions (biogenic and/or anthropogenic

emissions) (**Figure R3**). For the YRD and PRD, both NO_x and VOC emissions should be reduced based on C6 case (50% NO_x and VOC reduction), while C4 case (only NO_x 50% reduction) increased O₃ bias.

We also compared the sensitivity simulations with 12 mega cities in China and South Korea (**Figure R4**). VOC 50% reduction (C5 case) improved O₃ and NO₂ simulations in Chengdu and Chongqing. The lowest biases of O₃ and NO₂ were achieved with 50% NO_x and VOC reduction case (C6 case) for Shanghai, Nanjing, Guangzhou, Shenzhen, and Wuhan.

The detailed analysis, along with Figure R3 and R4, has been added to the revised manuscript.

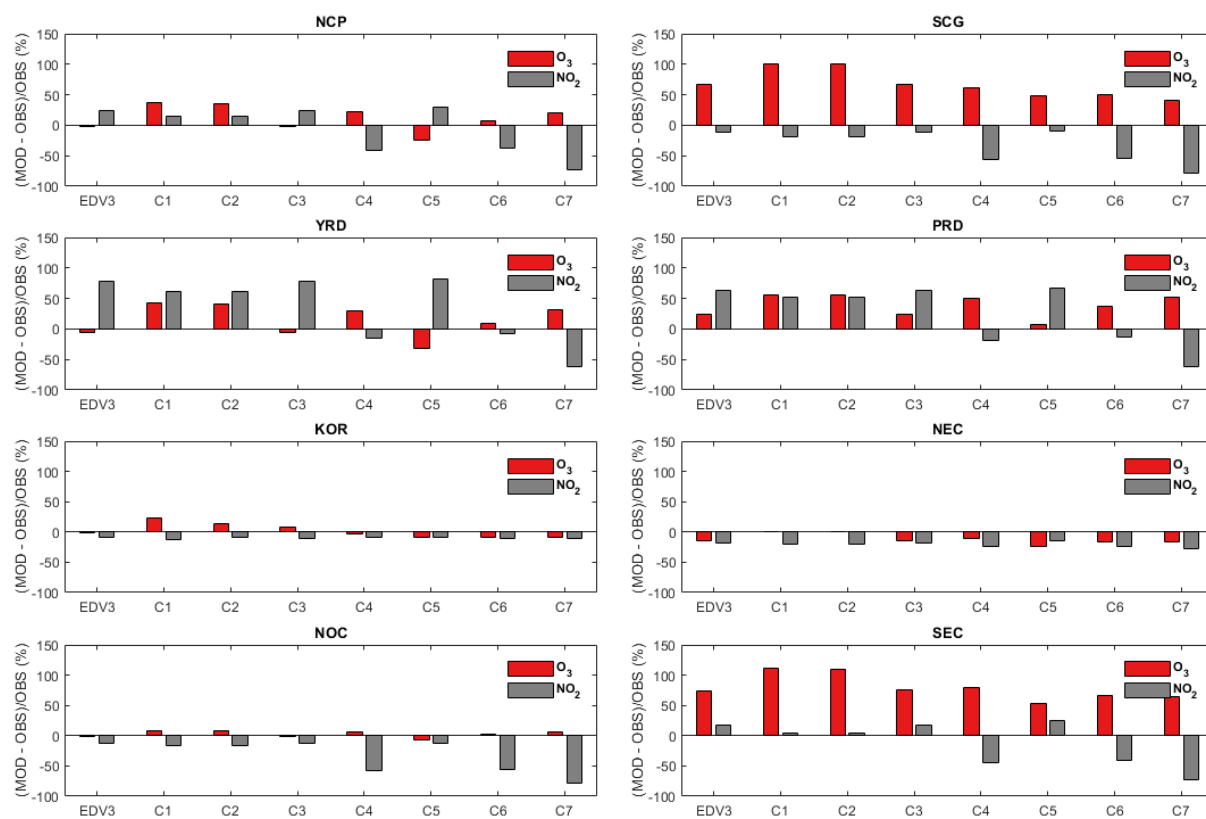


Figure R3. Comparison of relative biases ((Model-Observation)/Observation, unit=%) of daily O₃ and NO₂ at surface observation sites during the KORUS-AQ campaign period from sensitivity simulation (C1-7) with EDV3 in each region (NCP, SCG, YRD, PRD, KOR, NEC, NOC, and SEC). C1; EDGAR-HTAP v3 with double CO and VOC emission in China and South Korea, C2; EDGAR-HTAP v3 with double CO and VOC emission in China, C3; EDGAR-HTAP v3 with double CO and VOC emission in South Korea, C4; EDGAR-HTAP v3 with 50% NO_x reduction in China, C5; EDGAR-HTAP v3 with 50% VOC reduction in China, C6; EDGAR-HTAP v3 with 50% NO_x and VOC reduction in China, C7; EDGAR-HTAP v3 with 75% NO_x reduction in China.

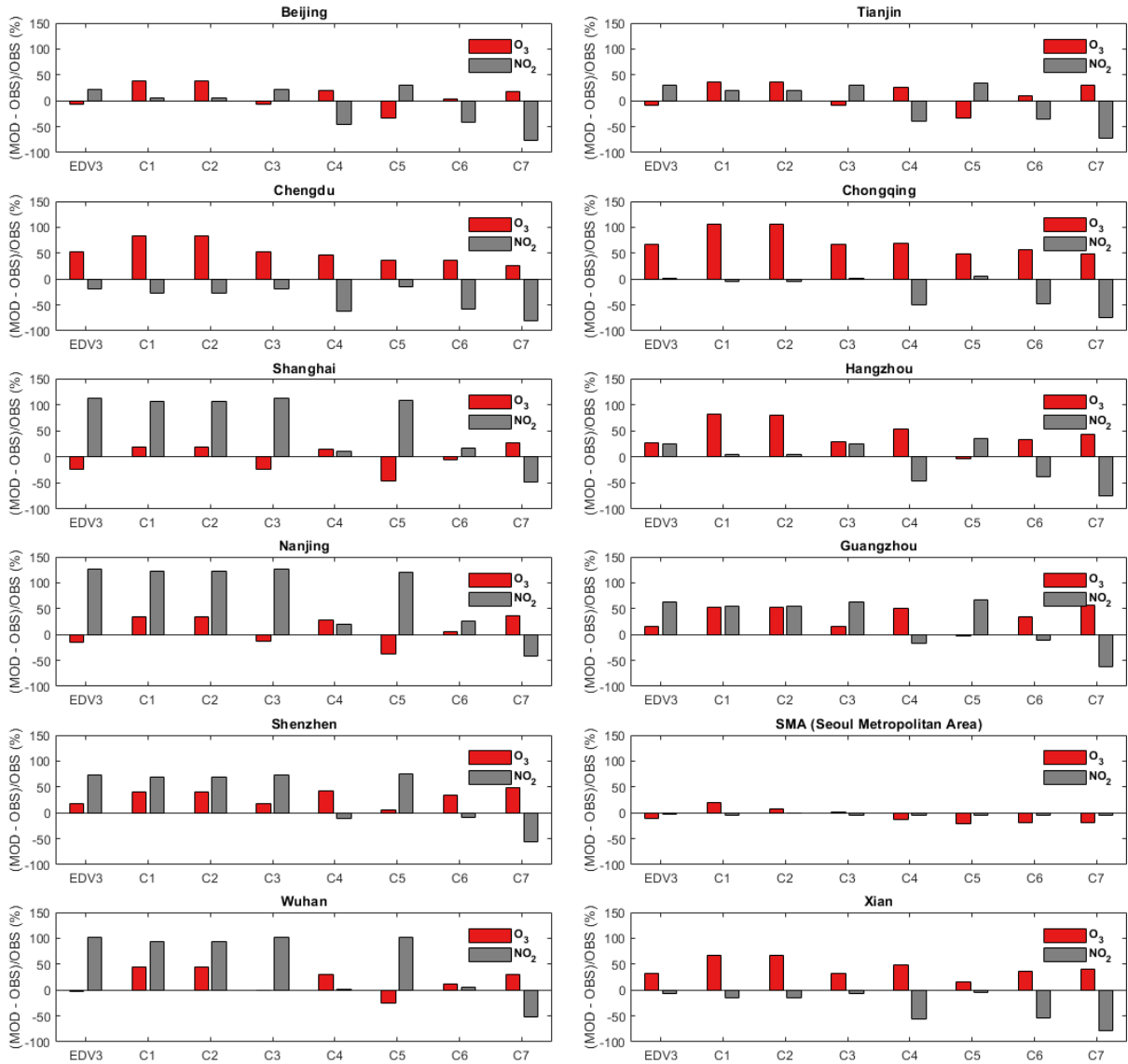


Figure R4. Same as **Figure R3** except that the region is changed to cities; Beijing (39.4-41.1N, 115.4-117.5E), Tianjin (38.55-40.25N, 116.7-118.1E), Chengdu (30.05-31.5N, 103-105E), Chongqing (28.15-32.25N, 105.3-110.2E), Shanghai (30.7-31.5N, 120.85-122E), Hangzhou (29.2-30.6N, 118.3-120.9E), Nanjing (31.2-32.65N, 118.35-119.25E), Guangzhou (22.55-24N, 112.9-114.05E), Shenzhen (22.4-22.9N, 113.7-114.65E), SMA (37.2-37.8N, 126.5-127.3E), Wuhan (29.95-31.4N, 113.65-115.1E), and Xian (33.65-34.75N, 107.65-109.9E).

3. Most of the conclusions in the manuscript are stated as “X is biased low with Y emissions inventory.” These statements would be more useful to the atmospheric chemistry community writ large if those statements were extended to say, “X is biased low with Y emissions inventory, which has Z implications for our understanding of emissions/chemistry.” For example, on page 20 line 12, can you add something to this sentence about the implications of having larger biases in the Transport case compared to the Local case? For a second example, on page 20, line 20, can you add something about the implications for NO_x emissions (based on O₃ being wrong but CO and HCHO being largely okay)? There are many other instances in the manuscript where this would be useful, but hopefully, the two examples I provided here are helpful illustrations.

→ To enhance discussions about causes of the model O₃ biases, we added a separate section of discussion about the chemical regimes in each region and city and the best way to reduce ozone biases accordingly, incorporating NO_x emissions information. Please refer to our response to Reviewer’s major comment 2. Furthermore, we added the sentences about analysis of Local and Transport case.

The excessive O₃ with double emissions in China is attributed to an overestimation of background O₃. We included Figure R5 to represent the overestimated O₃ from the downwind area (Yellow Sea) when CO and VOC emissions are doubled in China. This analysis is included in the revised manuscript. Furthermore, in section 4, causes of O₃ biases and directions to improvement for each region and cities are suggested in detail including Figure R3 and R4.

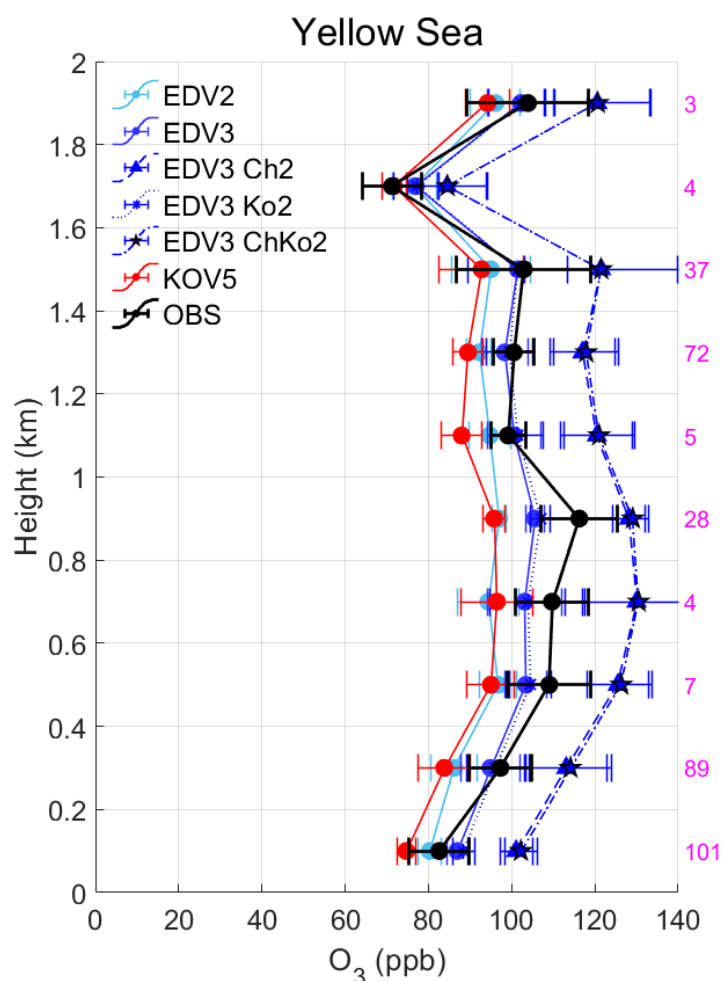


Figure R5. Vertically averaged O_3 from DC-8 (black), EDV2 (sky blue), EDV3 (blue), KOV5 (red), EDV3 with doubling Chinese CO and VOC emissions (dashed blue), EDV3 with doubling Korean CO and VOC emissions (dotted blue), and EDV3 with doubling Chinese and Korean CO and VOC emissions (dotted dashed blue) in Yellow Sea under 2 km height above ground level. The 1/2 of standard deviations are represented with whiskers in each 200m layer. The sample number is presented with magenta color on the right side of the plots.

4. The manuscript includes some contextualization of this work in the context of other emissions inventory comparisons (e.g., for CO on page 8). I think the paper would be strengthened by adding similar contextualization for the other comparisons (NO_x , O_3 , VOCs, etc.), especially given how many model-measurement comparisons have been done to date with KORUS-AQ (and related) data.

→ Our objective is to systematically identify and summarize potential issue associated with anthropogenic bottom-up emission inventories, investigating their potential impact on O_3

simulations in East Asia. We included relevant previous model studies in section 1 (Introduction) as explained below.

“Many modeling studies are done during this period including validations of CTM results with various observations. Miyazaki et al. (2019) adjusted emission inventories using various satellite data sets and Model for Interdisciplinary Research on Climate with chemistry (MIROC-Chem) resulting in O₃ simulations improvement. Goldberg et al. (2019) reported underestimations of NO_x emissions in South Korea including Seoul. Souri et al. (2020) also revealed the same issue in South Korea and analyzed sensitivity of O₃ formation to the NO_x and VOC emission adjustments derived from inverse modeling. Tang et al. (2019) revealed negative bias of simulated CO concentrations in East Asia by utilizing satellite data and the Community Atmosphere Model with Chemistry (CAM-Chem). Choi et al. (2022) modified anthropogenic VOC emissions using satellite HCHO observations and inverse modeling method with the Goddard Earth Observing System with Chemistry (GEOS-Chem), which reduced O₃ and HCHO biases.”

Specific comments:

Page 5, line 8: are the NMVOCs lumped or speciated?

→ It is lumped NMVOC. We added ‘total’ in front of ‘non-methane volatile organic compound’.

Page 5, line 9: do you apply any scale factors for using 2010 emissions data in a 2016 simulation?

→ We did not use scale factors.

Page 5, line 17: What does ‘specifically’ mean here?

→ We intended to describe that it is speciated NMVOCs from EDGAR-HTAP v2. We will change ‘specifically mapped EDGAR-HTAP v2 data’ to ‘speciated EDGAR-HTAP v2 VOC data’ to avoid confusion.

Page 7, lines 1-2: should read “...toluene and less *reactive* aromatics...”

→ Thank you. We added ‘reactive’ in front of ‘aromatics’ in the revised manuscript.

Page 7, line 14: what species are you referring to that is larger in South Korea by 263%?

→ It’s TOL as mentioned in previous sentence. We added ‘of TOL’ behind ‘relative difference’ to avoid confusion in the revised manuscript.

Page 8, line 1: add “respectively” after “(HCHO)”

→ Thank you for the comment. We included ‘respectively’ after ‘(HCHO)’.

Page 8, line 13: I think it would be clearer to say “For all emission inventories...” rather than “With all emission...”

→ Thank you for the comment. We changed “With all emission inventories” to “For all emission inventories” in the updated version of manuscript.

Page 8, lines 14-17: It was hard for me to figure out which simulations correspond to which numbers in these sentences. Reword to clarify?

→ We changed the sentence to “we conducted two additional model simulations using EDGAR-HTAP v3 that shows lowest bias of O₃ concentrations compared to DC-8 than

EDGAR-HTAP v2 and KORUS v5 over the SMA (mean bias = EDV2: -16.9, EDV3: -14.2, KOV5: -18.1 ppb)” adding bias information between parentheses in the revised manuscript.

Page 10, line 19: What kind of interpolation method was used?

→ The linear interpolation method is used for the vertical interpolation. We added “using linear interpolation method” after “vertically interpolated to the aircraft data” in the revised manuscript.

Page 11, lines 11-19: Can you say something about how much these meteorological biases affect your comparisons? For example, how much would temperature-dependent evaporative VOC emissions change over these temperature ranges?

→ Both anthropogenic and biogenic VOC emissions can be affected by air temperature (Huang et al., 2022; Song et al., 2019). In this response, we could calculate the impact of temperature on biogenic VOC emissions. The isoprene emission in MEGAN is calculated following the equation below (Guenther et al., 2006).

$$Emission = [EF][\gamma][\rho]$$
$$\gamma_{Temp} = E_{opt} \cdot \frac{C_2 \cdot \exp(C_1 \cdot x)}{C_2 - C_1 \cdot (1 - \exp(C_2 \cdot x))}$$
$$x = \frac{\left(\frac{1}{T_{opt}} - \frac{1}{T}\right)}{0.00831}$$

EF is emission factor ($\text{mg m}^{-2} \text{h}^{-1}$). ρ is normalized ratio. γ is an emission activity factor that can vary for different conditions such as leaf area index, temperature, vegetation type, leaf age, soil moisture, and canopy environment. E_{opt} and T_{opt} are empirical coefficients. C_1 and C_2 are constants. We calculated isoprene (ISO) emission sensitivity to temperature bias at each SYNOP station by changing T. The negative temperature biases resulted in reduced

isoprene emissions in South Korea (**Figure R6**). However, as discussed in 3.3.1, ISO is still overestimated for all regions.

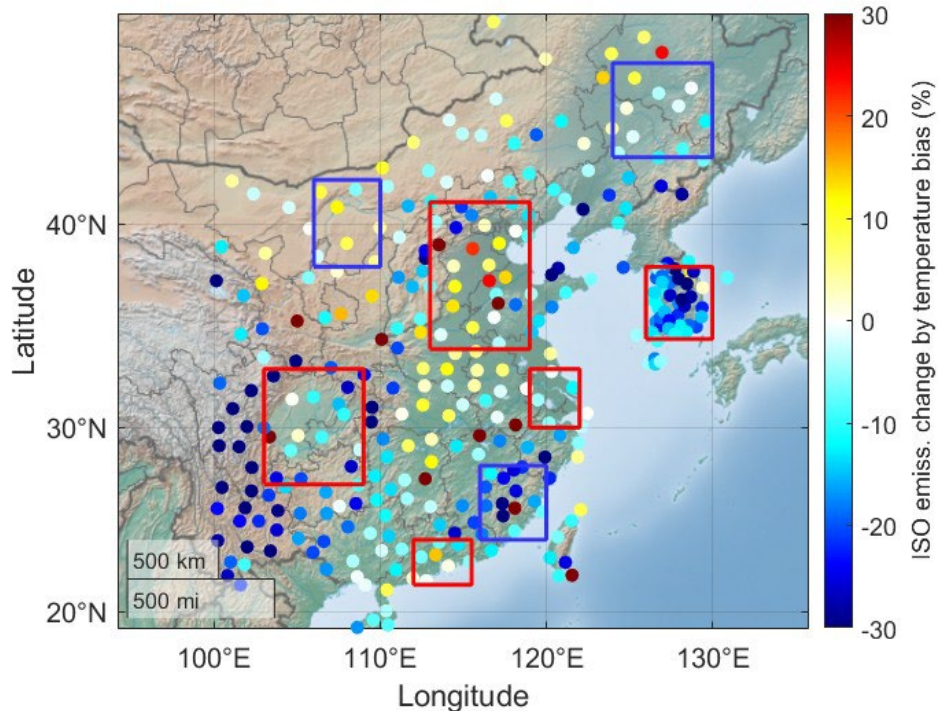


Figure R6. Relative isoprene (ISO) emission change from the temperature bias at the surface (unit = %).

Page 12, line 11: “all emissions inventories” instead of “all emissions”

→ Thank you for the comment. We changed “all emissions” to “all emission inventories”.

Page 17, line 3: ISO definition should be moved earlier to where it’s first used.

→ Agreed. We first defined isoprene as ISO in line 24 of page 13 in the revised manuscript.

Page 19, lines 23-24: can you use the biases calculated during the local case to draw some conclusions about the emissions inventory over China?

→ We added “, which implies that the insufficient local emissions of O₃ precursors in the emission inventories are much important than the Chinese emissions.” after “15.5-18.2 ppb” in the revised manuscript to clarify local VOC emission issues to the low model O₃ concentrations in South Korea.

Title: Unclear what ‘precursor’ refers to here. Is it O₃ and HCHO precursors? If so, perhaps rephrasing it as “ozone, formaldehyde, and their precursors” would be clearer.

→ Agreed. We changed the title.

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Responses to Reviewer 2's comments

The reviewer's comments are written in blue and our responses are in black.

The authors provide an overview of emission inventories and model simulations in East Asia using WRF-Chem v4.4 with three different emission inventories. To do so, they compare model results with various observations and conduct a sensitivity test by doubling CO and VOC emissions in China and South Korea. This study is significant as it shows the current state of emission inventories and represents points to improve simulated ozone. However, the following comments should be considered to enhance the study.

General comments:

1. In my opinion, the authors need to clarify the main topic and purpose of this study. I think that the research aims to evaluate model simulations using three emission inventories, comparing them with various observations and to conduct an analysis and sensitivity test of underestimated ozone. However, these aspects do not seem to be adequately explained in the introduction and abstract. Additionally, it should be made clear whether the analysis of underestimated ozone will focus on East Asia or be specific to South Korea. If the authors want to focus on East Asia, additional analysis for China has to be included, as discussed for South Korea. The scope of regions needs to be clarified.

→ To emphasize the focus and purpose of this paper, we clarified our objectives in the introduction and abstract. Previous modeling studies during the KORUS-AQ campaign period utilized old version of anthropogenic bottom-up emission inventories, using inversion or data assimilation method for the accurate air pollutants simulations (O₃, CO, HCHO, etc.). In contrast, our study employed recent versions of bottom-up emission datasets (EDGAR-HTAP v3 and KORUS v5) and expanded the analysis domain from South Korea to East Asia, including China. The revised manuscript includes brief descriptions of previous works (Choi et al., 2022; Goldberg et al., 2019; Miyazaki et al., 2019; Souri et al., 2020; Tang et al., 2019) are included in the introduction, clarifying our focus on O₃ and its precursors in both the abstract and introduction.

2. The paper discusses which emissions should improve and what causes the underestimation of ozone, comparing simulated species with observations. However, the discussion needs to be organized more. It mainly focuses on the underestimation of VOC emissions about underestimated ozone, but the impact of VOC emissions can vary depending on the ozone production regime (relative ratio to NO_x emissions). The paper independently compares VOC, NO_x, and O₃ mixing ratios, but these species are related to each other, and more effort considering them together is needed to understand model performance and emissions. Therefore, additional explanations about these regimes are necessary for the analysis during KORUS-AQ. In addition, NO_x emissions could contribute to VOC chemistry, resulting in ozone changes, so it would be great to describe them in the sensitivity analysis.

→ We agree with the suggestion that the simulated O₃ should be interpreted along with NO₂ and VOC emissions. We included **Table R1** in the revised manuscript to fully represent the differences between emission inventories and their simulated chemical regimes in different regions. Also, we explained the NO_x, VOC, biogenic isoprene emissions, and formaldehyde-to-NO₂ ratio in each region.

Specifically, in NCP, as the model simulated NCP as NO_x-saturated regime (FNR < 1), KOV5 simulated O₃ concentrations well because of higher reactive VOC emissions (TOL and XYL). In SCG and SEC, relatively high biogenic emissions from MEGAN compared to TOL and XYL led to high FNR (FNR > 1). These interpretations are incorporated into section 3.2.

Table R1. Comparison of total NO_x, TOL, XYL, biogenic isoprene emissions, and formaldehyde-to-NO₂ ratio (FNR) for different emission datasets in each regional box. The MEGAN biogenic isoprene emissions are equally applied to all simulations using different emission data. (unit = mol/s for emissions)

Type	emissions	NCP	SCG	YRD	PRD	KOR(SMA)	NEC	NOC	SEC
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	EDV3	0.44	1.30	0.32	0.52	0.43(0.18)	0.93	0.94	1.33
	KOV5	0.72	2.33	0.48	1.00	0.71(0.22)	1.44	1.49	1.91

To address the limitations of interpreting the efficient O₃ production regime with FNR, we additionally conducted sensitivity simulations with different emission; EDV3_Ch0.5NO_x, EDV3_Ch0.5VOC, EDV3_Ch0.5NO_xVOC, and EDV3_Ch0.25NO_x, representing EDGAR-HTAP v3 with 50% NO_x reduction, 50% VOC reduction, 50% NO_x and VOC reduction, and 75% NO_x reduction in China, respectively, as discussed in Kim et al. (2023).

Comparing relative biases of O₃ and NO₂ in each region and city (**Figure R1** and **R2**), the C5 case (50% VOC emission reduction only) exhibited the lowest O₃ biases in SCG and SEC, implying the need to reduce VOC emissions (biogenic and/or anthropogenic emissions) (**Figure R1**). In YRD and PRD, a 50% reduction in both NO_x and VOC emissions (C6 case) produced the most reasonable O₃ and NO₂ simulations.

Additionally, we evaluated O₃ and NO₂ simulations with different emissions at 12 mega cities in China and South Korea (**Figure R2**). EDV3 simulated O₃ and NO₂ well for the cities such as Beijing, Tianjin, Hangzhou, SMA, and Xian. In Chengdu and Chongqing, high O₃ and NO₂ biases are alleviated with 50% VOC emission reduction. For Shanghai, Nanjing,

Guangzhou, Shenzhen, and Wuhan, the C6 case shows the most reasonable O₃ and NO₂ simulations, while a simple 50% reduction of NO_x slightly increased O₃ biases.

Those analysis are detailed in section 4 (discussion) in the revised manuscript.

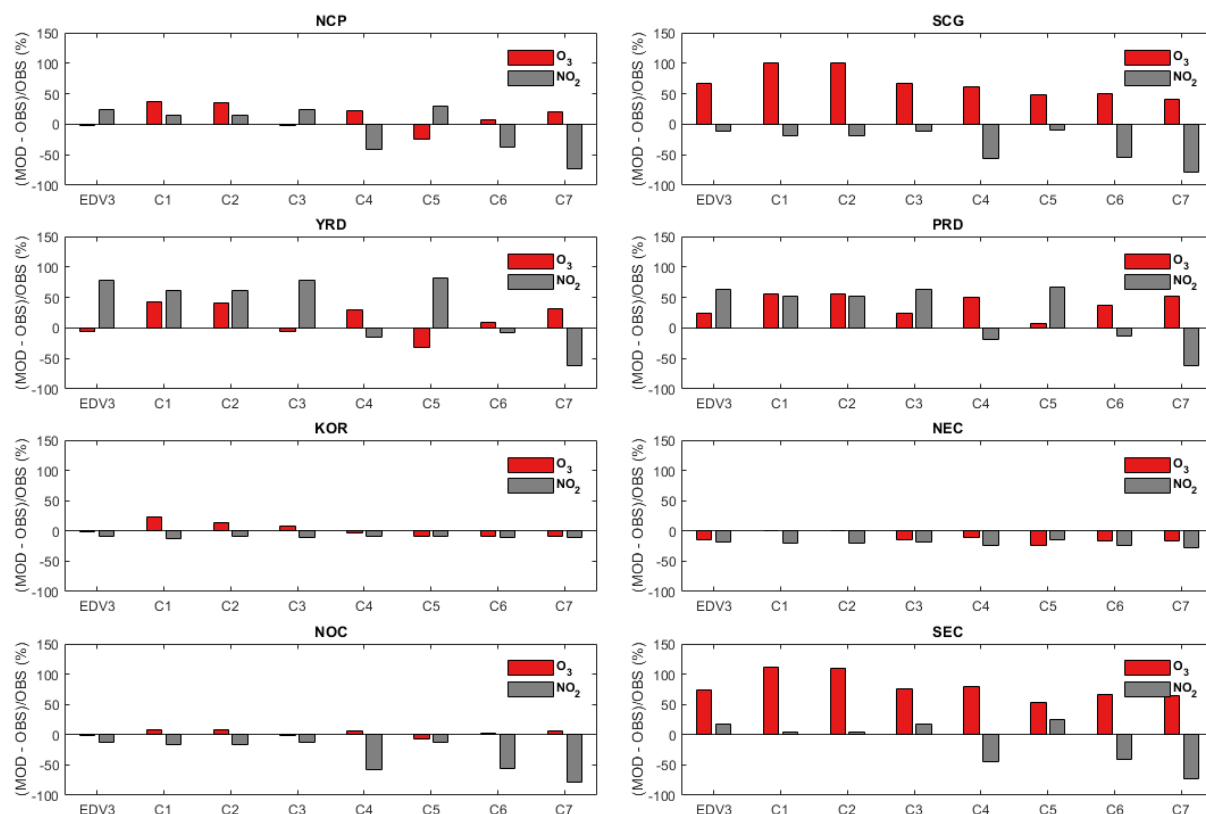


Figure R1. Comparison of relative biases ((Model-Observation)/Observation, unit=%) of daily O₃ and NO₂ at surface observation sites during the KORUS-AQ campaign period from sensitivity simulation (C1-7) with EDV3 in each region (NCP, SCG, YRD, PRD, KOR, NEC, NOC, and SEC). C1; EDGAR-HTAP v3 with double CO and VOC emission in China and South Korea, C2; EDGAR-HTAP v3 with double CO and VOC emission in China, C3; EDGAR-HTAP v3 with double CO and VOC emission in South Korea, C4; EDGAR-HTAP v3 with 50% NO_x reduction in China, C5; EDGAR-HTAP v3 with 50% VOC reduction in China, C6; EDGAR-HTAP v3 with 50% NO_x and VOC reduction in China, C7; EDGAR-HTAP v3 with 75% NO_x reduction in China.

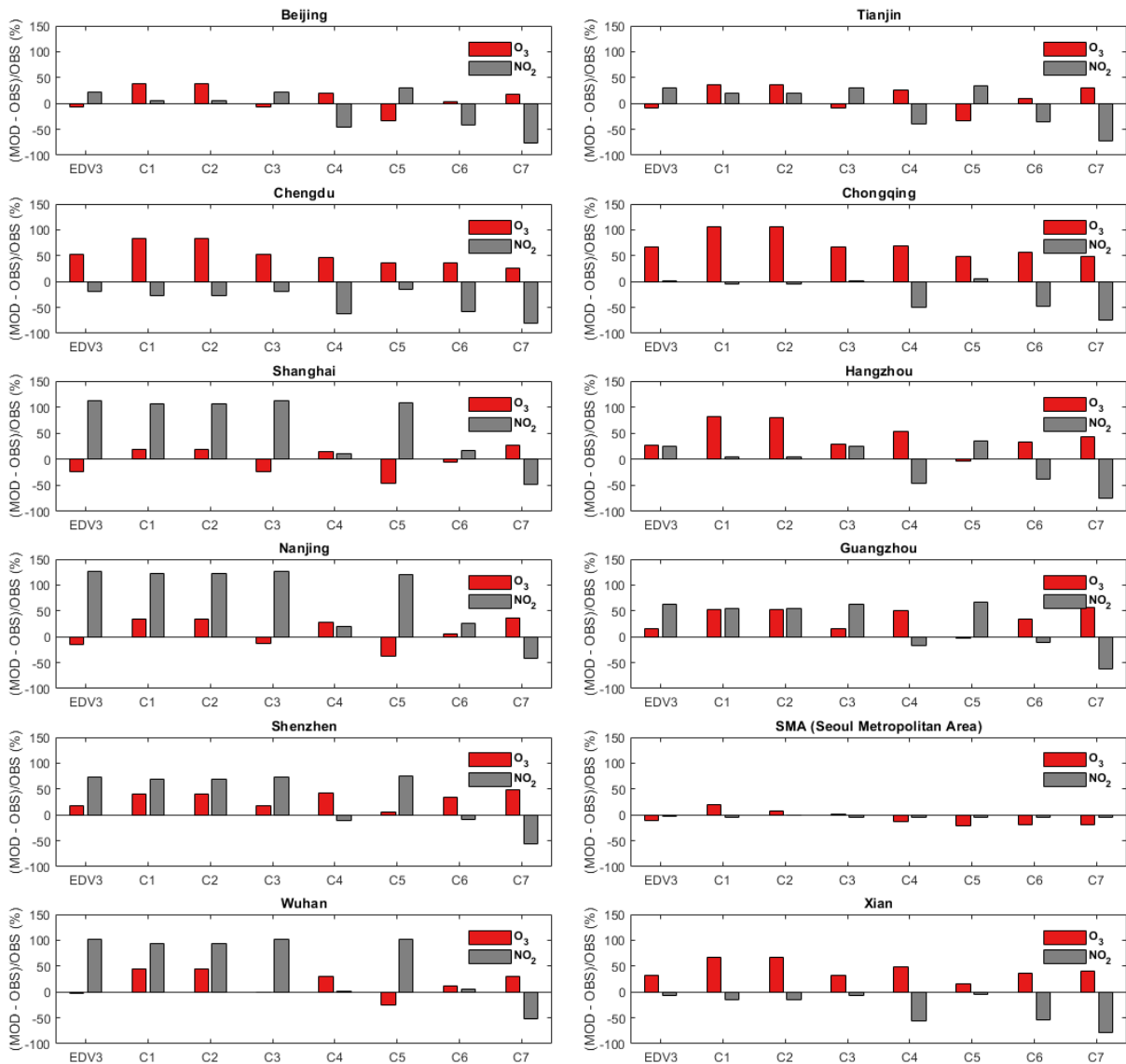


Figure R2. Same as **Figure R1** except that the region is changed to cities; Beijing (39.4-41.1N, 115.4-117.5E), Tianjin (38.55-40.25N, 116.7-118.1E), Chengdu (30.05-31.5N, 103-105E), Chongqing (28.15-32.25N, 105.3-110.2E), Shanghai (30.7-31.5N, 120.85-122E), Hangzhou (29.2-30.6N, 118.3-120.9E), Nanjing (31.2-32.65N, 118.35-119.25E), Guangzhou (22.55-24N, 112.9-114.05E), Shenzhen (22.4-22.9N, 113.7-114.65E), SMA (37.2-37.8N, 126.5-127.3E), Wuhan (29.95-31.4N, 113.65-115.1E), and Xian (33.65-34.75N, 107.65-109.9E).

3. The font sizes in the figures are small, and the figure resolutions in both the manuscript and supplementary document are low. Please enhance their readability.

→ The resolution issue may be made when the file is converted to pdf file. It is fixed and all the figures are updated with the large font size and high resolution in the revised manuscript.

4. Many paragraphs are overly lengthy, attempting to cover multiple topics within a single paragraph. Please ensure that paragraphs are concise.

→ The paragraphs are organized based on the analysis region and species for clearer representation. The paragraphs are segmented in P3 L9, P9 L19, P11 L20, P12 L13, P12 L23, P16 L9, P16 L12, P17 L5, P17 L17, P18 L7, P18 L14, P20 L14, and P20 L20.

Specific comments:

P3 L3-15: In my opinion, the paragraph should be separated at L9. Additionally, before providing an overall description of the paper, it is important to clearly explain what the authors want to convey through the paper and what scientific significance it holds.

→ We added some references (Choi et al., 2022; Goldberg et al., 2019; Miyazaki et al., 2019; Souril et al., 2020; Tang et al., 2019;) that are previously conducted for emission adjustments using chemical transport model and added the descriptions of our purpose of this paper at P3 L9.

P4 L18: The link is not open.

→ As we checked again, this link is still working but direct connection to this site through PDF file is not working. We changed the link to “<https://rda.ucar.edu/datasets/ds313.7/>”. We hope now this link is working.

P4 L21-22: Even so, in South Korea, the authors analyzed model performance over China. Therefore, the authors should discuss the effects of fire emissions on China.

→ We additionally simulated the WRF-Chem model using Fire Inventory from NCAR (FINN) v2.5 emissions (Wiedinmyer et al., 2022). The fire emission slightly increased averaged MDA8 O₃ concentrations by 1 ppbv (~ 1.6 %) in China. We added simple descriptions of this sensitivity test after “small impact on air quality simulations during the KORUS-AQ campaign period” at P4 L22 in the revised manuscript.

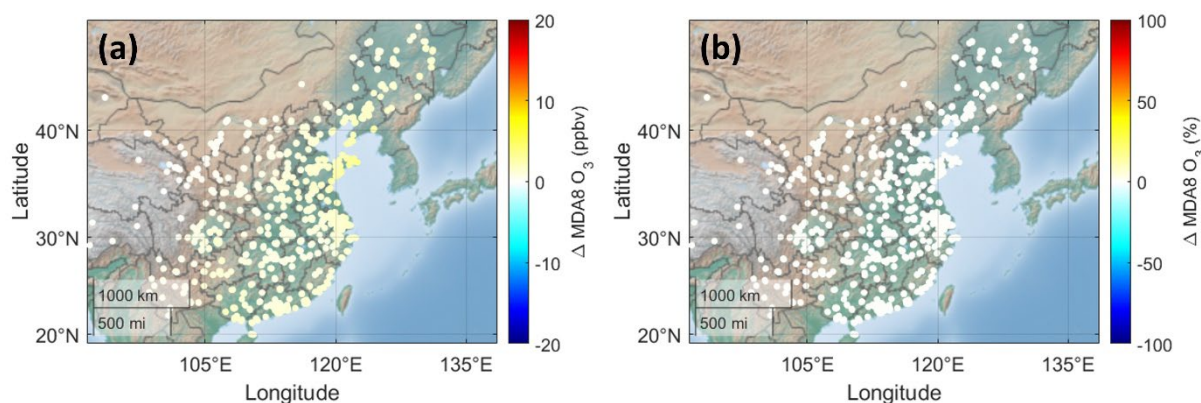


Figure R3. The (a) absolute and (b) relative differences of averaged MDA8 O₃ during the KORUS-AQ campaign period at the Chinese surface observations sites between WRF-Chem simulations with EDGAR-HTAP v3 (EDV3) and EDGAR-HTAP v3 with fire emissions (EDV3_Fire) (EDV3_Fire – EDV3).

P5 L2 What about “Anthropogenic bottom-up emission data” or something similar?

→ We changed “emissions” to “anthropogenic bottom-up emission inventories”.

P5 L4: The bottom row of Figure 1 is for only toluene or TOL (toluene +less reactive aromatics)?

→ It is model emission data that represents the sum of toluene and less reactive aromatics. We agree that it would be confusing to the readers whether it is toluene or TOL (toluene + less aromatics). We added “(toluene + less reactive aromatics)” after “TOL” in the revised manuscript.

Please rearrange the order of figures and tables in the manuscript and supplementary document. Table S3 appears before Table S2. Similarly, Figure 7 needs to be rearranged.

→ Thanks for your comment. It is fixed now and Figure 7 is rearranged to Figure 2.

P6 L23: Only toluene or TOL emissions?

→ It is sum of toluene and less reactive aromatics. We changed “toluene” to “TOL (toluene + less reactive aromatics)” in the revised manuscript.

P7 L1-4: Is CO a major precursor affecting ozone formation?

→ CO is one of the important precursors but has less impact than NO₂ and VOC in urban area. The term “major” can lead to misunderstanding, suggesting that CO is the primary precursor significantly affecting O₃ concentrations. To avoid this confusion, we removed the term “major” in this line.

P7 L4: It would be great to include boxes of three regions in Figure 1.

→ Agreed. We changed Figure 1 as **Figure R4** in the revised manuscript.

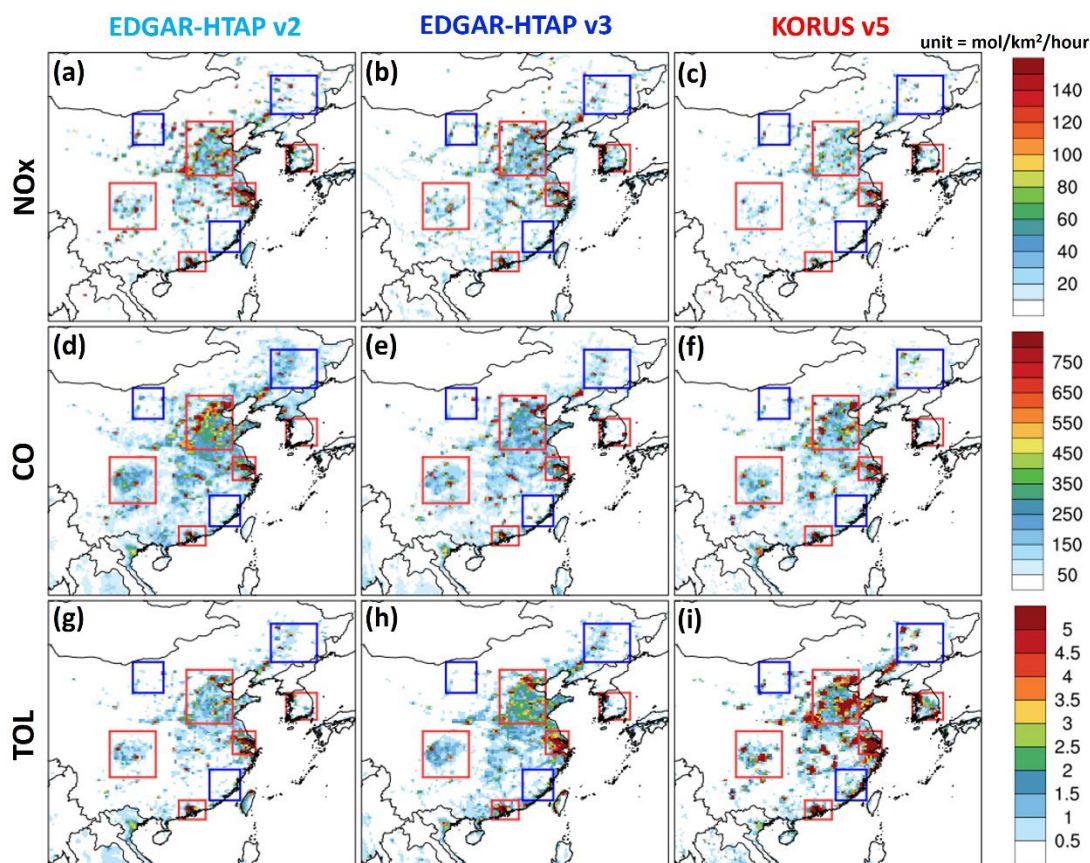


Figure R4. The averaged spatial distribution map of the NO, CO, and TOL (toluene + less reactive aromatics) emissions from (a, d, g) EDGAR-HTAP v2, (b, e, h) v3, and (c, f, i) KORUS v5 in May.

P7 L12-18: What about other VOC emissions? Toluene is one of reactive species with OH, but isoprene, ethene, and other species are also reactive with OH, leading to ozone production. It would be helpful to show how different total reactive VOC emissions are between three inventories.

→ We added an additional row of total NMVOC amounts for each region in **Table S3** (Supporting information). The EDGAR-HTAP v3 has larger total non-methane VOC (NMVOC) emissions over China compared to EDATA-HTAP v2 and KORUS v5 by 38 and 27 % respectively. The descriptions of total NMVOC differences are added to the revised manuscript.

P7 L23-P8 L1: The sentence could potentially mislead about VOC emissions, as formaldehyde is produced by the oxidation of many VOCs. Some VOC species might overestimate or underestimate.

→ We changed “CO and VOC for all emissions by -40% ($\pm 2\%$) and -25% ($\pm 1\%$) (HCHO)” to “HCHO, TOL, and XYL” to “CO, HCHO, TOL, and XYL for all emissions by -40% ($\pm 2\%$), -25% ($\pm 1\%$), -67% ($\pm 21\%$), -53% ($\pm 18\%$) respectively” to avoid misreading.

P8 L14-21: Regarding general comment #1, the authors should clarify the scope of this study for ozone underestimations.

→ The introduction and abstract sections are revised in the updated manuscript. It is changed as mentioned in the previous reply to general comment #1.

P9 L11-P10 L2: The paragraph could be separated at L19, and the sentence at L22-23 might be relocated after the description of NO₂ in South Korea. Please revise.

→ It is revised. And the L22-23 is relocated to P9 L17.

Section 3.2:

It would be helpful to explain what causes discrepancies between models and observations. In addition, the authors mentioned that low MDA8 in the models could be related to low VOC emissions. However, determining the regimes to which certain areas belong should involve considering both NO_x and VOCs. Underestimated ozone could result from both high NO_x concentrations in the model and low VOC emissions. In Section 3.2, the comparison of ozone and NO₂ between models and observations was discussed separately. Combining the analysis and discussion would enhance the manuscript and the understanding of emissions.

→ We acknowledge the comment to the section 3.2. We anticipate that the reply to general comment #2 is enough to explain this reply as previously discussed about NO_x and VOC emissions with FNR in **Table R1**.

P13 L16-19: The authors mentioned ground-based NO₂ observations over China and South Korea have positive biases in Section 2.3.2. If these positive biases are corrected, FNR might exceed 1, indicating a transition or NO_x-limited regime. In addition, it would be useful to provide the range of the FNR to determine ozone production regimes.

→ As previously explained in the reply of general comment #2, we also analyzed FNR from different emission inventories for each regional box. The model FNR is not affected by molybdenum issue because it is the model value itself. In NCP and KOR (or SMA), all emission inventories show very low FNR value (< 1) indicating those areas as highly NO_x-saturated regime. So, even though the NO₂ observations from molybdenum converter are corrected, the regime will not be dramatically changed.

Figure S5: Please plot NO₂ figures together with FNR and HCHO.

→ **Figure S5** is replaced to **Figure R6** in the revised Supporting Information.

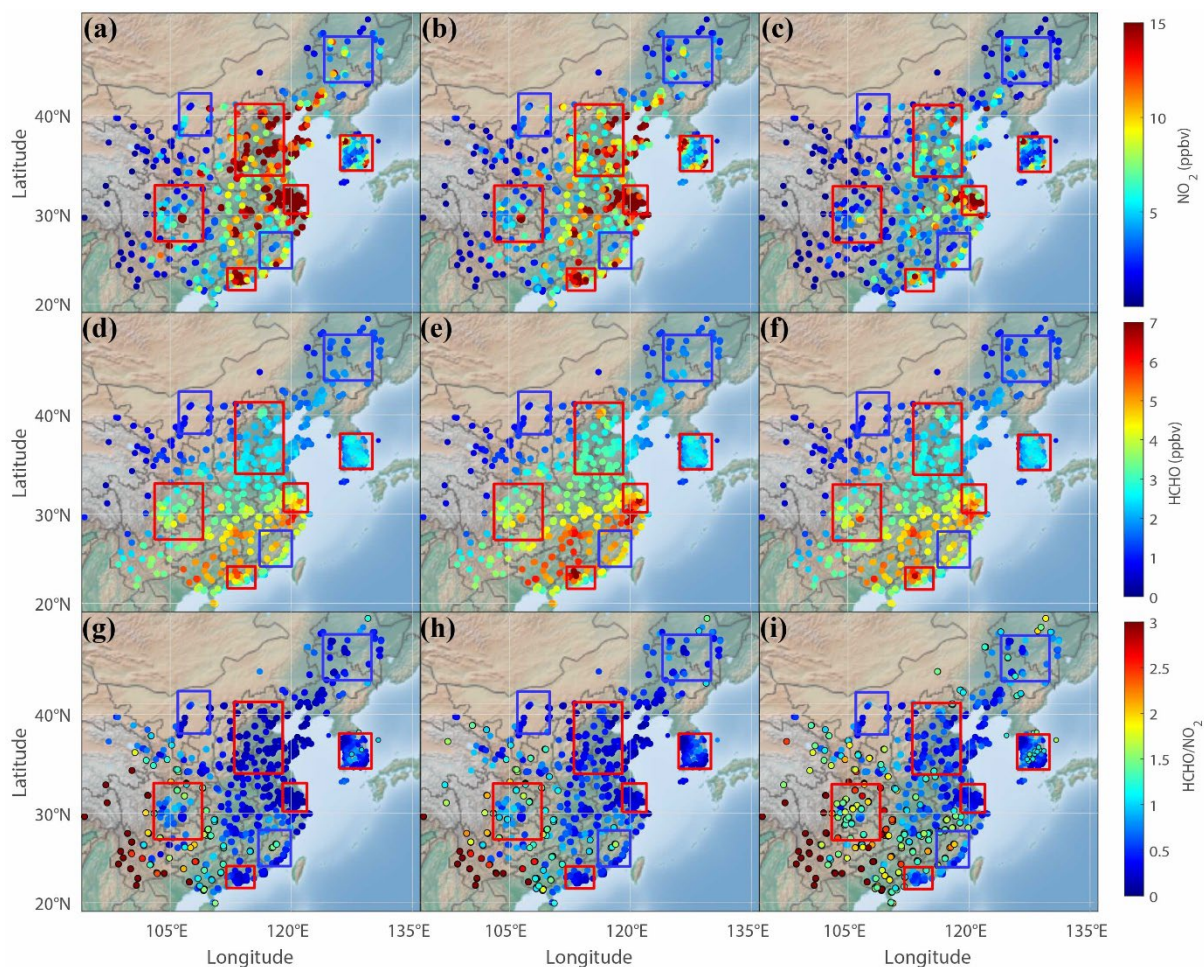


Figure R5. Simulated surface (a-c) NO_2 and (d-f) HCHO concentrations and (g-i) HCHO to NO_2 ratio (FNR) with (a, d, g) EDV2, (b, e, h) EDV3, and (c, f, i) KOV5 emissions for 14-16 LST. FNR greater than 1 is marked with black circles. The simulated NO_2 , HCHO, and FNR are linearly interpolated to ground-based observation sites.

P14 L11-13: In Figure 2, ozone mixing ratios simulated with EDV3, KOV5 are not substantially lower than the observations, and MDA8 ozone also appears as yellow in Figure 3.

→ We wanted to mention that there are small biases in YRD region. To avoid misreading, we changed “The lower bias of O_3 in YRD” to “The reason why O_3 is well simulated in YRD, even though NO_2 is highly overestimated in this region,” in the revised manuscript.

P14 L16-17: The lowest bias of -5.5 ppbv was observed for KOV5 in South Korea.

→ We wanted to emphasize that EDV3 shows the lowest absolute value of biases. For the NO₂, EDV3 shows the smallest bias in South Korea by -1.9 ppb. We changed ‘the lowest’ to ‘the smallest’ in the revised manuscript.

P16 L10: It would be helpful to describe why the authors showed HCHO.

→ We used HCHO as one of the proxies of VOC concentrations, though it is the product of many other VOCs in reaction with reactive gases. The sentence, “we also evaluated the model HCHO, which can be formed by oxidation of other VOCs but also directly emitted by anthropogenic sources, to investigate potential issue of anthropogenic VOC emissions”, is added after “Additionally,”.

P17 L5-6: Overall ozone production regimes were discussed in Section 3.2. However, to lead to your conclusion that underestimated ozone results from low VOCs during KORUS-AQ, it would be helpful to show FNR during KORUS-AQ. Also, the lifetime of CO is too long to significantly contribute to ozone. How much does the CO oxidation contribute to ozone? Is it related to background ozone concentration, and then how much?

→ We anticipate that the newly added Table 3 will explain the FNR in South Korea and SMA. It represents South Korea and SMA as highly NO_x-saturated regime. We included **Figure R6** to the revised Supporting Information to show the CO contribution to O₃ concentrations in SMA. The reduced bias of CO derived from doubled China anthropogenic CO emissions (-96 to -63 ppb) slightly increased O₃ by 1.4 ppb compared to DC-8 for all flight observations. The overall descriptions of this results will be added at the P16 L9 to explain the CO impact on O₃ concentrations in SMA.

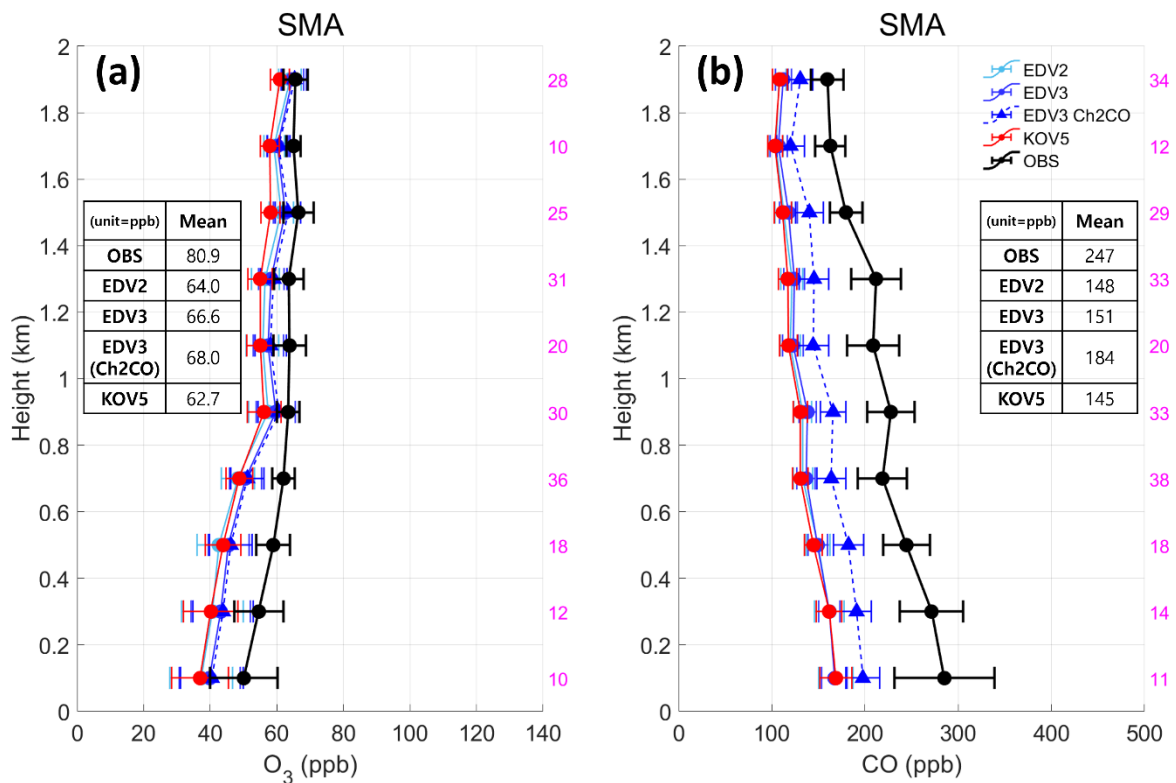


Figure R6. Vertically averaged (a) O₃ and (b) CO from DC-8 (black), EDV2 (sky blue), EDV3 (blue), EDV3 with double CO emission in China (EDV3 Ch₂CO) (blue dashed), and KOV5 (red) 3 in SMA under 2 km height above ground level. The 1/2 of standard deviations are represented with black whiskers in each 200m layer. The sample number is presented with magenta color on the right side of the plots.

P17 L14-16: During KORUS-AQ, ground-based NO₂ was measured from a photolytic converter following the description in P10 L6-8, correct? If data from both molybdenum and photolytic converters were available, why did you choose to use data from molybdenum converter for the comparison? Also, low NO_x from the model is a result influenced by various factors, including emissions, chemistry, PBL, and others. It would be helpful to describe which factors are affected rather than just saying the comparison with observations.

→ NO₂ observations by photolytic converter is only available at the Olympic Park. So, we used NO₂ observed by molybdenum converter, which is measurement instrument of Airkorea, for the analysis in KOR and SMA domain.

There are still other possibilities of NO₂ underestimations; 1) the emission factor used in this study is from Los Angeles basin that might be not adequate to SMA, 2) the uncertainty of HO_x and RO_x radicals from other sources can affect the NO₂ concentrations. We included those possible uncertainties at P17 L17.

P17 L 20-22: Simulated TOL from KOV5 also shows significant differences from observations below 1 km except at the surface.

→ “below 1 km” is replaced by “at surface level and had the lowest bias of -0.9 and -0.1 ppb respectively under 2 km”.

P18 L13-14: It would be great to explain why surface and vertical columns exhibit different diurnal patterns.

→ The diurnal patterns of surface NO₂ concentration are attributed to the diurnal cycle of PBL height. In the morning, NO₂ is concentrated near the surface layer due to under-developing mixed layer height. In the afternoon, as the PBL height grows, NO₂ is more mixed and distributed to vertically higher altitudes. On the other hand, vertical column NO₂ density is high in the afternoon because of consistent emission of NO_x during the daytime. Those explanations are included in the revised manuscript.

P18 L15: VCD patterns differ between model and Pandora.

→ “The simulated and observed HCHO show similar diurnal variations” is deleted.

P19 L 2-4: How did diurnal profiles contribute to reducing biases?

→ Diurnal profiles did not reduce the negative biases directly. We wanted to point out that TOL and XYL are still underestimated compared to surface observations at Olympic Park as

DC-8 shows underestimation of TOL and XYL under 2 km in SMA. We revised “reduced the model negative biases from EDV2 and EDV3” to “exhibited smaller negative biases than EDV2 and EDV3” to avoid confusion.

P19 L18-19: Even though the authors cite Peterson et al. (2019) to separate local and transport cases, please provide a brief description for readers.

→ “Stagnant and Blocking is the period that large anticyclone is located over South Korea, and Transport case is the period that South Korea is largely affected by long-range transport of air pollutants by westerly wind” is added after the sentence.

P20 L22-24: When considering the contributions of South Korea’s CO and VOCs emissions, calculated as the difference between ch2 and chko2, the increase of CO and VOC emissions does not lead to improvements in CO and VOCs. The difference in CO is only 5 ppbv, while the difference in HCHO is 32 ppb, resulting in overestimation of the model. In addition, in chko2, doubling emissions in China might affect O₃, CO, and HCHO in Korea because of transport. Also, local cases may be influenced by the transport of species with relatively long lifetimes. To describe the effects of South Korea’s emissions on ozone, CO, and HCHO, the author could simulate the model with only doubling CO and VOC emissions in Korea.

→ To avoid the transport impact when interpreting the emissions in South Korea, we conducted an additional simulation using EDGAR-HTAP v3 with double CO and VOC emission in South Korea only (EDV3_Ko2). In the Local case, although the EDV3_Ko2 reduced biases of O₃, CO, and HCHO over the SMA, doubled CO and VOC emissions in both South Korea and China (EDV3_ChKo2) showed the lowest biases. For the Transport case, doubling CO and VOC emissions in South Korea (EDV3_Ko2) slightly reduced O₃ and CO biases, but resulted in an overprediction of HCHO. We included those results in section 3.4 in the revised manuscript.

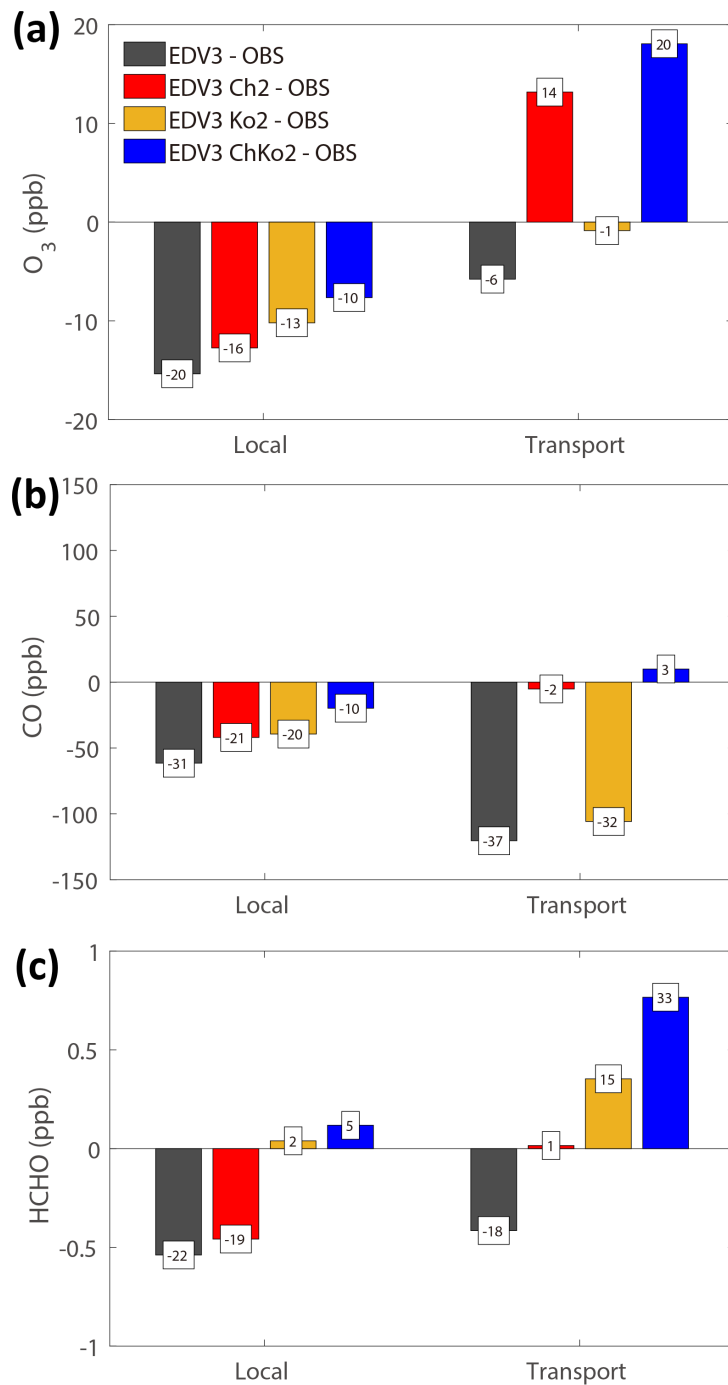


Figure R7. The biases in (a) the model O₃, (b) CO, and (c) HCHO concentrations (bars) relative to the DC-8 observations under 2 km height over SMA (dark gray: EDV3, red: EDV3 Ch2, orange: EDV3 Ko2, red: EDV3_ChKo2): (left panel) Local and (right panel) Transport case. Fractional differences (%) are shown in the white boxes.

Figures 2-6: Adding boxes to the plots in the second and third rows would be helpful for easily recognizing the regions.

→ The boxes are included in Figure 3, 5, and 6.

TableS4: Please provide definitions for the species names in MOZART and SAPRC-99.

→ The Table R2-R3 are included in the revised Supporting Information.

Table R2. The list of MOZART species (Emmons et al., 2010).

Species	Atomic composition	Note
ISOP	C ₅ H ₈	isoprene
SO2	SO ₂	sulfur dioxide
NO	NO	nitric oxide
NO2	NO ₂	nitrogen dioxide
CO	CO	carbon monoxide
C2H6	C ₂ H ₆	ethane
C2H5OH	C ₂ H ₅ OH	ethanol
CH3OH	CH ₃ OH	methanol
C3H8	C ₃ H ₈	propane
BIGALK	C ₅ H ₁₂	lumped alkanes C>3
TOLUENE	C ₆ H ₅ (CH ₃)	lumped aromatics
C2H4	C ₂ H ₄	ethene
BIGENE	C ₄ H ₈	lumped alkenes C>3
CH2O	CH ₂ O	formaldehyde
CH3CHO	CH ₃ CHO	acetaldehyde
CH3COCH3	CH ₃ COCH ₃	acetone
MEK	CH ₃ C(O)CH ₂ CH ₃	methyl ethyl ketone
NH3	NH ₃	Ammonia

Table R3. The list of SAPRC99 species (Carter, 2000).

Species	Note
ISOP	Isoprene
SO2	Sulfur dioxide
NO	Nitric oxide
NO2	Nitrogen dioxide
CO	Carbon monoxide
ALK1	Alkanes and other non-aromatic compounds that react only with OH, and have kOH < 5 x 10 ² ppm ⁻¹ min ⁻¹ . (Primarily ethane)
ALK2	Alkanes and other non-aromatic compounds that react only with OH, and have kOH between 5 x 10 ² and 2.5 x 10 ³ ppm ⁻¹ min ⁻¹ . (Primarily propane and acetylene)
ALK3	Alkanes and other non-aromatic compounds that react only with OH, and have kOH between 2.5 x 10 ³ and 5 x 10 ³ ppm ⁻¹ min ⁻¹ .
ALK4	Alkanes and other non-aromatic compounds that react only with OH, and have kOH between 5 x 10 ³ and 1 x 10 ⁴ ppm ⁻¹ min ⁻¹ .
ALK5	Alkanes and other non-aromatic compounds that react only with OH, and have kOH greater than 1 x 10 ⁴ ppm ⁻¹ min ⁻¹
ARO1	Aromatics with kOH < 2x10 ⁴ ppm ⁻¹ min ⁻¹ .
ARO2	Aromatics with kOH > 2x10 ⁴ ppm ⁻¹ min ⁻¹ .
MEOH	Methanol
ETHE	Ethene
OLE1	Alkenes (other than ethene) with kOH < 7x10 ⁴ ppm ⁻¹ min ⁻¹ . (Primarily terminal alkenes)
PHEN	Phenol
CRES	Cresols
HCHO	Formaldehyde
CCHO	Acetaldehyde and Glycolaldehyde
RCHO	Lumped C3+ Aldehydes
BALD	Aromatic aldehydes (e.g., benzaldehyde)
GLY	Glyoxal
MGLY	Methyl Glyoxal
BACL	Biacetyl
MACR	Methacrolein
ACET	Acetone
MEK	Ketones and other non-aldehyde oxygenated products which react with OH radicals slower than 5 x 10 ⁻¹² cm ³ molec ⁻² sec ⁻¹
PRD2	Ketones and other non-aldehyde oxygenated products which react with OH radicals faster than 5 x 10 ⁻¹² cm ³ molec ⁻² sec ⁻¹
MVK	Methyl Vinyl Ketone
IPRD	Lumped isoprene product species
NH3	Ammonia

Table S7-S8: Please clarify the time period for the data.

→ Those are corrected in the revised manuscript. We added ‘for the KORUS-AQ campaign period’ in the Table captions.

Figure 8: Please indicate the color for observations.

→ It is corrected in the revised manuscript.

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