

Reply to comments from Referee #1

First of all, thank you for your valuable comments and suggestions. Following your comments, we attempt to clarify and improve the manuscript by eliminating, modifying, and adding several parts from/into the original text. The added or modified parts are painted in a blue color in the revised manuscript.

General Comment

In this study, the author attempted to sequentially incorporate several potential HONO sources and processes into the CMAQ modeling framework. And the simulation performances of the modified CMAQ models were then evaluated by comparing the modeled HONO mixing ratios with the HONO mixing ratios observed at the Olympic Park station in Seoul, South Korea. The simulation results have been improved than the Original CMAQ model. However, there are several issues in the article.

Specific Comments

Comment 1 In Lines 179-180, the average values of simulation results in Table S2 are not sufficient to confirm the good prediction of the model, and the comparisons of time series of observation and modeled outputs are needed.

Reply: Considering your comments, we added Fig. S1 and revised Table S2 into the supplementary material. Fig. S1 shows the temporal variations of modeled and observed relative humidity (RH), temperature (T), pressure (P), wind speed (WS), and wind direction (WD) during the target periods. Please, refer to Fig. S1 and Table S2. The purpose of original Table S2 was that our meteorological model simulations produced reasonable quality of meteorological fields.

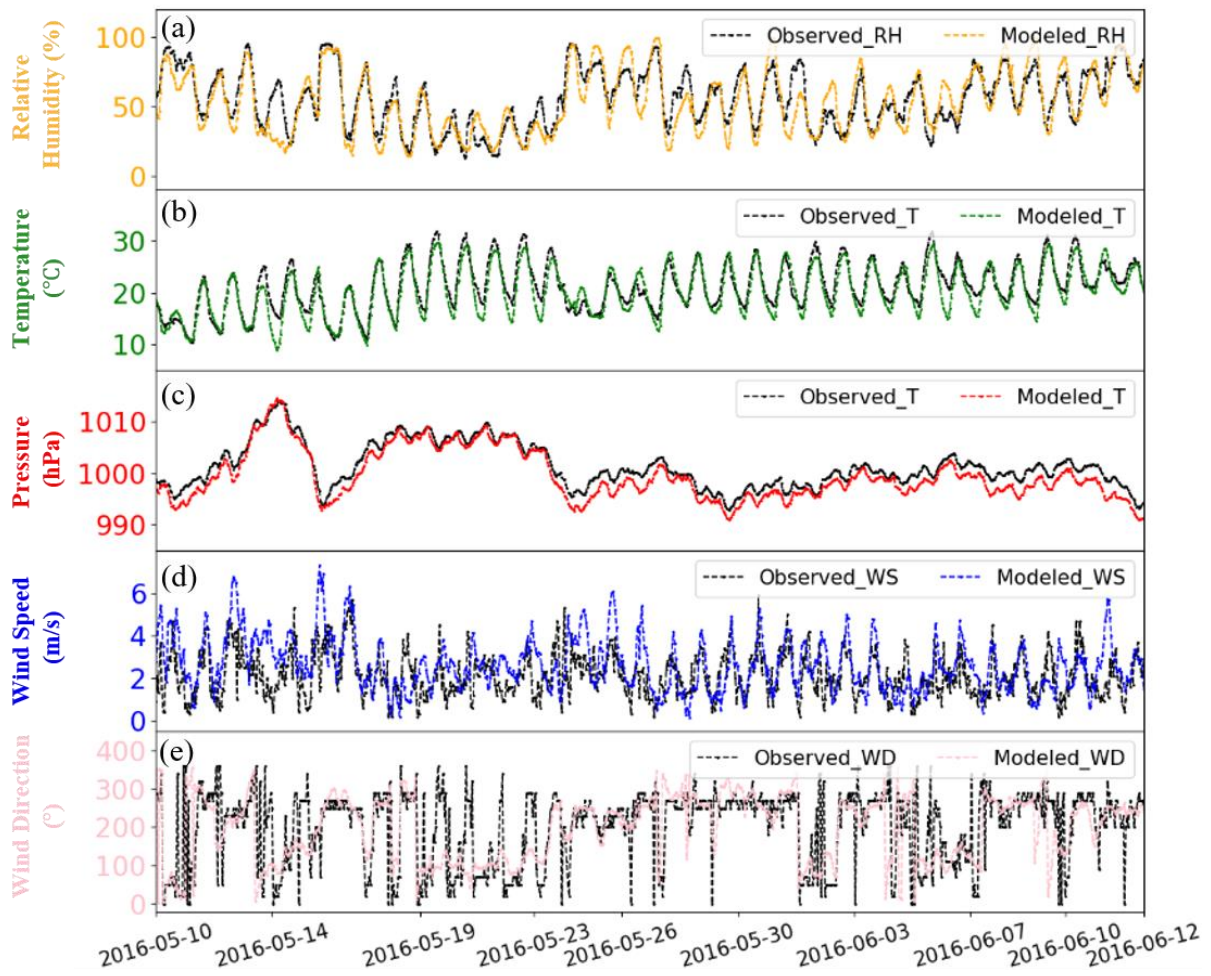


Fig. S1. Temporal variations of (a) relative humidity (RH); (b) temperature (T); (c) pressure; (d) Wind speed (WS) at 10m above the surface; and (e) wind direction (WD) at 10m above the surface during period of the KORUS-AQ campaign.

Table S2. Statistical analysis of modeled and observed meteorological parameters at the Olympic Park station during the period of the KORUS-AQ campaign.

Parameter	Observed mean	Modeled mean	R	RMSE	MB	IOA
RH (%)	55.81	53.33	0.85	11.95	-2.48	0.92
T (°C)	21.27	20.28	0.93	1.96	-0.99	0.96
Pressure (hPa)	1001.38	999.62	0.98	2.01	-1.77	0.95
WS (m s ⁻¹)	2.14	2.65	0.47	1.30	0.51	0.66
WD (°)	202.71	196.01	0.53	88.87	-6.70	0.75

Comment 2 In Table 1, the emission factor of gasoline and diesel vehicles are 0.8% and 2.3% in all studies area. However, there are significant differences in traffic volumes and vehicle types in different areas.

Reply: The traffic emissions used in this study were from the anthropogenic emissions inventory of KORUSv5.0. The KORUSv5.0 was developed based on detailed considerations of vehicle fuel-types and traffic volumes across our domain (East Asia) (Please, refer to Woo et al., 2012; Woo et al., 2020).

Comment 3 In lines 205-207, is the equation for the relationship between J_{NPHE} and J_{NO_2} also in this reference (Stockwell et al., 1990)? And how J_{HONO} is calculated?

Reply: The relationship between J_{NPHE} and J_{NO_2} was first proposed by Bejan et al. (2006). Since then, it has been utilized in many modeling mechanisms, including SAPRC07TC (Carter et al., 2010). Regarding this, please refer to lines 207-208 in our revised manuscript. The J values for HONO and other chemical species were calculated in CMAQ modeling via PHOT subroutine, using quantum yields and cross-section data. In our CMAQ model simulations, they were calculated based on a look-up table involving J_values at different latitudes, altitudes, and solar zenith angles. After the initial J values were calculated under the clear-sky conditions, such values were corrected with cloud coverage data (Byun and Ching, 1999).

Comment 4 In Table 3, how were HONO emission rates of traffic calculated?

Reply: The HONO emissions were determined based on the emission ratios of HONO to NO_x from gasoline and diesel vehicle exhausts. These ratios were discussed in lines 233-235 in our revised manuscript.

Comment 5 In lines 276-277, why are the uptake coefficients of NO_2 at nighttime and daytime are 8.0×10^{-6} and 1.3×10^{-4} , respectively? Also how is the 900 for this factor taken into account? The same issue in lines 292-293.

Reply: We adopted the uptake coefficients of NO_2 on aerosol and ground surfaces from the studies of Czader et al. (2012) and VandenBoer et al. (2013). Also, the factor was selected to be $900 \text{ W}\cdot\text{m}^{-2}$ based on a sensitivity test (Czader et al., 2012). We clarified this point in our revised manuscript (Please, refer to lines 281 – 282, 296 – 297, and Table R1).

Table R1. Statistical analysis with different parameterization of γ_{NO_2} for modeled and observed HONO mixing ratios at the Olympic Park stations, Seoul, Korea.

EXP	Parameterizations of γ_{NO_2}	References	Observed mean	Modeled mean	IOA	MB	RMSE
SEN_A	$\gamma_{a,\text{NO}_2} = 2.0 \times 10^{-5} \times \left(\frac{\text{light intensity}}{400}\right)$ (daytime) $\gamma_{a,\text{NO}_2} = 1.0 \times 10^{-6}$ (nighttime) $\gamma_{g,\text{NO}_2} = 1.3 \times 10^{-6} + 4.8 \times 10^{-8} \times [\text{SWR}]$ (daytime) $\gamma_{g,\text{NO}_2} = 6.5 \times 10^{-7}$ (nighttime)	Kleffmann et al. (1998) Vogel et al. (2003) Zhang et al. (2023) Marion et al. (2021)	1.35	1.44	0.654	0.09	1.28
SEN_B	$\gamma_{a,\text{NO}_2} = 1.3 \times 10^{-4} \times \left(\frac{\text{light intensity}}{400}\right)$ (daytime) $\gamma_{a,\text{NO}_2} = 2.0 \times 10^{-6}$ (nighttime) $\gamma_{g,\text{NO}_2} = 5.8 \times 10^{-6} \times \left(\frac{\text{light intensity}}{400}\right)$ (daytime) $\gamma_{g,\text{NO}_2} = 3.1 \times 10^{-7}$ (nighttime)	Xue et al. (2021) Mong et al. (2009) Yu et al. (2021)	1.35	1.18	0.713	-0.17	1.11
SEN_C	$\gamma_{a,\text{NO}_2} = 4.0 \times 10^{-5} \times \left(\frac{\text{light intensity}}{400}\right)$ (daytime) $\gamma_{a,\text{NO}_2} = 8.0 \times 10^{-6}$ (nighttime) $\gamma_{g,\text{NO}_2} = 5.8 \times 10^{-6} \times \left(\frac{\text{light intensity}}{400}\right)$ (daytime) $\gamma_{g,\text{NO}_2} = 3.1 \times 10^{-7}$ (nighttime)	Stemmler et al. (2007) Liu et al. (2019) Yu et al. (2021)	1.35	1.13	0.729	-0.22	1.06
SEN_D	$\gamma_{a,\text{NO}_2} = 2.0 \times 10^{-5} \times \left(\frac{\text{light intensity}}{400}\right)$ (daytime) $\gamma_{a,\text{NO}_2} = 1.0 \times 10^{-6}$ (nighttime) $\gamma_{g,\text{NO}_2} = 5.8 \times 10^{-6} \times \left(\frac{\text{light intensity}}{400}\right)$ (daytime) $\gamma_{g,\text{NO}_2} = 3.1 \times 10^{-7}$ (nighttime)	Kleffmann et al. (1998) Vogel et al. (2003) Yu et al. (2021)	1.34	1.62	0.733	0.27	1.02
SEN_E	$\gamma_{a,\text{NO}_2} = 4.0 \times 10^{-5} \times \left(\frac{\text{light intensity}}{900}\right)$ (daytime) $\gamma_{a,\text{NO}_2} = 8.0 \times 10^{-6}$ (nighttime) $\gamma_{g,\text{NO}_2} = 5.8 \times 10^{-6} \times \left(\frac{\text{light intensity}}{900}\right)$ (daytime) $\gamma_{g,\text{NO}_2} = 3.1 \times 10^{-7}$ (nighttime)	Stemmler et al. (2007) Liu et al. (2019) Yu et al. (2021) Vandenboer et al. (2013)	1.35	1.05	0.742	-0.30	1.06
SEN_F	$\gamma_{a,\text{NO}_2} = 1.3 \times 10^{-4} \times \left(\frac{\text{light intensity}}{900}\right)$ (daytime) $\gamma_{a,\text{NO}_2} = 8.0 \times 10^{-6}$ (nighttime) $\gamma_{g,\text{NO}_2} = 5.8 \times 10^{-6} \times \left(\frac{\text{light intensity}}{900}\right)$ (daytime) $\gamma_{g,\text{NO}_2} = 5.0 \times 10^{-7}$ (nighttime)	This study	1.35	1.18	0.764	-0.17	1.12

Comment 6 In Line 412, is there much improvement in model results if NO₂ daytime sources are enhanced?

Reply: The consideration of photo-induced NO₂-to-HONO conversion improved the mean bias from -0.79 to -0.28 ppb (please, refer to Fig. 3 or Table 4). However, there is still room for further improvements, with respect to direct HONO emissions and vertical parameterizations of NO₂ (Gilgorovski et al., 2016; Guo et al., 2020; Tang et al., 2024).

Comment 7 In Line 447, “Its contribution increases to 4.2% during the daytime”, which sources is this contribution?

Reply: The “Its contribution” meant the contribution from the HET_A during the daytime. However, this sentence was removed in our revised manuscript, because we thought it is a little bit out of context.

Reference cited in this response:

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