Supporting Information for *Atmos. Chem. Phys.* manuscript:

Atmospheric Measurements at the Foot and the Summit of Mt. Tai - Part II: HONO Budget and Radical (RO_x + NO_3) Chemistry in the Lower Boundary Layer

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1. Lifetimes of HONO and NO_x and direct HONO emissions ($HONO_{emi}$)

As discussed in the main text (Section 3.2.2.1), the contribution of direct emission on the observed HONO could be overestimated when using a constant $\Delta HONO/\Delta NO_x$, especially during the daytime due to the distinctly different lifetimes of HONO (τ(HONO)) and NO_x ($\tau(NO_x)$). Therefore, during the daytime, when τ(HONO) was shorter than 1 h, $HONO_{emi}$ was corrected by multiplying the ratio of $\tau(HONO)/\tau(NO_x)$ (see (Eq-3) in the main text). τ(HONO) against OH and photolysis was directly obtained from F0AM model simulations (Wolfe et al., 2016). $\tau(NO_x)$ depends NO_2 lifetime and NO/NO_2 ratio regarding the net loss of NO_x is mainly in the form of HNO_3 produced through OH or NO_3 induced reactions. The equation is shown in (Eq-S1) (Seinfeld and Pandis, 2016).

$$\tau(NO_x) = \tau(NO_2) * (1 + \frac{NO}{NO_2}), \quad \text{(Eq-S1)}$$

Net NO_2 loss was through reactions of NO_2 + OH → HNO_3, NO_3 + VOCs → HNO_3, and NO_3 + NO_2 + wet surface → HNO_3, which were considered to calculate $\tau(NO_2)$. Results on HNO_3 production rate were presented in Figure 12 and discussed in Section 3.3.3.1 of the main text. Results on daytime τ(HONO), $\tau(NO_x)$, and $HONO_{emi}$ were shown in Figure S6.

Figures

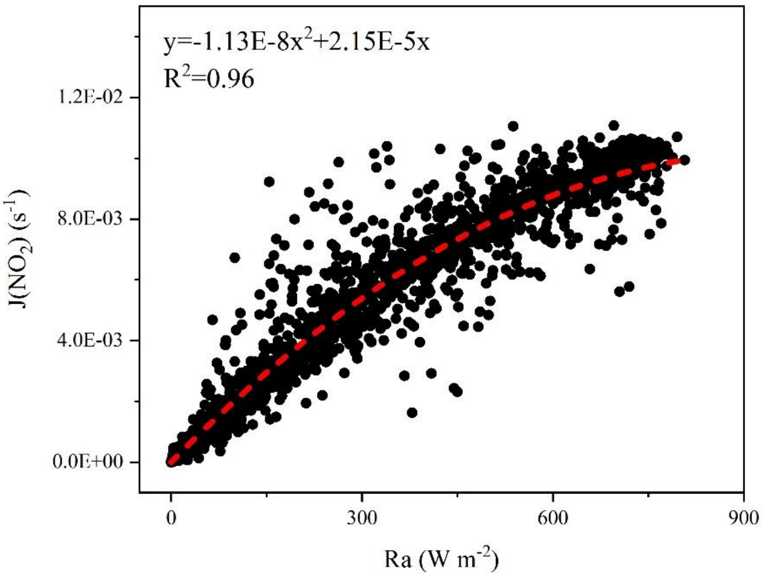


Figure S1: Correlation between the measured $J(NO_2)$ and solar irradiance (Ra).

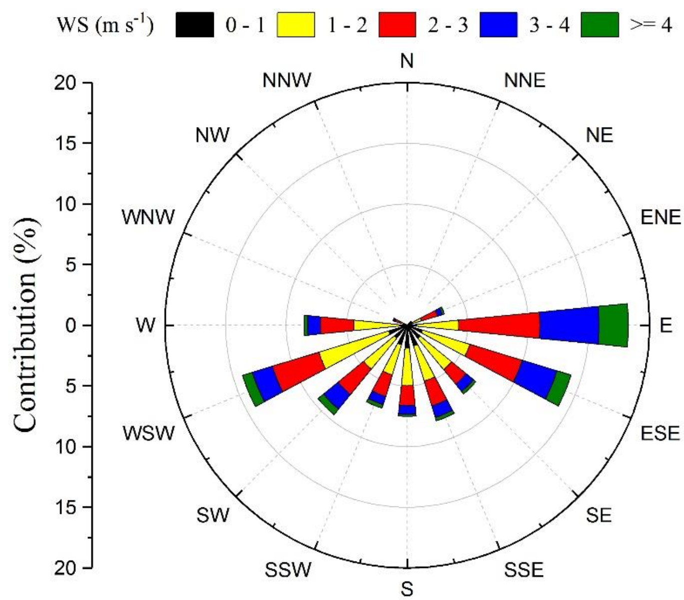


Figure S2: Wind rose plot for the wind measurements at the foot of Mt. Tai

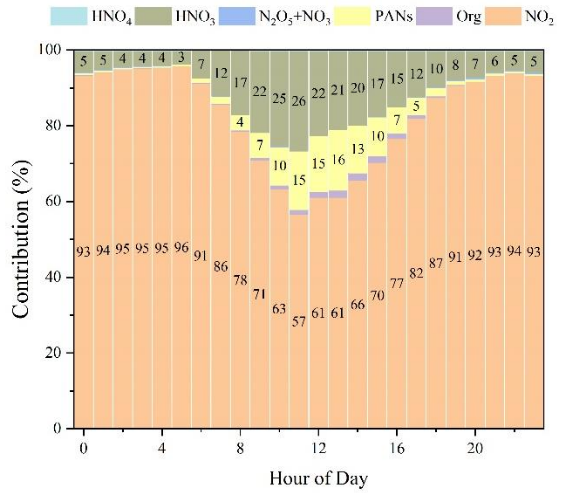
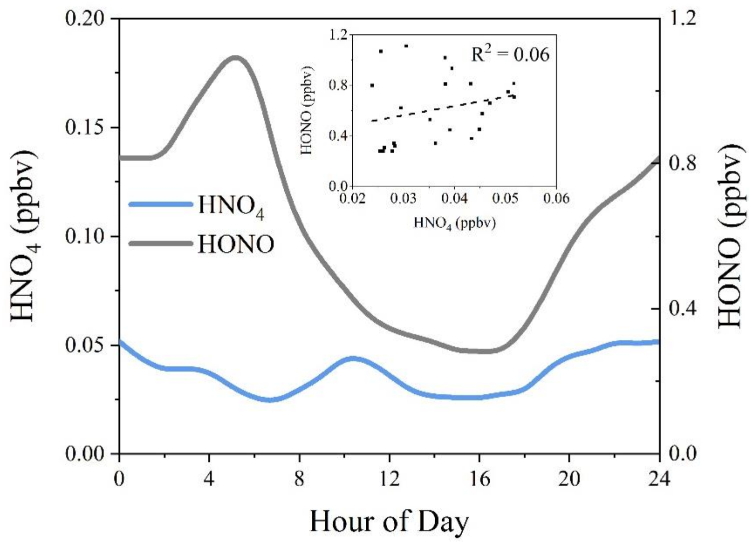


Figure S3: Relative contribution of each NO_z* species. PANs = PAN + PPN + MPAN, and Org represents organic nitrates*.



75 **Figure S4: Diurnal variations of HONO and HNO_4 and their correlations.**

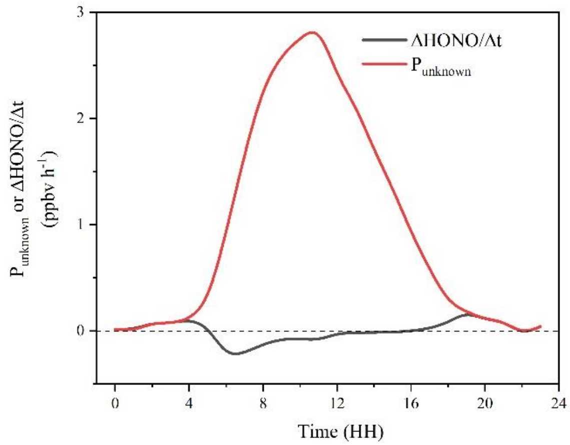


Figure S5: Diurnal variations of $P_{unknown}$ and ΔHONO/Δt.

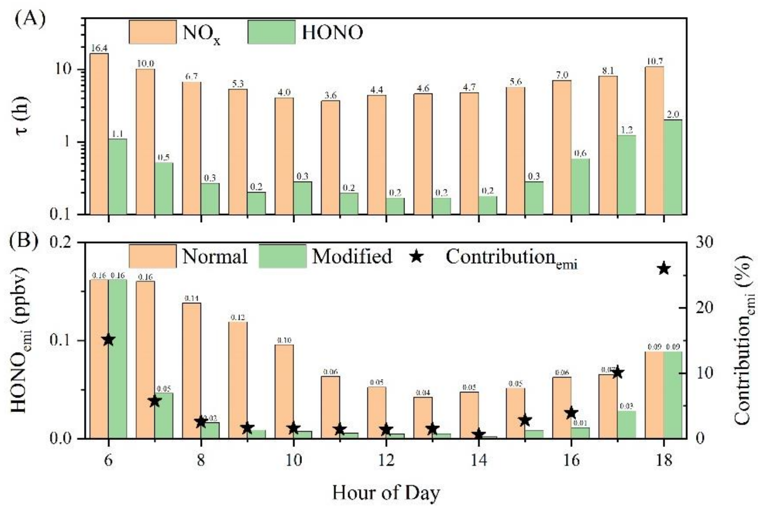


Figure S6: (A): Daytime lifetimes of HONO and NO_x; (B): $HONO_{emi}$ with a constant $\Delta HONO/\Delta NO_x$ ratios (Normal) or modified according to lifetimes (Modified) and the contribution of modified $HONO_{emi}$ to the observed HONO.

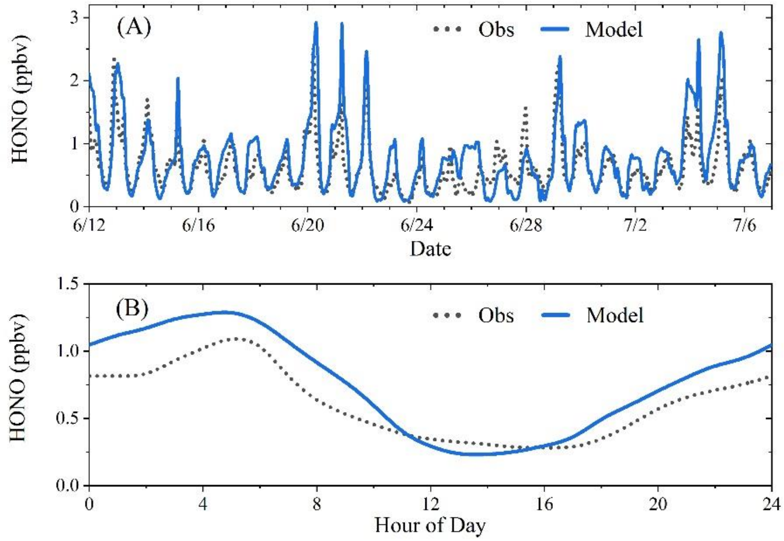


Figure S7: Modeled (Sce-3 with reduced γ_g and enlarged γ_a of 1.2×10^{-3}) HONO mixing ratios (Model, in blue) in comparison with observations (Obs, in black). (A): time series; (B): diurnal variations.

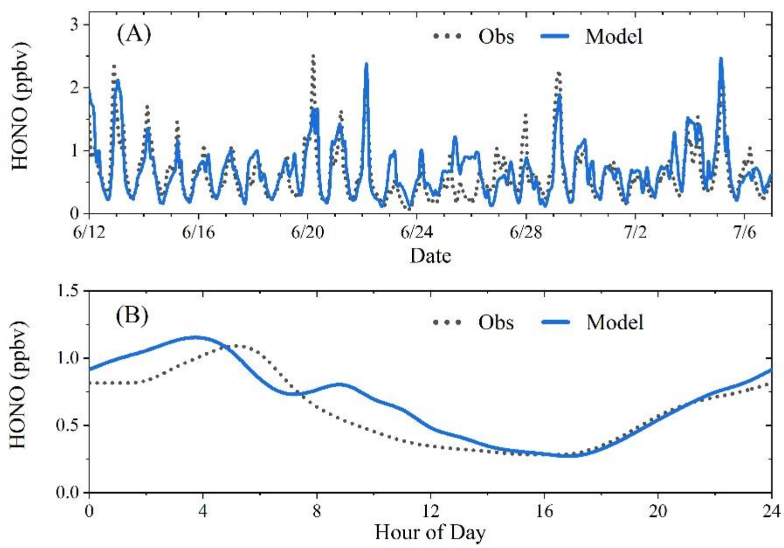


Figure S8: Modeled (Sce-3 with reduced γ_g and enlarged EF of 400) HONO mixing ratios (Model, in blue) in comparison with observations (Obs, in black). (A): time series; (B): diurnal variations.

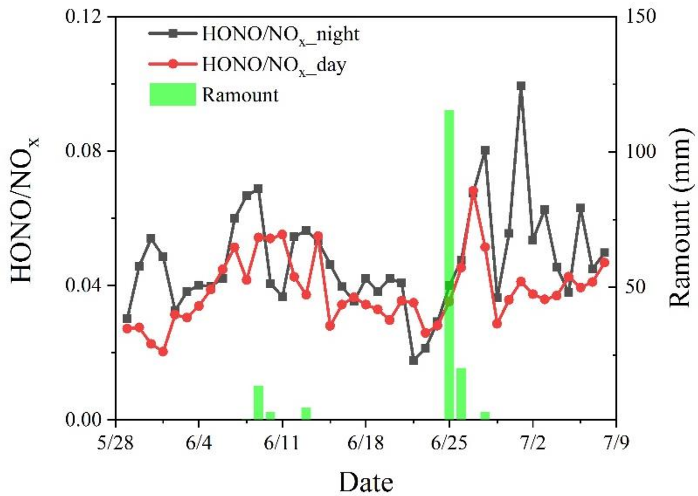


Figure S9: Daily rainfall amount (Ramount) and the daytime or the night-time HONO/NO$_x$.

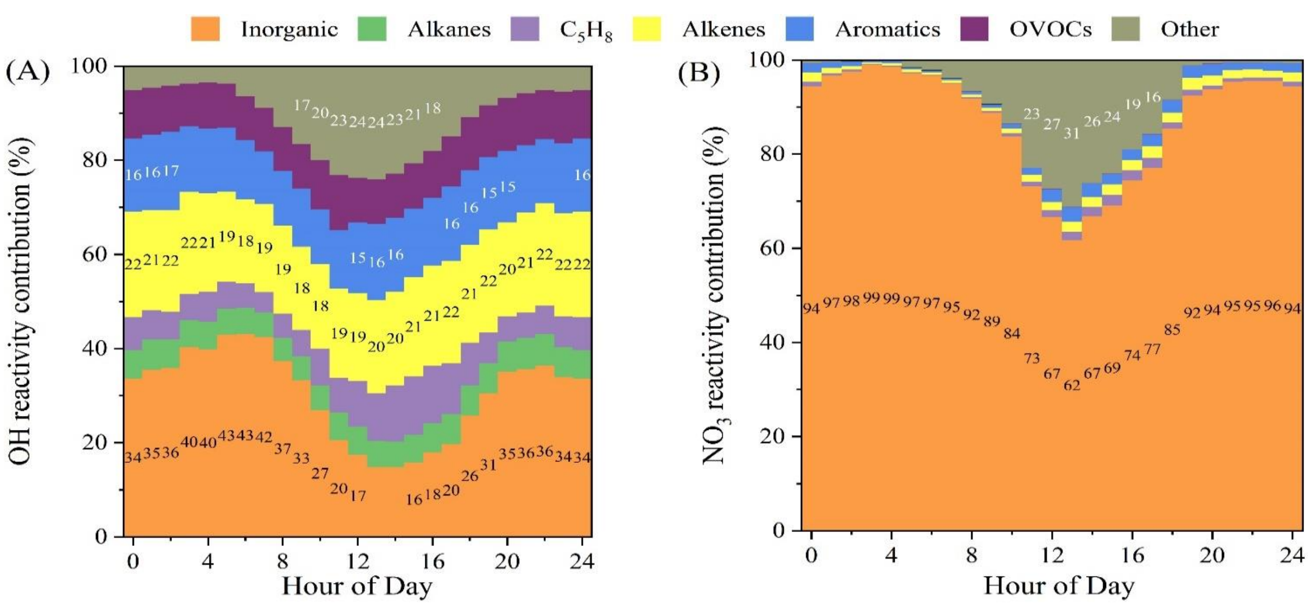


Figure S10: (A): OH and (B): NO$_3$ reactivity contributions. Reactivity with other unmeasured species was classified as "other". Note that alkenes here do not include C$_5$H$_8$.

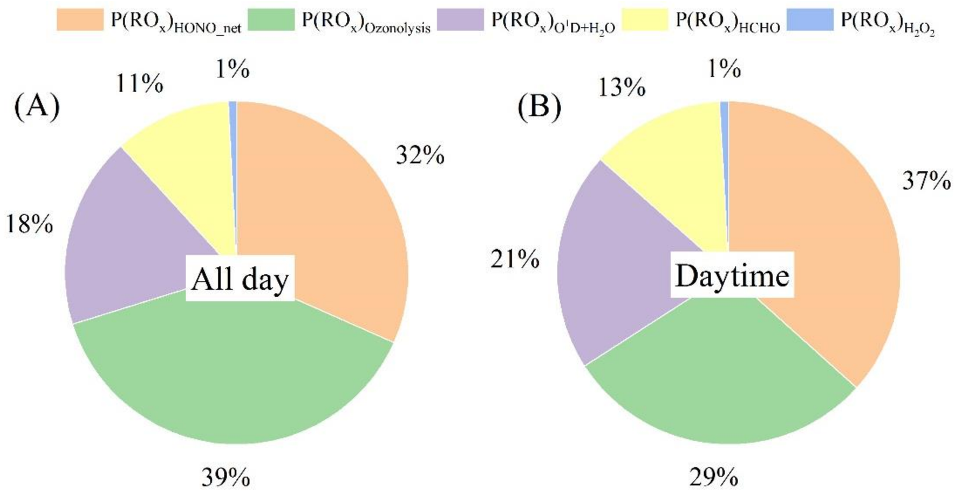


Figure S11: Relative contributions of different primary RO_x paths (A): throughout the whole day or (B): during the daytime.

References

Seinfeld, J. H. and Pandis, S. N.: Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, John Wiley & Sons., 2016.

Wolfe, G. M., Marvin, M. R., Roberts, S. J., Travis, K. R. and Liao, J.: The framework for 0-D atmospheric modeling (F0AM) v3.1, Geosci. Model Dev., 9(9), 3309–3319, doi:10.5194/gmd-9-3309-2016, 2016.