## Supplements

Table S 1: Additional simulations for sensitivity tests

No.	Simulation ID	HR: N <sub>2</sub> O <sub>5</sub>	HR: HO <sub>2</sub>	HR: RO <sub>2</sub>
1	FCTHR.ho2-10	х	×10	Х
2	FCTHR.ho2-5	х	$\times 5$	х
3	FCTHR.ho2-2	х	$\times 2$	х
4	FCTHR.ho2-1.5	х	×1.5	х
5	FCTHR.ho2-0.5	х	×0.5	х
6	FCTHR.n2o5-10	×10	х	х
7	FCTHR.n2o5-5	×5	х	х
8	FCTHR.n2o5-2	$\times 2$	х	х
9	FCTHR.n2o5-1.5	×1.5	х	х
10	FCTHR.n2o5-0.5	×0.5	х	х



Figure S1: Tropospheric column ozone (DU) in the Northern Hemisphere (first and second panels) and North Pacific (third panel) by OMI (black) and CHASER (red for STD; blue for noHR).



10 Figure S2: Seasonal and annual mean distributions of cloud droplet (left) and total aerosols SAD (right).



Figure S3: Observations and simulations for SO<sub>4</sub> and NO<sub>3</sub> mass concentrations at EANET stations. Legend: dotted-black, observed; red, CHASER (STD).



15 Figure S4: Observations and simulations for HNO<sub>3</sub>, NO<sub>x</sub>, and O<sub>3</sub> gaseous concentrations at EANET stations. Legend: dotted-black, observed; red, CHASER (STD).



Figure S5: Observations and simulations for PM<sub>2.5</sub> mass concentrations at EANET stations. Legend: dotted-black, observed; red, CHASER (STD).



Figure S6: Observations and simulations for PM<sub>2.5</sub> mass concentrations at EMEP stations. Legend: dotted-black, observed; red, CHASER (STD).



25 Figure S7: Observations and simulations for SO<sub>4</sub> and CO concentrations at EMEP stations. Legend: dotted-black, observed; red, CHASER (STD).



Figure S8: Observations and simulations for particulate NO<sub>3</sub><sup>-</sup> and gaseous HNO<sub>3</sub> concentrations at EMEP stations. Legend: dottedblack, observed; red, CHASER (STD).



Figure S9: Observations and simulations for NO<sub>x</sub> concentrations at EMEP stations. Legend: dotted-black, observed; red, CHASER (STD).



Figure S10: Observations and simulations for O<sub>3</sub> concentrations at EMEP stations. Legend: dotted-black, observed; red, CHASER 35 (STD).



Figure S10: (cont.)



Figure S11: Observations and simulations for NO<sub>2</sub>, OH, O<sub>3</sub>, and CO levels during ATom1. Legend: black, ATom1; red, CHASER 40 (STD).



Figure S12: Model correlations and biases of STD run with EANET (upper row) and EMEP (lower row) observations for aerosol and gaseous concentrations. 3 sigma-rule outlier detection is applied for each station before calculating all data. For NO<sub>x</sub>, all data was filtered once more time by the 2 sigma-rule. *N* is the number of available stations.



Figure S13: Correlations and bias of STD run with MIRAI's CO and O<sub>3</sub> data.





Figure S14: Correlation between CHASER (STD) and ATOM1 for NO<sub>2</sub>, OH, CO, and O<sub>3</sub>. First, second, and third rows respectively show data from all flight altitudes, lower troposphere (P > 600 hPa), and the North Pacific (140–240 E, 40–60 N, P > 600 hPa). Two sigma-rule outlier detection was used for observational NO<sub>2</sub> and OH; a 3 sigma-rule was applied for CO and O<sub>3</sub>.



Figure S15: Effects of HRs(Clouds) (a-b) and HRs(Aerosols) (c-d) in zonal mean and at the surface. The contour interval is 2% in the plots for zonal mean.



Figure S16: Effects of HR(N<sub>2</sub>O<sub>5</sub>) in zonal mean calculated by FCTHR\_n2o5-10 run (a) and effects of HR(HO<sub>2</sub>) at the surface calculated by FCTHR\_ho2-10 run (b)