Supplement of

Ozone formation sensitivity based on the secondary formaldehyde to nitrogen dioxide ratio (FNR_{sec}) derived from ground-based remote sensing measurements and chemical transport model

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Table S1: The Pandora stations used in this study

Location name	Latitude (deg)	Longitude (deg)	Instrument number	Location altitude (m)	Local Operator
Sapporo	43.0727	141.3459	P196	46	Masatomo Fujiwara
Tsukuba-NIES	36.0513	140.1210	P176	45	Tamaki Fujinawa
Tokyo-TMU	35.6200	139.3834	P194	135	Shungo Kato
Fukuoka	33.5491	130.3660	P199	55	Hisahiro Takashima

Table S2: Emission inventories employed in the GEOS-Chem model simulations

Source	Inventory	Resolution	Reference
Anthropogenic			
+ Global	CEDSv2	0.5° × 0.5°	McDuffie et al. (2020)
+ Japan	REASv3.2.1	0.25° × 0.25°	Kurokawa and Ohara (2020)
Aircraft	AEIC 2019	1° × 1°	Simone et al. (2013)
Biomass burning	GFED4	0.25° × 0.25°	van der Werf et al. (2017)
Lightning	-	0.5° × 0.625°	Murray et al. (2012)
Off-line biogenic VOCs, Soil NO_x, sea salt aerosol	-	0.5° × 0.625°	Weng et al. (2020)
Off-line dust	-	0.5° × 0.625°	Meng et al. (2021)
Shipping	CEDSv2	0.5° × 0.5°	McDuffie et al. (2020)

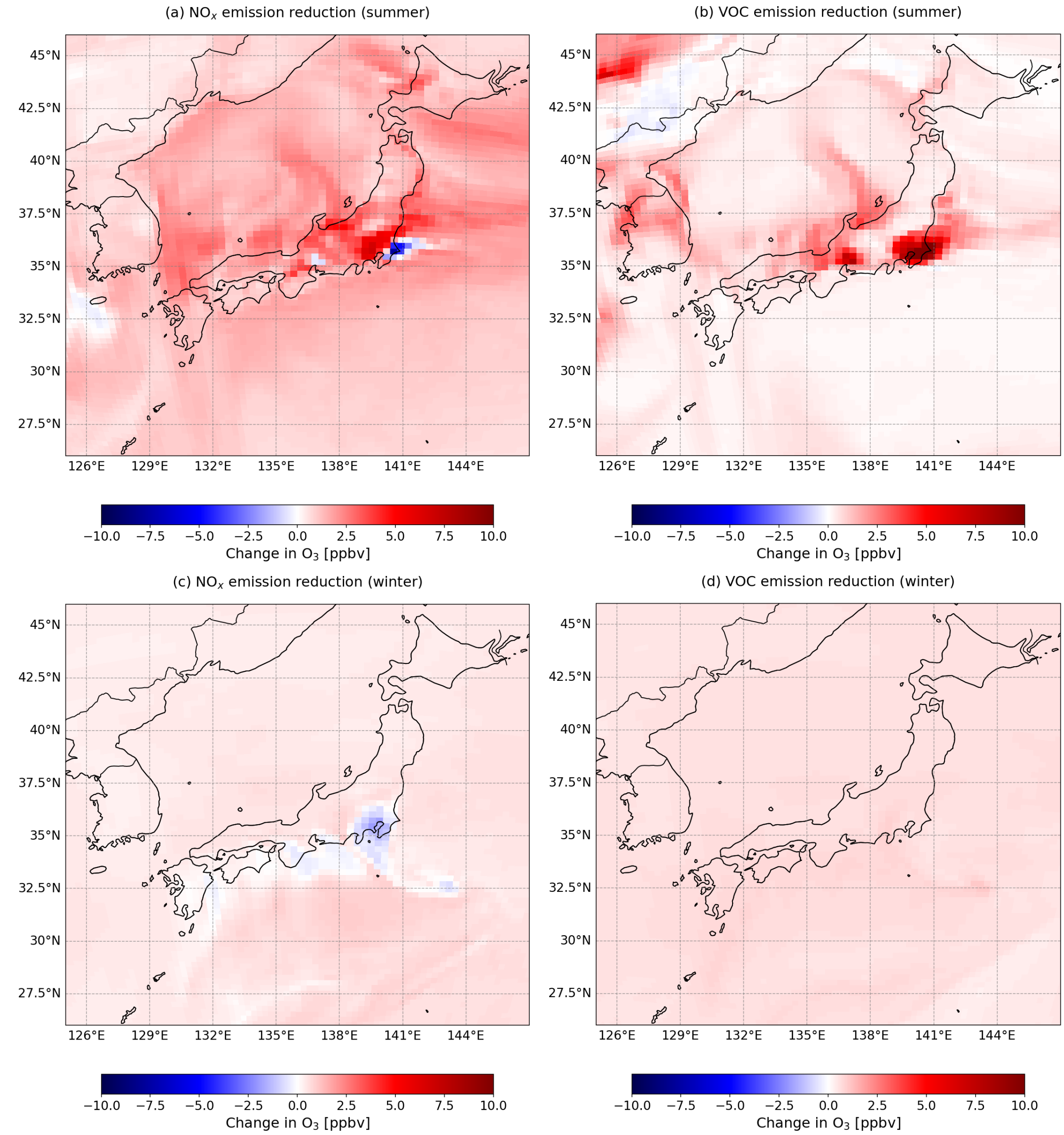


Figure S1: GEOS-Chem model simulations of changes in the surface ozone concentration at 13:00 in response to NO$_x$ emission reduction (a, c) and VOC emission reduction (b, d). The top panels present changes in summer, while the bottom panels present changes in winter. Red and blue colors indicate positive and negative change in the surface ozone concentration, respectively.

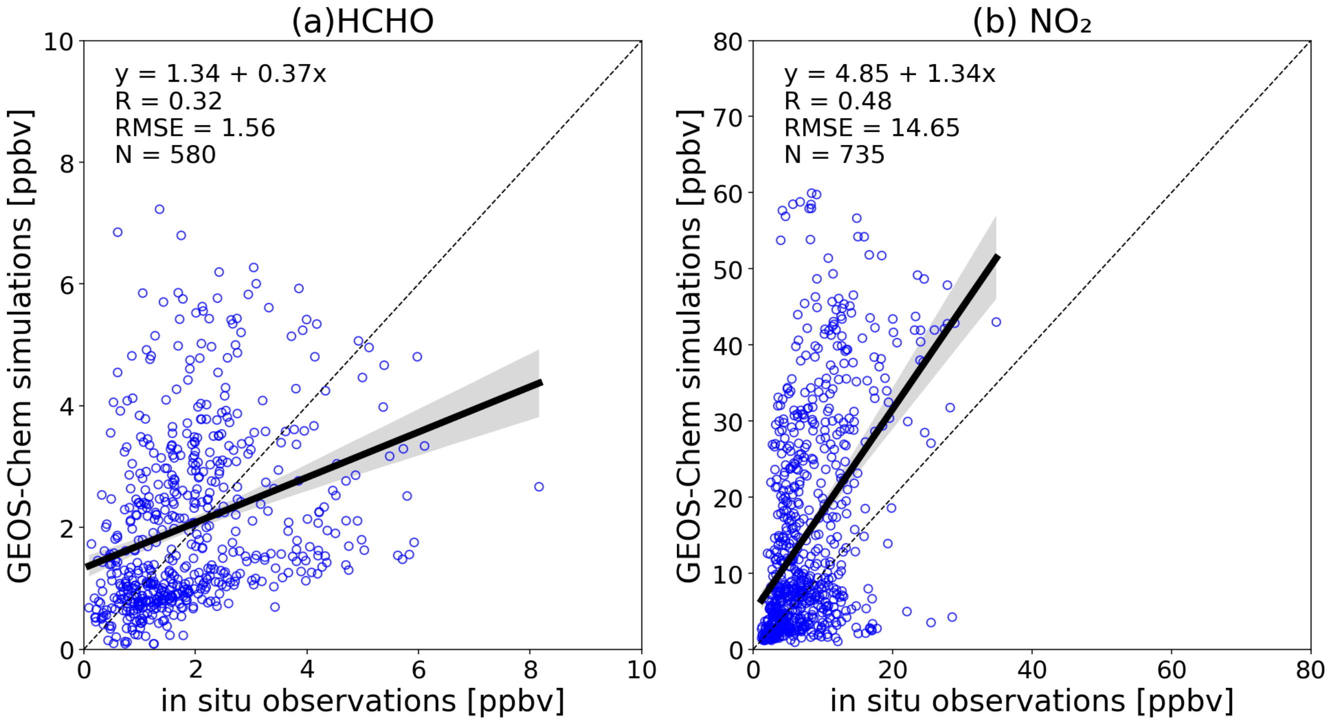


Figure S2: Scatter plots of the surface HCHO (a) and NO_2 (b) comparing in situ measurements with GEOS-Chem model simulation at Tokyo-TMU. The y-axis represents GEOS-Chem simulations, and the x-axis represents in situ observations. The dashed line indicates the 1:1 line.

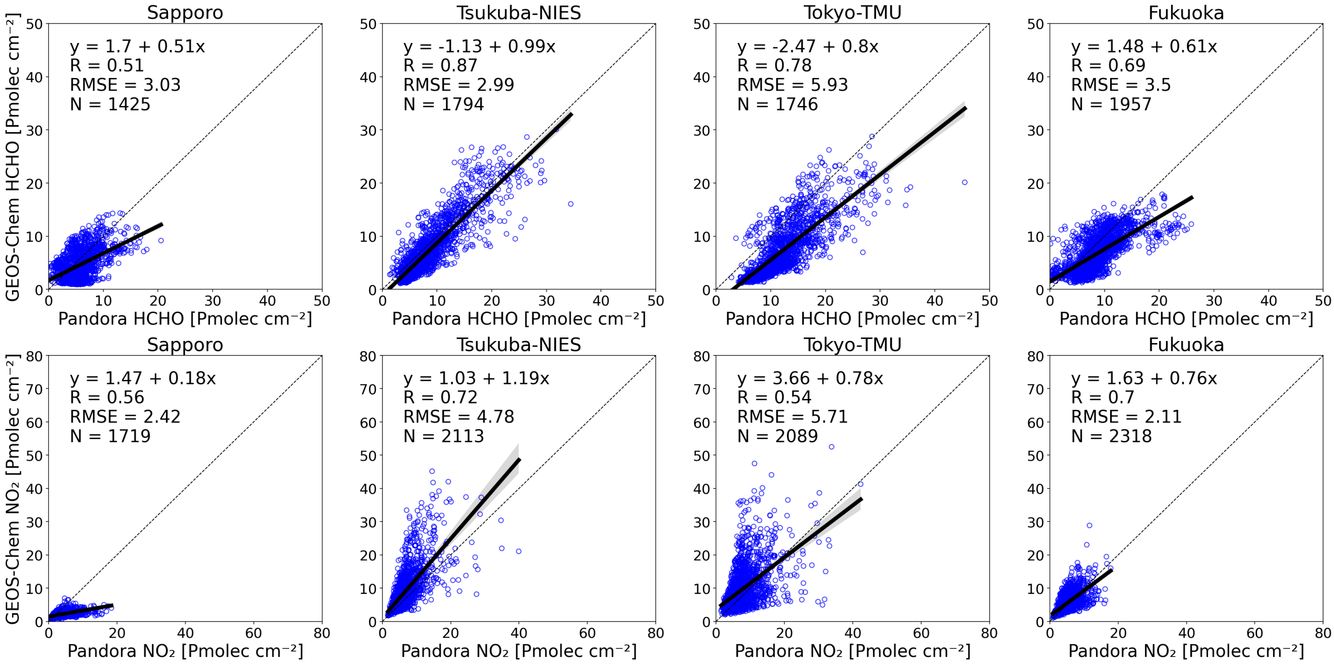


Figure S3: Scatter plots of tropospheric vertical column densities of HCHO (top panels) and NO_2 (bottom panels) between the Pandora direct-sun measurements and GEOS-Chem model simulations. The dashed line indicates the 1:1 line.

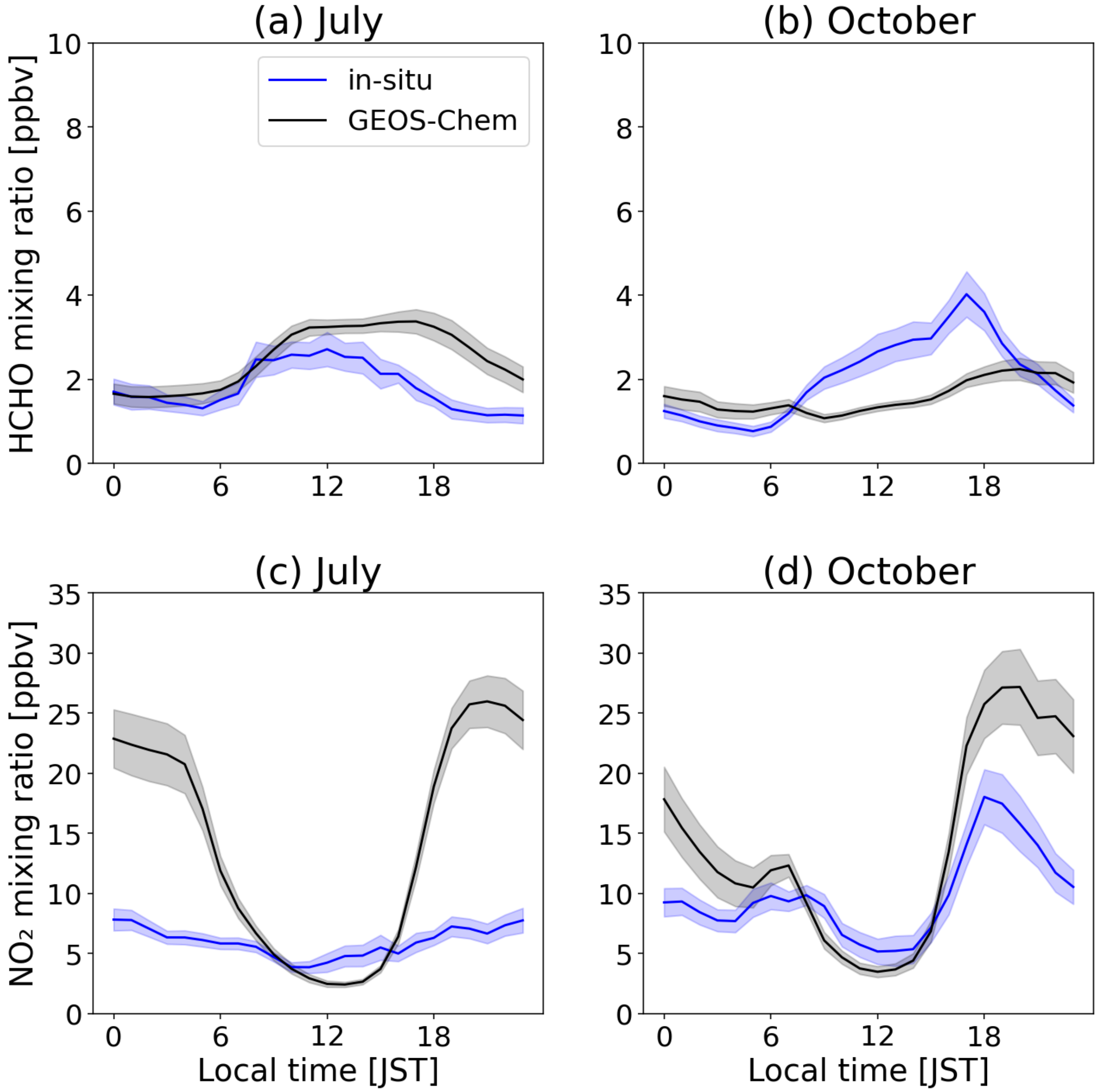


Figure S4: Diurnal cycle of the surface HCHO (top panels) and NO_2 (bottom panels) obtained from in situ measurements (blue) and GEOS-Chem simulations (black) at the Tokyo-TMU site in July (a and c) and in October (b and d) 2022. Shaded error bands indicate ±1 standard error.

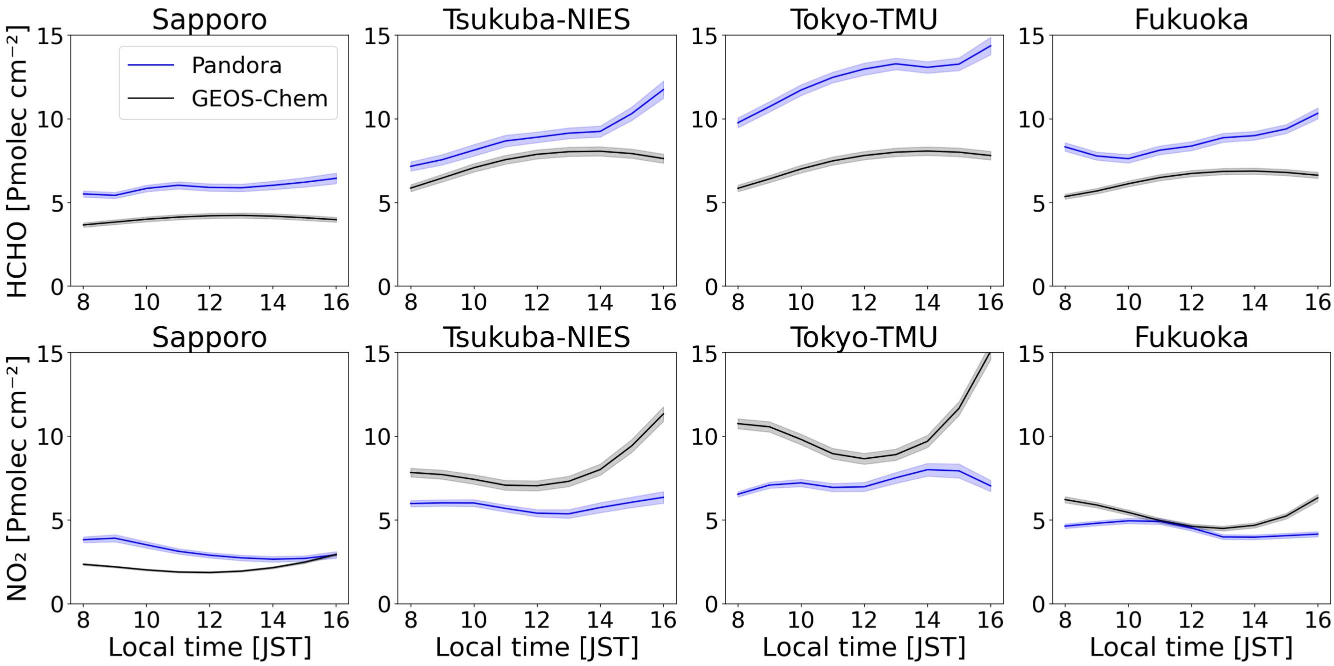


Figure S5: Diurnal variation of annual tropospheric vertical column densities of HCHO and NO_2 obtained from Pandora (blue) and GEOS-Chem model simulation (black). Shaded error bands indicate ±1 standard error.

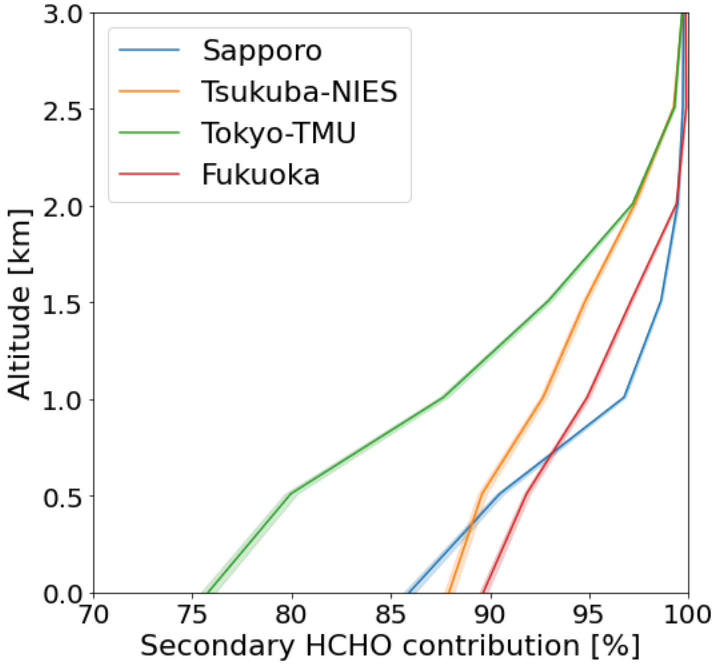
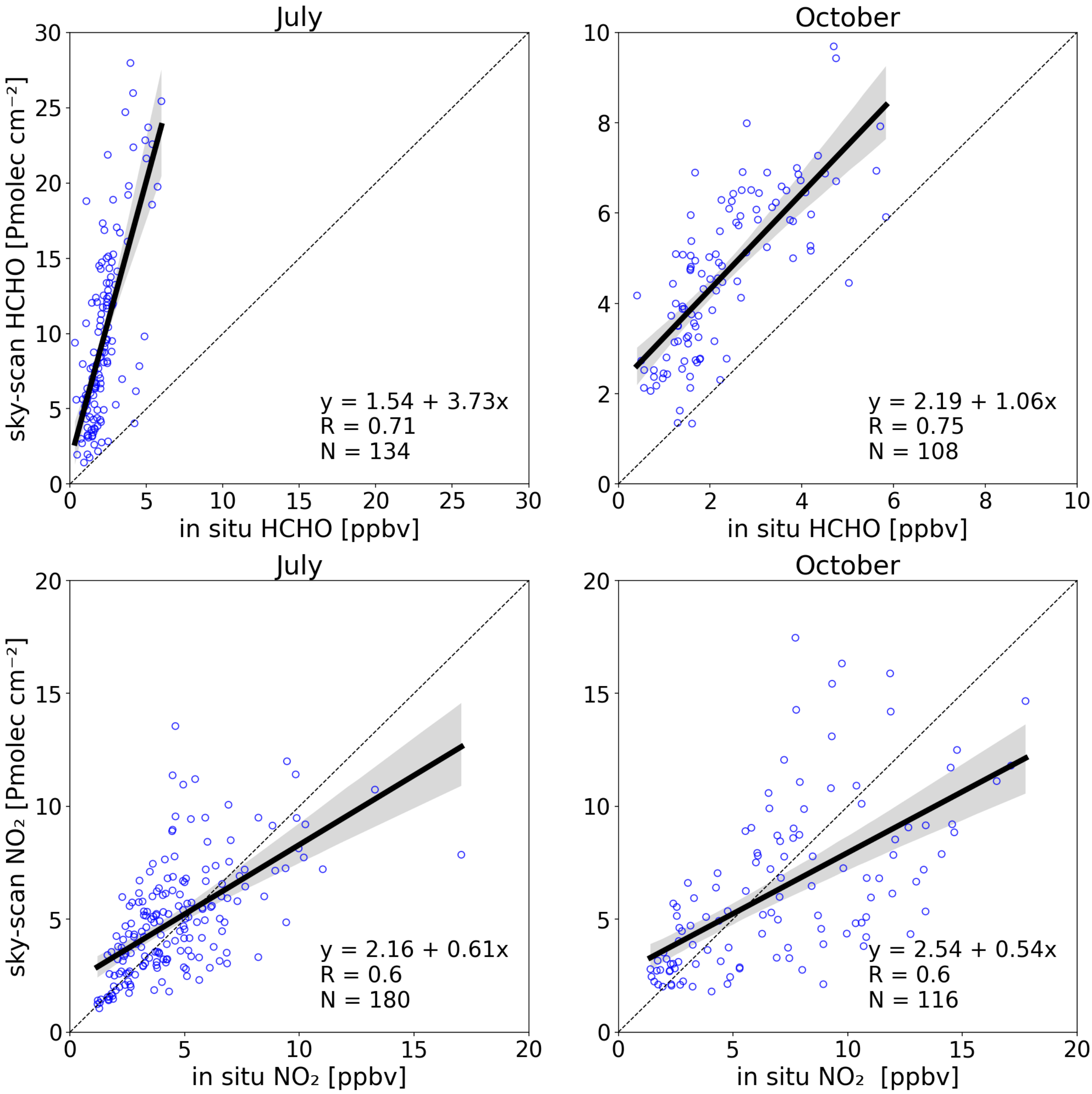


Figure S6: Vertical contributions of secondary HCHO (annual mean) at the study JPN sites obtained from GEOS-Chem model. Shaded error bands indicate ±1 standard error.



45 **Figure S7: Relationship in HCHO (top panels) and NO_2 (bottom panels) between the lower tropospheric column (Pandora sky-scan) and the surface concentrations (in situ measurements). The dashed line indicates the 1:1 line.**

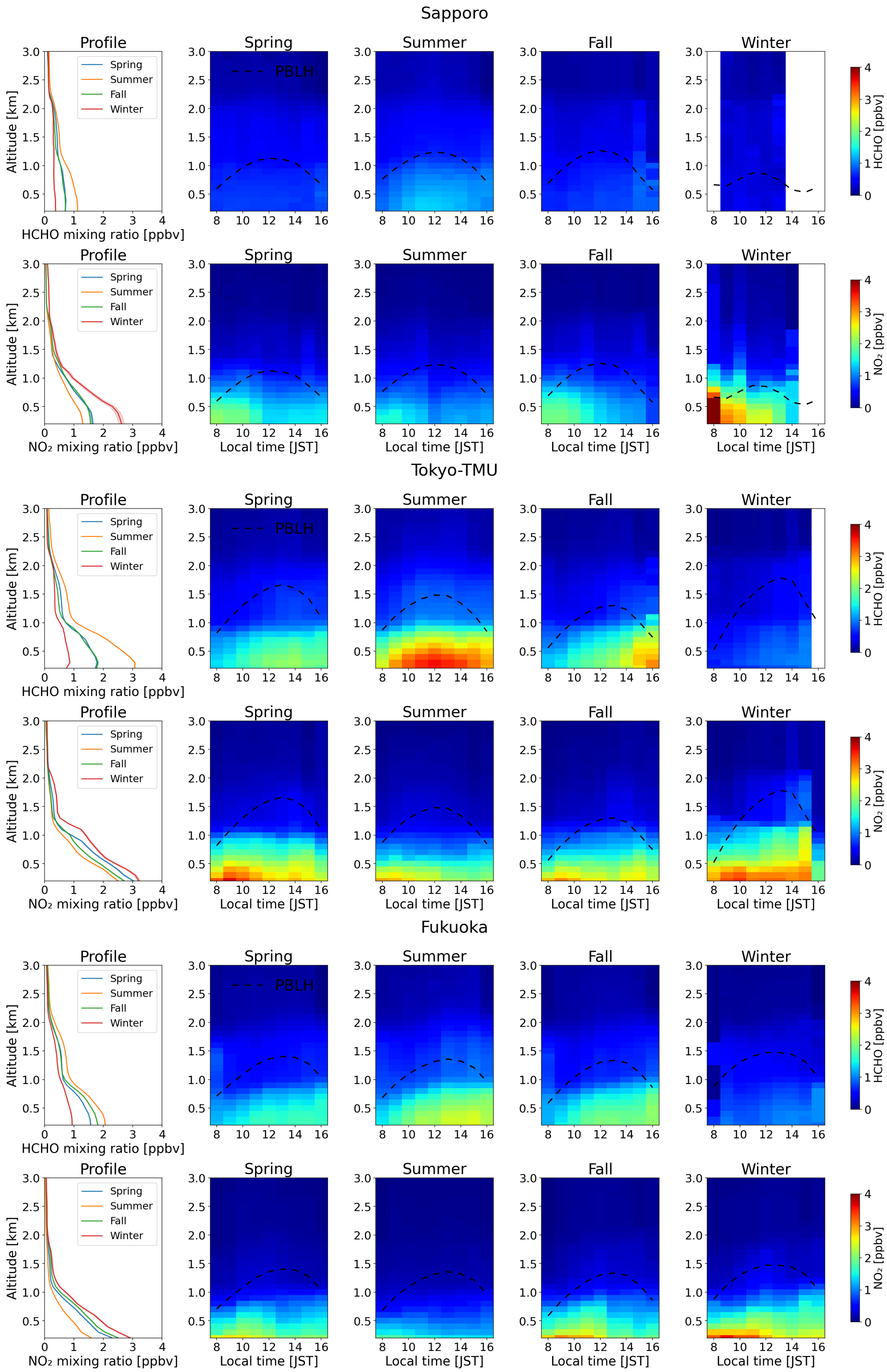


Figure S8: Vertical profiles of HCHO (top panels) and NO_2 (bottom panels) derived from the Pandora sky-scan mode at Sapporo, Tokyo-TMU, and Fukuoka. The leftmost panels show the seasonal vertical profiles, with shaded error bands indicating ±1 standard error. Color bars exhibit the diurnal profiles. Black dashed curves present planetary boundary layer height (PBLH).

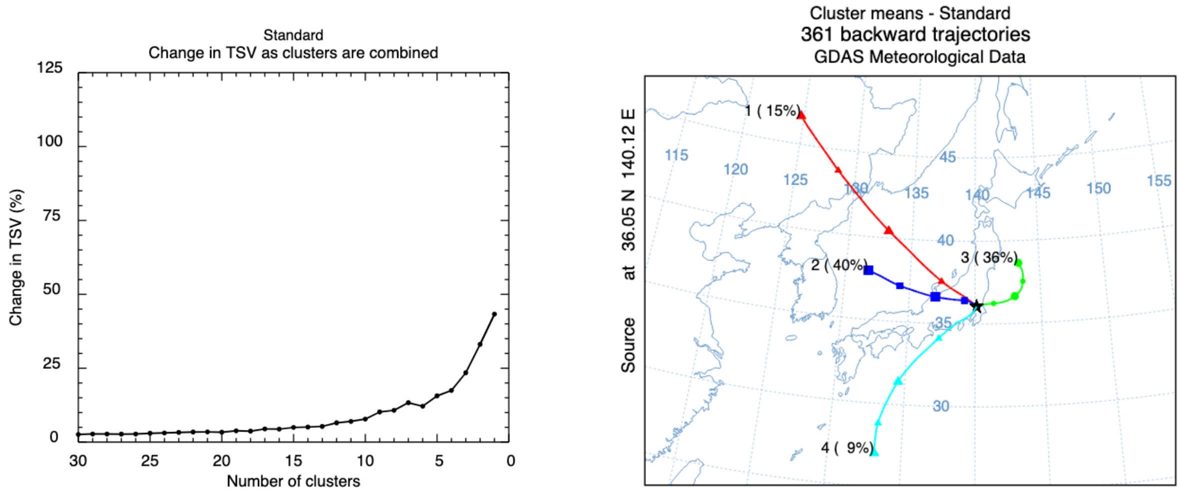


Figure S9: Cluster analysis based on the change in total spatial variance (TSV) (left panel) and cluster means (right panel) of one-year backward trajectories arriving at 13:00 at Tsukuba-NIES site using HYSPLIT model. Four clusters include c1 (red), c2 (blue), c3 (green), and c4 (cyan).

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