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Causes of growing middle-to-upper tropospheric ozone over the northwest Pacific region

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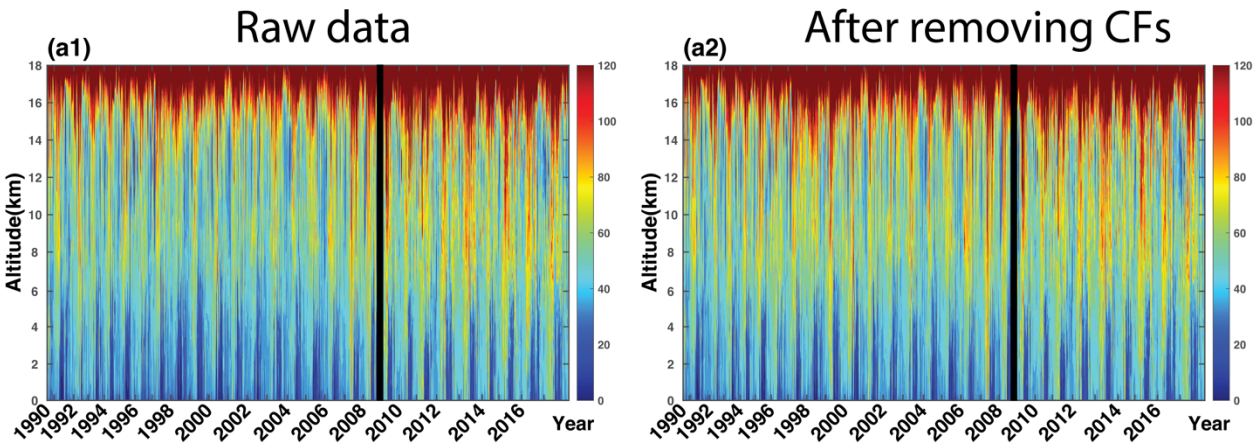

Figure S1. All O_3 profile samples were used in the analysis at Naha from (a1) WOUDC and (a2) after removing CFs. Black lines indicate the transition time from the CI to ECC ozonesonde at the Japanese stations around 2009.

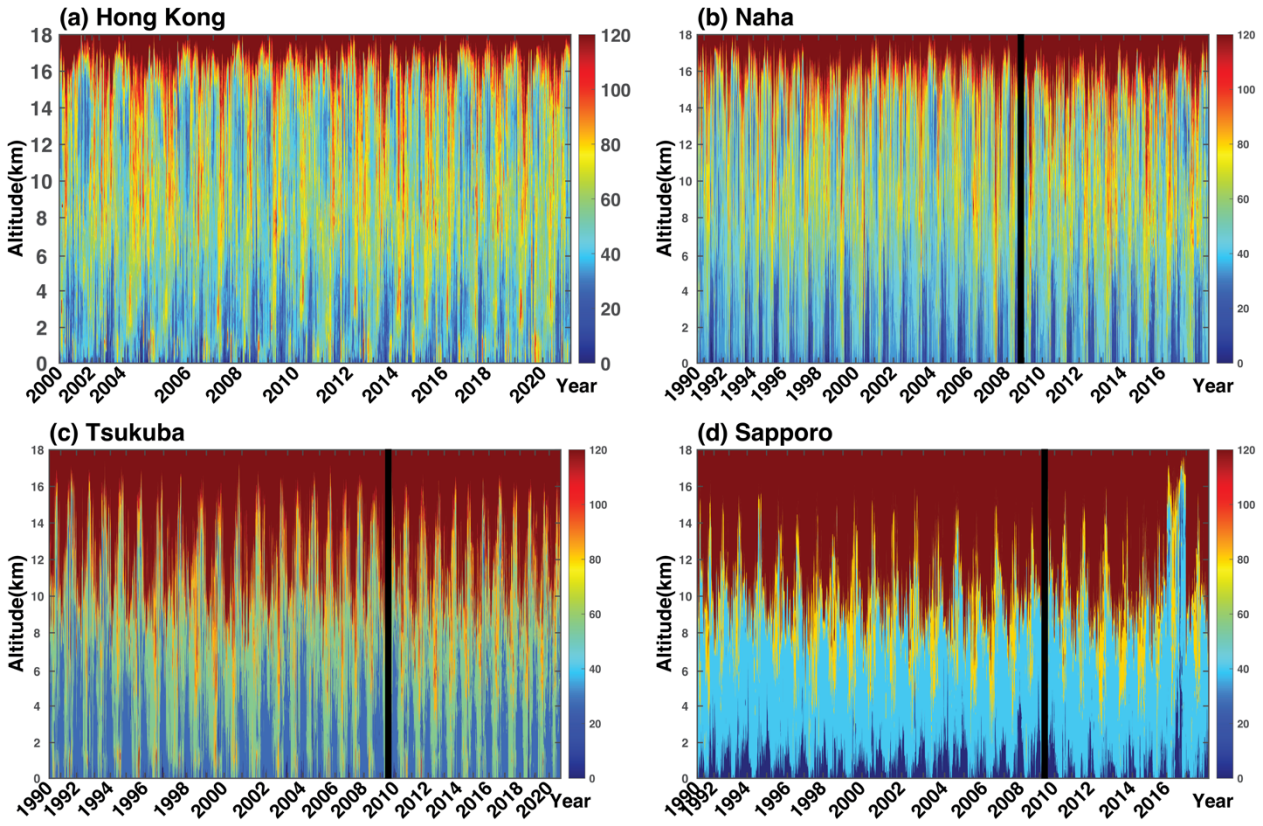

Figure S2 All O_3 profile samples used in the analysis at (a) Hong Kong, (b) Naha, (c) Tsukuba, and (d) Sapporo. Black lines indicate the transition time from CI to ECC ozonesonde at the Japanese stations around 2009.

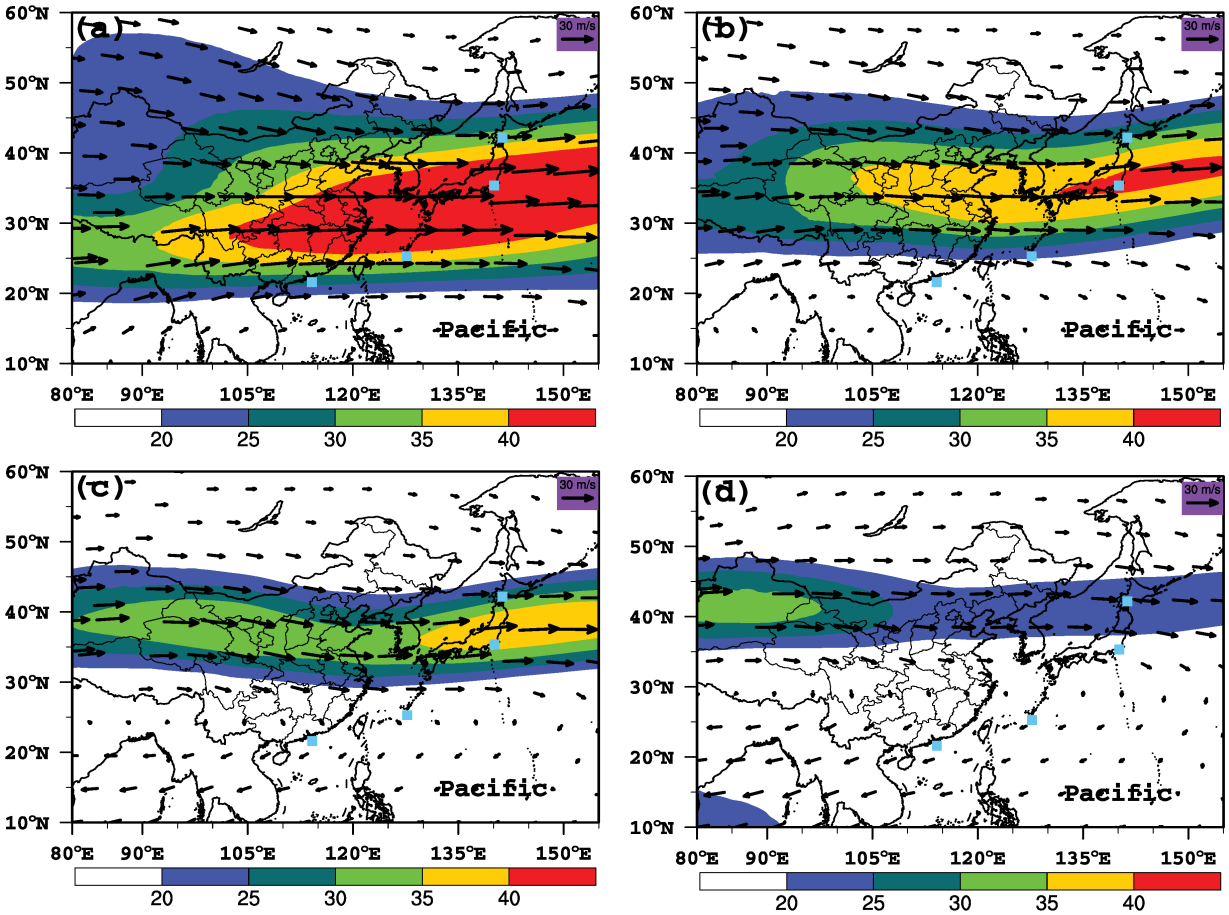

Figure S3. The wind field (vector) and wind speed (color shades) retrieved from ERA5 (the fifth generation ECMWF reanalysis) at 200hPa in (a) April, (b) May, (c) June, and (d) July averaged over 1990-2020. Four O_3-sounding sites are indicated in the blue squares.

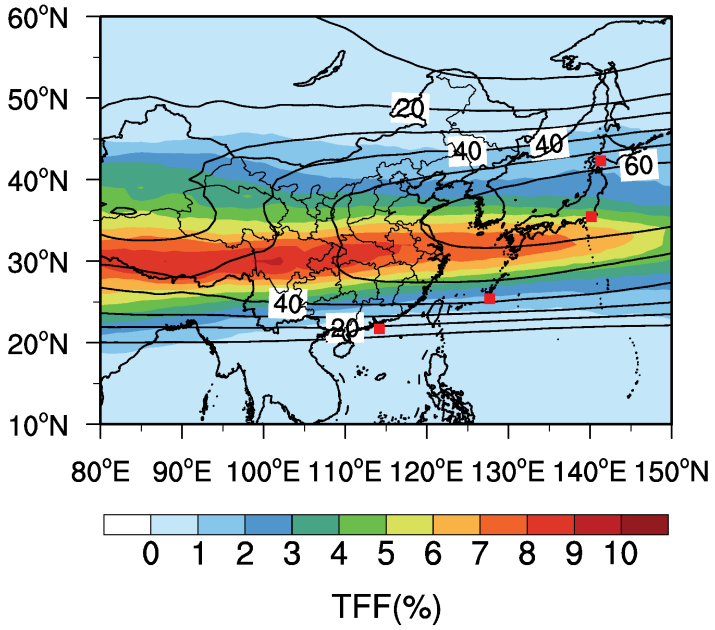

Figure S4 Climatological distribution of tropopause folding frequency (shaded color) and jet frequency (contour lines, units: %) during 2000-2018, products provided by ETH Zurich. Four O3-sounding sites are indicated with the red squares.

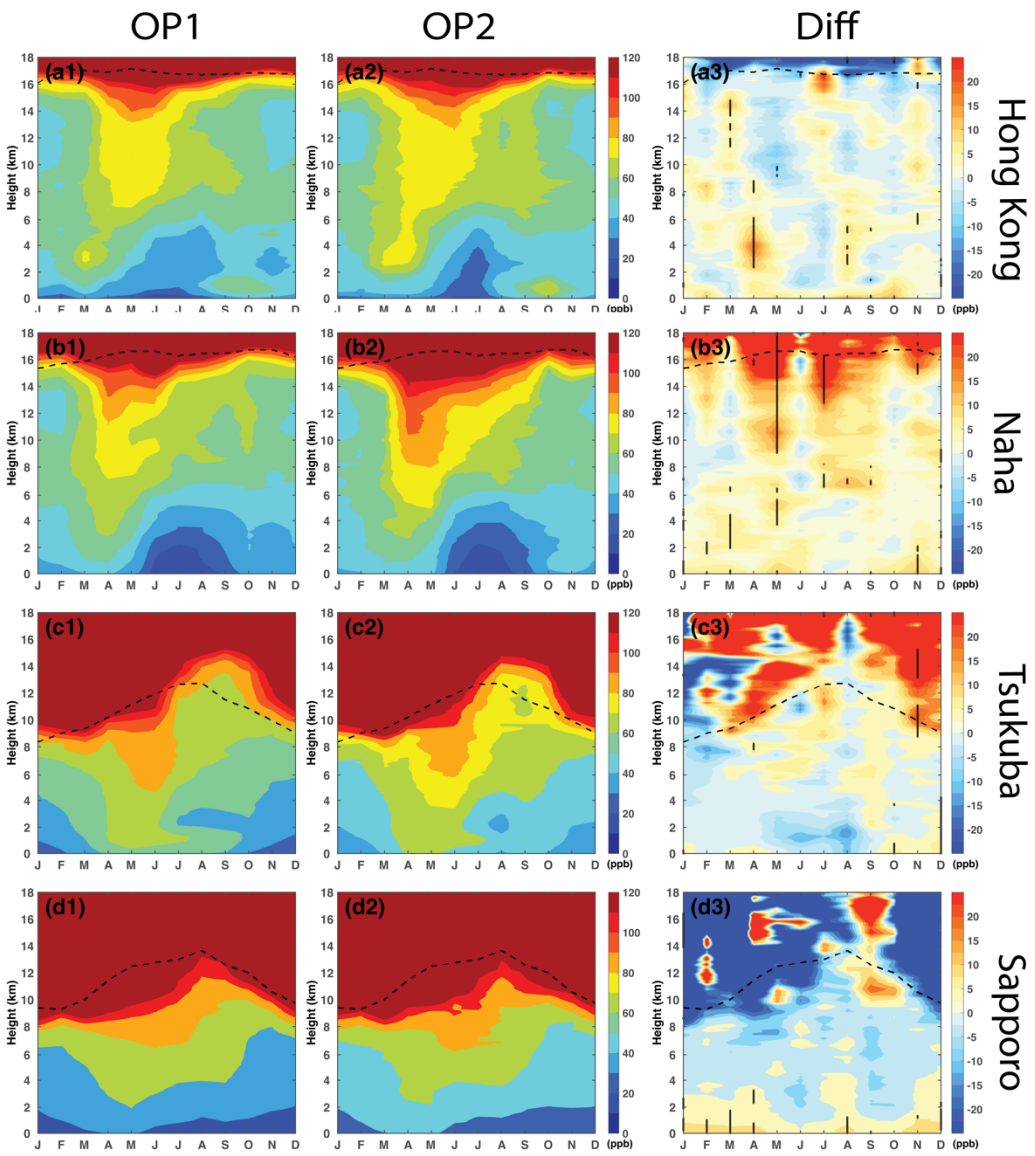

Figure S5. Monthly evolution of the vertical distribution of mean O_3 in the first overlapping period (OP1: 2000-2008), the last overlapping period (OP2: 2009-2017), and the difference between OP2 and OP1 of O_3 at four observation sites (a1-a3) Hong Kong, (b1-b3) Naha, (c1-c3) Tsukuba and (d1-d3) Sapporo. Black dash lines indicate tropopause height. Dots in the i-l represent the layer with statistically significant changes according to a paired two-sided t-test ($p < 0.05$).

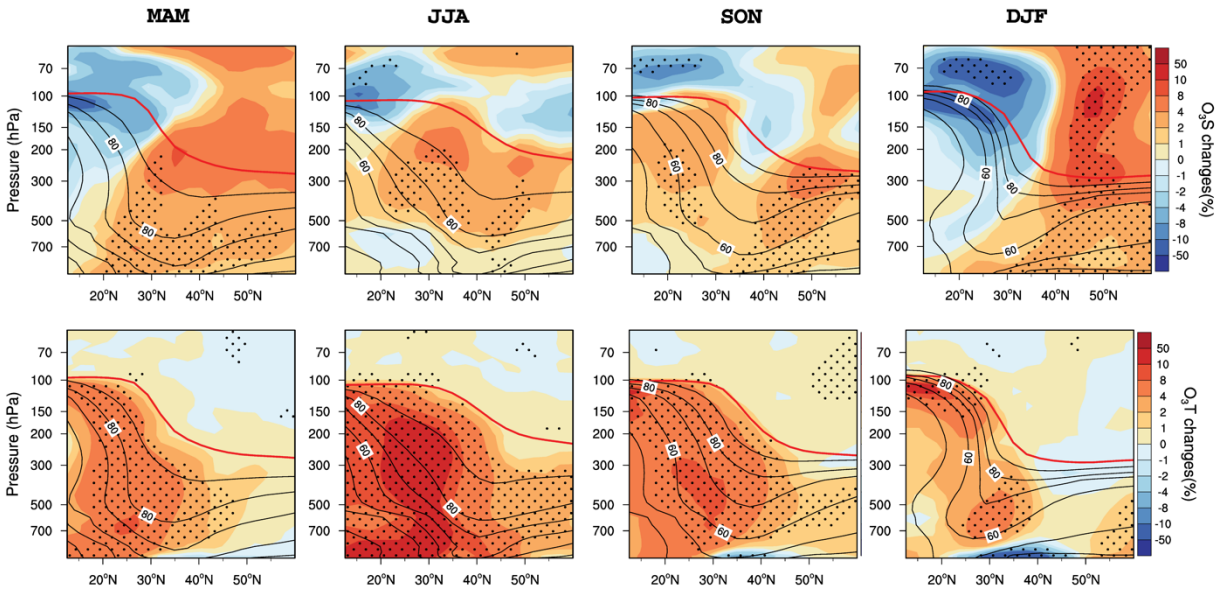

Figure S6. Latitude-pressure cross sections of difference of O_3S, and O_3T relative to climatological O_3 (%) between the 2010s and 1990s along the Northwest Pacific region (zonal mean over 110°E to 150°E) in four seasons. Black lines indicate the climatological distribution of O_3S, and O_3T, respectively. Red solid lines denote the tropopause height. Dots represent the layer with statistically significant changes according to a paired two-sided t-test ($p < 0.05$).

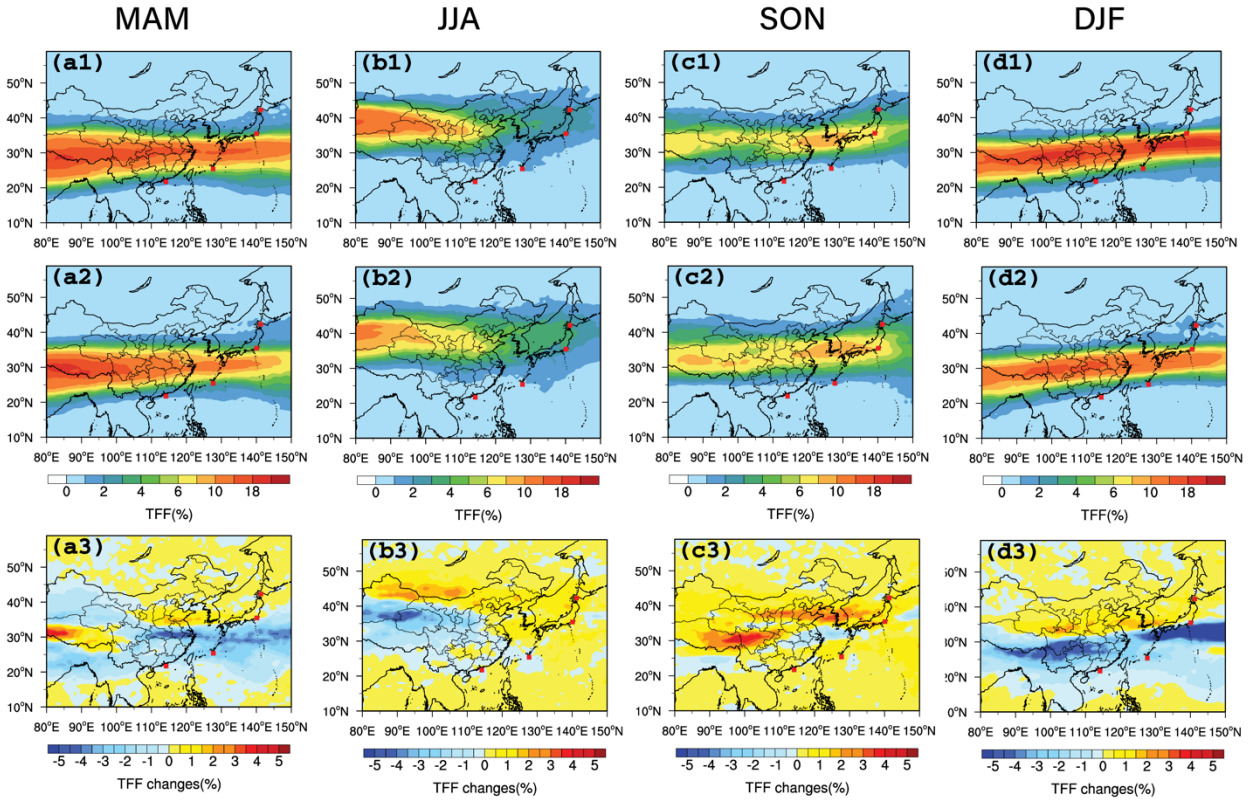

Figure S7. Distribution of tropopause folding frequency, a product provided by ETH Zurich, during the 1990s (a1-d1), 2010s (a2-d2) and its changes (a3-d3) at (a) spring, (b) summer, (c) autumn and (d) winter.