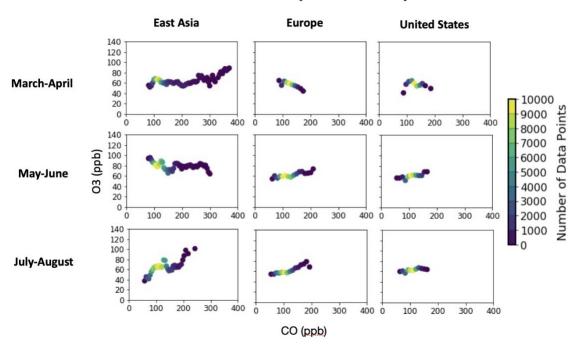
## Supplementary Materials for

# "Large and increasing stratospheric contribution to tropospheric ozone over East Asia"

Nadia K. Colombi<sup>1\*</sup>, Daniel J. Jacob<sup>1,2</sup>, Xingpei Ye<sup>3</sup>, Robert M. Yantosca<sup>2</sup>, Kelvin H. Bates<sup>4</sup>, Drew C. Pendergrass<sup>2</sup>, Laura Hyesung Yang<sup>2</sup>, Ke Li<sup>5</sup>, and Hong Liao<sup>5</sup>

### O3 vs CO (350-750 hPa)



**Figure S1** IAGOS ozone versus CO concentrations in the free troposphere (750-350 hPa) over East Asia (30.0-45.0 ° N, 109.0-145.0 ° E), Europe (35.0-54.0 ° N, -11.0-30.0 ° E), and the United States (28.0-49.0 ° N, -124.6.0- -66.8 ° E) for March-August 2015-2019. Ozone and CO column mixing ratios are computed from individual vertical profiles averaged over 25 hPa pressure levels. Ozone column mixing ratios are then averaged over bins of 5 ppb CO. Only bins with 50 or more observations are included.

<sup>&</sup>lt;sup>1</sup>Harvard University, Department of Earth and Planetary Science, Cambridge, MA 02138, USA

<sup>&</sup>lt;sup>2</sup>Harvard University, John A. Paulson School of Engineering and Applied Sciences, Cambridge, MA 02138, USA

<sup>&</sup>lt;sup>3</sup>Peking University, Department of Atmospheric and Oceanic Science, Beijing 100084, China

<sup>&</sup>lt;sup>4</sup>University of Colorado, Department of Mechanical Engineering, Boulder, CO 80309, USA

<sup>&</sup>lt;sup>5</sup>Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control, Jiangsu Collaborative Innovation Center of Atmospheric Environment and Equipment Technology, School of Environmental Science and Engineering, Nanjing University of Information Science & Technology, Nanjing 210044, China

<sup>\*</sup>Correspondence to: Nadia Colombi (ncolombi@g.harvard.edu)

#### Stratospheric ozone at the surface: Difference between Oy vs Ox tagging method

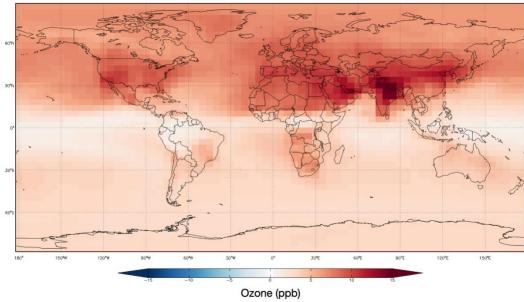
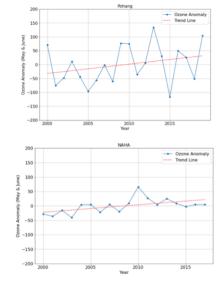
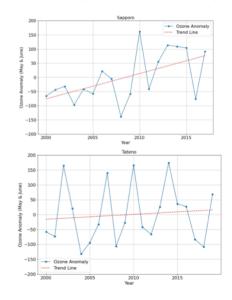


Figure S2 Stratospheric contribution to surface ozone for May 2016 using the expanded odd oxygen family to include hydrogen oxide radicals produced from ozone photolysis as in Bates and Jacob (2020).

# Anomalies in lower stratospheric ozone (150-80 hPa)





**Figure S3** Anomalies in lower stratospheric (150-80 hPa) ozone relative to the 2000-2019 May-June mean from four ozonesonde stations in East Asia. Data is obtained from the World Ozone an Ultraviolet Radiation Data Centre (https://woudc.org/data/explore.php?lang=en).

#### References

[1] Bates, K. H. and Jacob, D. J.: An Expanded Definition of the Odd Oxygen Family for Tropospheric Ozone Budgets: Implications for Ozone Lifetime and Stratospheric Influence, Geophysical Research Letters, 47, e2019GL084486, <a href="https://doi.org/10.1029/2019GL084486">https://doi.org/10.1029/2019GL084486</a>, 2020.