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*Supplement of*

## **Modeling stratospheric intrusion and trans-Pacific transport on tropospheric ozone using hemispheric CMAQ during April 2010 – Part 1: Model evaluation and air mass characterization for stratosphere–troposphere transport**

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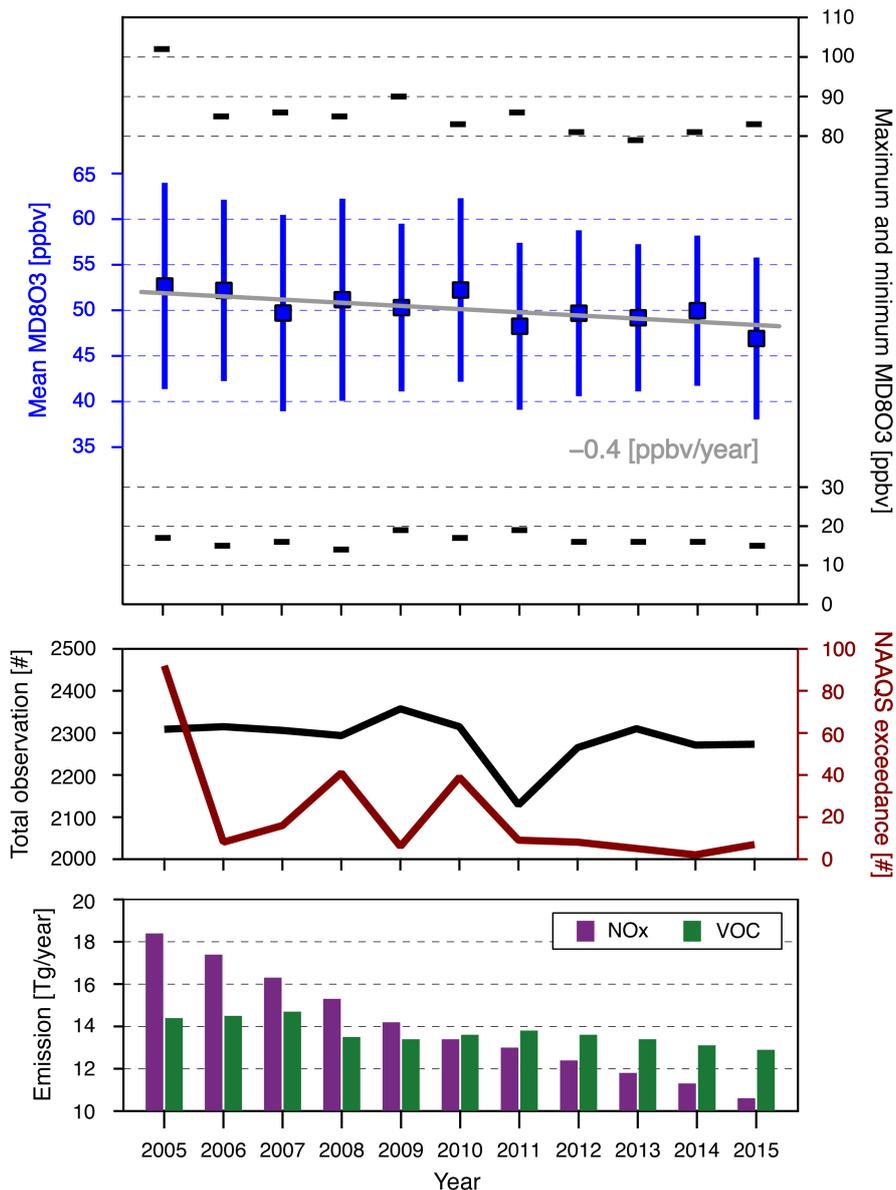
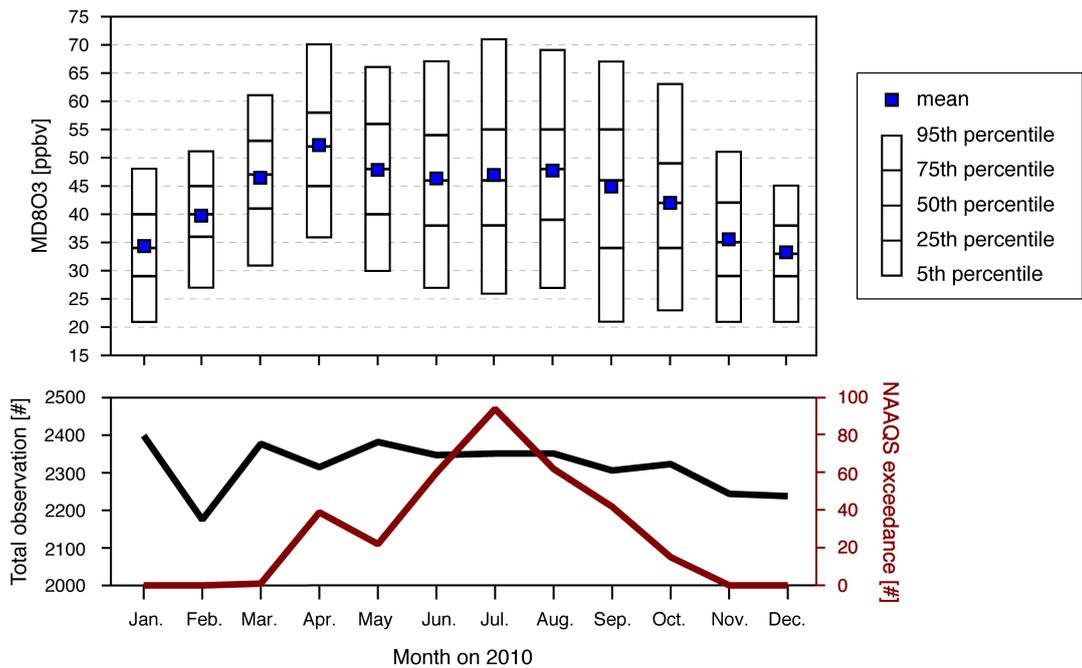
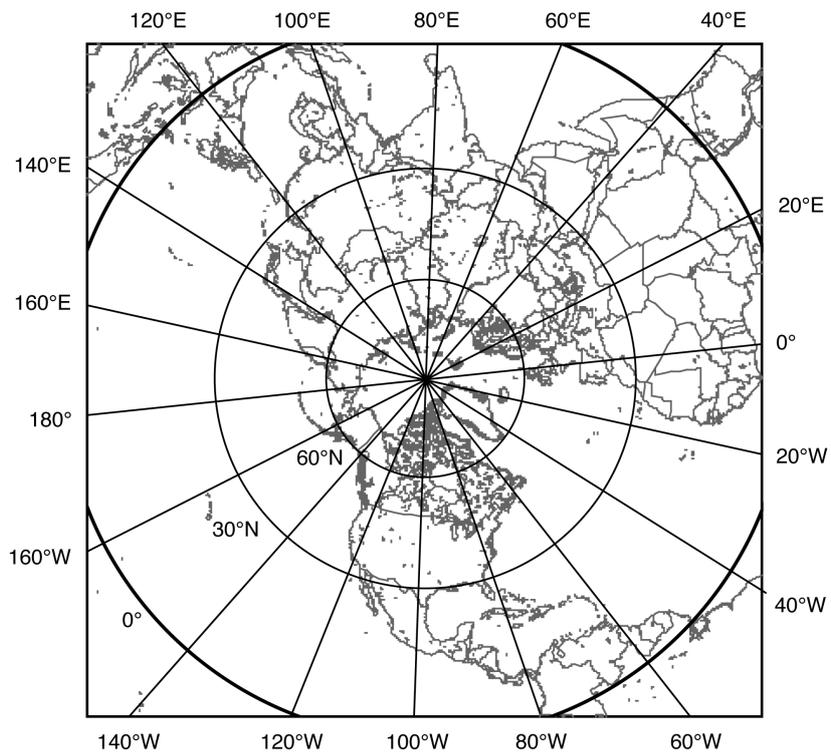


Figure S1. Long-term trends from 2005 to 2015 as before and after 5 years comparison to 2010. (Top) Mean (blue color, left-axis) and maximum and minimum (black color, right-axis) MD8O3 on April. (Center) Number of total observations (black color, left-axis) and exceedance of NAAQS (dark red color, right-axis; 75 ppbv is used as a criterion as 2010) on April. (Bottom) Annual NO<sub>x</sub> (purple) and VOCs (green) emissions in the U.S.A. except wildfire (<https://www.epa.gov/air-emissions-inventories/air-pollutant-emissions-trends-data>).



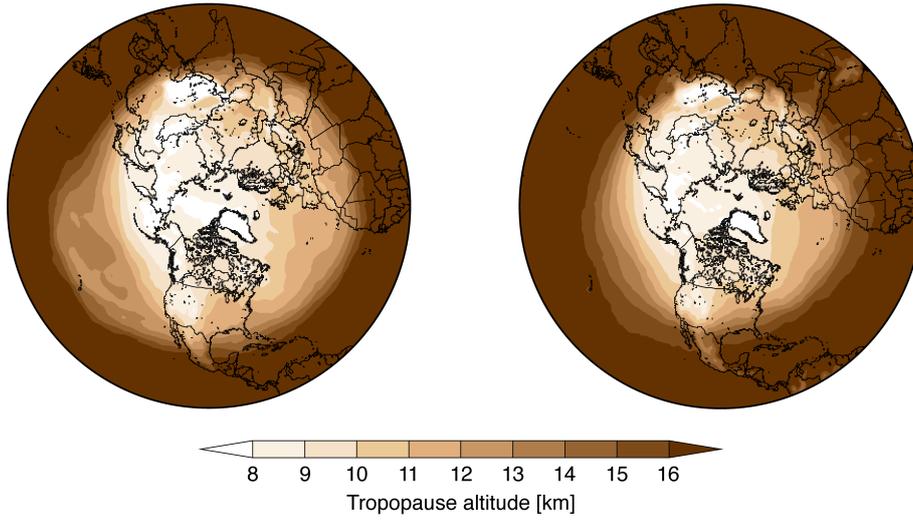
**Figure S2. (Top) Monthly mean and percentiles of MD8O3 on 2010. (Bottom) Number of total observations (black color, left-axis) and exceedance of NAAQS (dark red color, right-axis; 75 ppbv is used as a criterion as 2010) on 2010.**



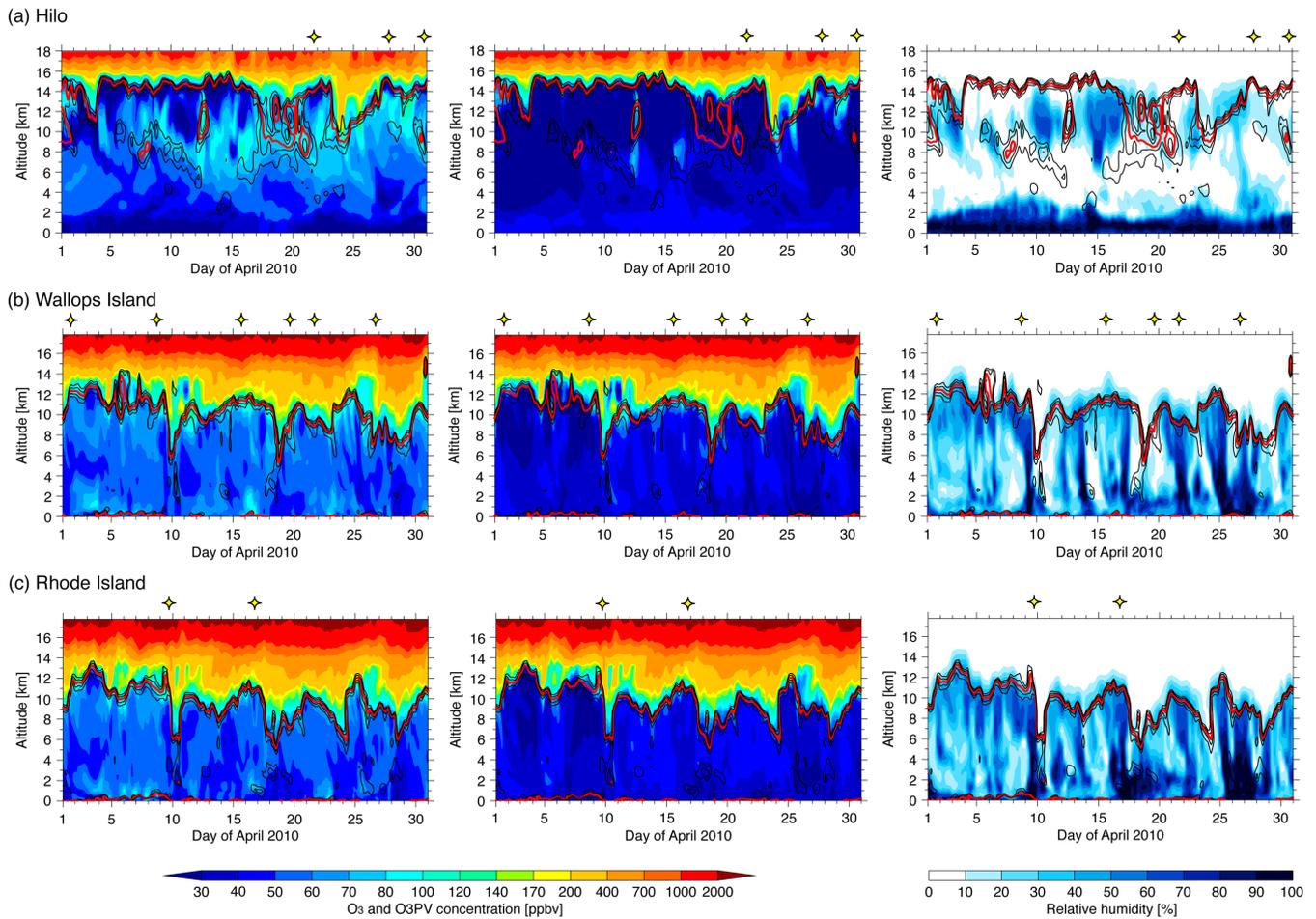
**Figure S3. The information of longitude and latitude in H-CMAQ modelling system.**

Dynamic tropopause

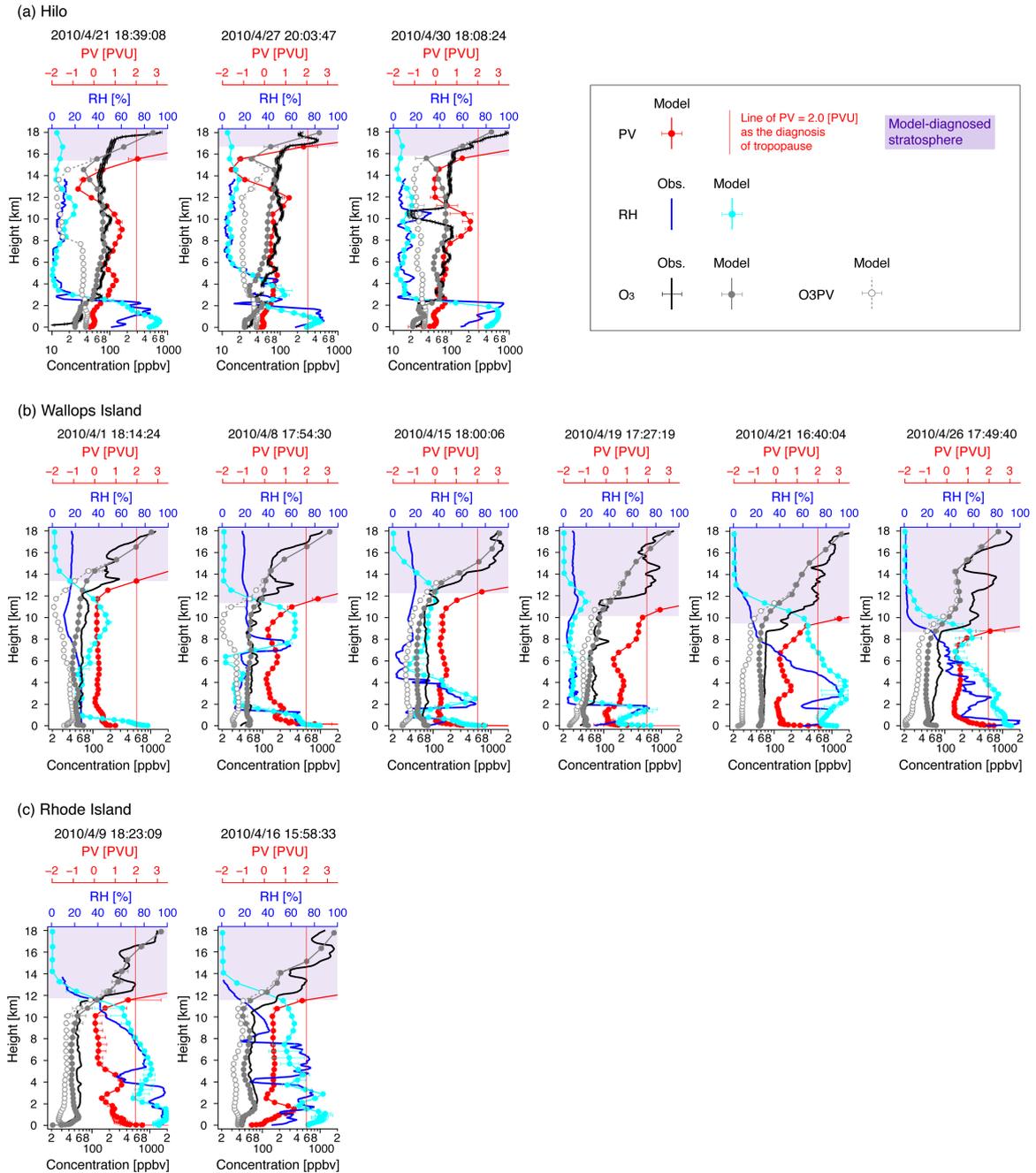
Thermal tropopause



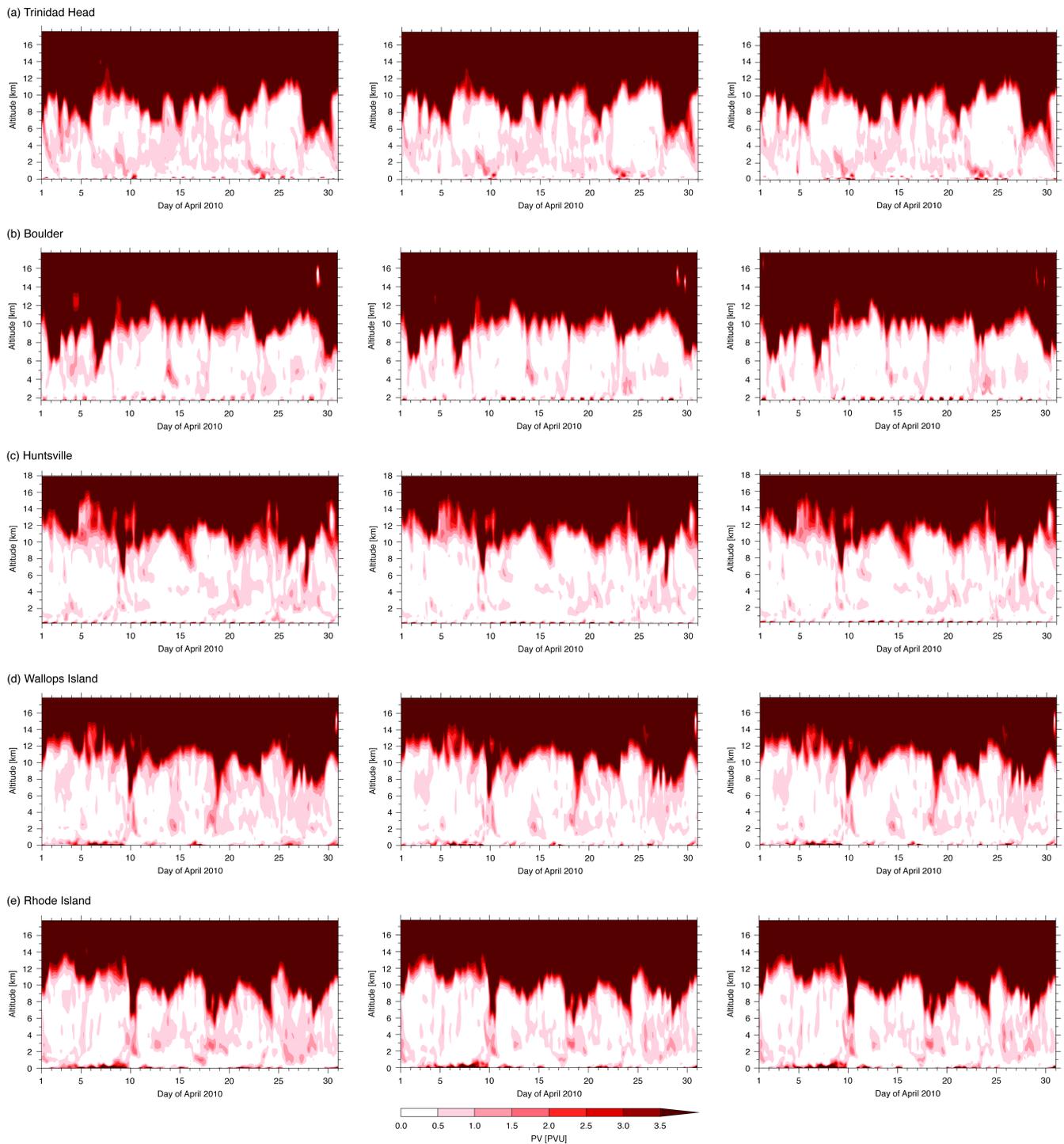
**Figure S4. Estimated tropopause altitude averaged over April 2010 by (left) the dynamic approach using PV in this work and (right) the thermal approach using the lapse rate.**



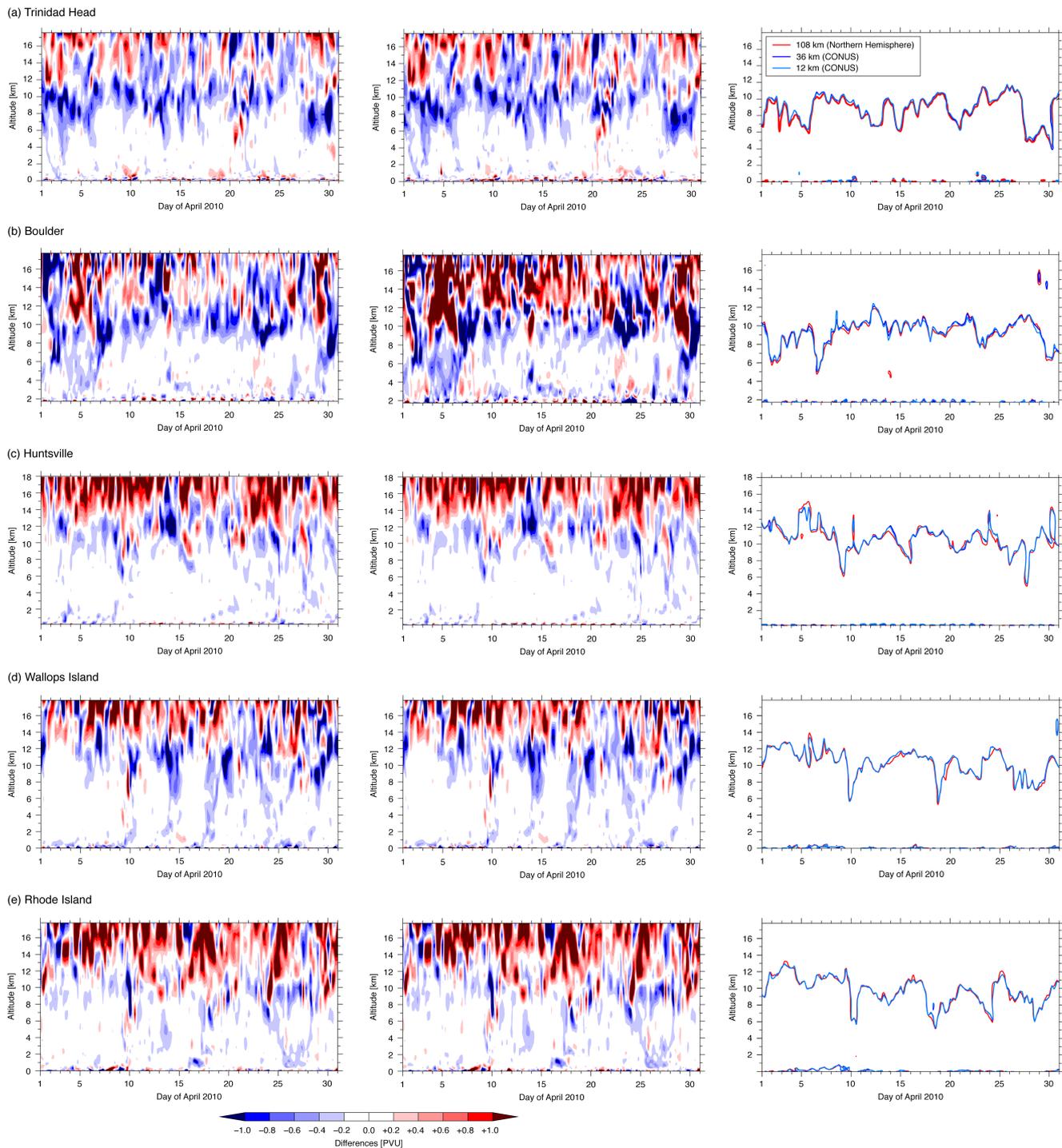
**Figure S5.** Curtain plots of modeled (left) O<sub>3</sub>, (center) O<sub>3</sub>PV, and (right) RH at U.S. ozonesonde sites of (a) Hilo (HI), (b) Wallops Island (VA), and (c) Rhode Island (RI) during April 2010. Yellow stars indicate the time of available ozonesonde measurements. Contour lines of modeled PV are also inserted for contours of 1.0, 1.5, 2.0, 2.5, and 3.0 PVU with thick red lines denoting the 2.0 PVU contour as an index to diagnose the tropopause. See also Figure 4.



**Figure S6. Vertical profiles of observed and modeled O<sub>3</sub> and RH at U.S. ozonesonde sites of (a) Hilo (HI), (b) Wallops Island (VA), and (c) Rhode Island (RI). Also see Figure S5 for ozonesonde measurement times. For modeled O<sub>3</sub> and RH, the hourly result corresponding to the ozonesonde measurement time is shown by circles, and the maximum and minimum model results within  $\pm 2$  hours of the measurement time are shown by whiskers. For observed O<sub>3</sub> at Hilo and Boulder, the range of uncertainties of the O<sub>3</sub> observations is shown by whiskers. Modeled O<sub>3</sub>PV and PV are also shown. Modeled PV profiles are plotted in red, and vertical lines corresponding to a PV value of 2 PVU are inserted as an index of the tropopause, and the layer range diagnosed as stratospheric air mass is colored in purple. See also Figure 5.**

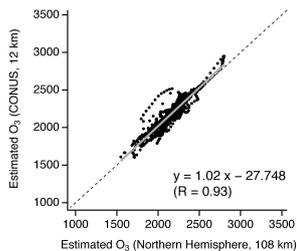
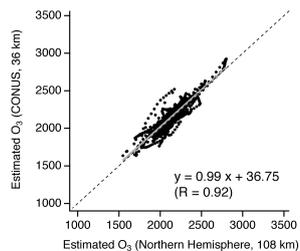


**Figure S7. Curtain plots of modeled PV by (left) WRF over northern hemisphere with a 108 km horizontal grid resolution, (center) WRF over CONUS domain with a 36 km horizontal grid resolution, and (right) WRF over CONUS domain with a 12 km horizontal grid resolution at U.S. ozonesonde sites of (a) Trinidad Head (CA), (b) Boulder (CO), (c) Huntsville (AL), (d) Wallops Island (VA), and (e) Rhode Island (RI) during April 2010.**

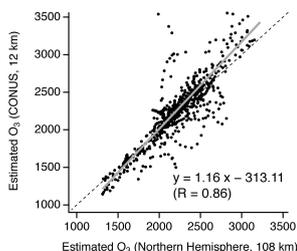
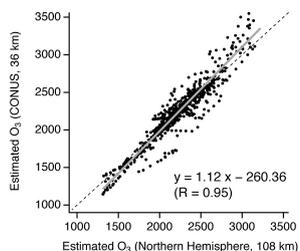


**Figure S8.** Curtain plots of (right) the difference of modeled PV calculated from 36 km–108 km, (center) from 12 km–108 km, and (left) modeled PV lines of 2 PVU by 108 km (red), 36 km (blue), and 12 km (sky blue) at U.S. ozonesonde sites of (a) Trinidad Head (CA), (b) Boulder (CO), (c) Huntsville (AL), (d) Wallops Island (VA), and (e) Rhode Island (RI) during April 2010.

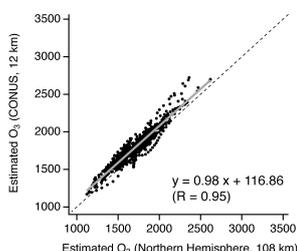
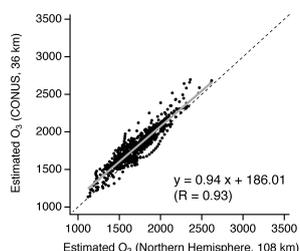
(a) Trinidad Head



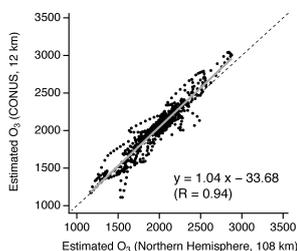
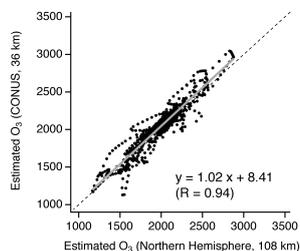
(b) Boulder



(c) Huntsville



(d) Wallops Island



(e) Rhode Island

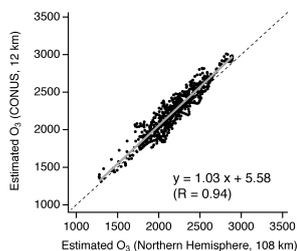
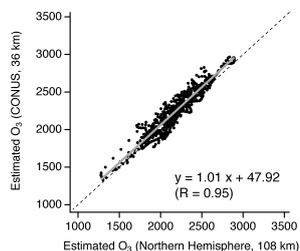
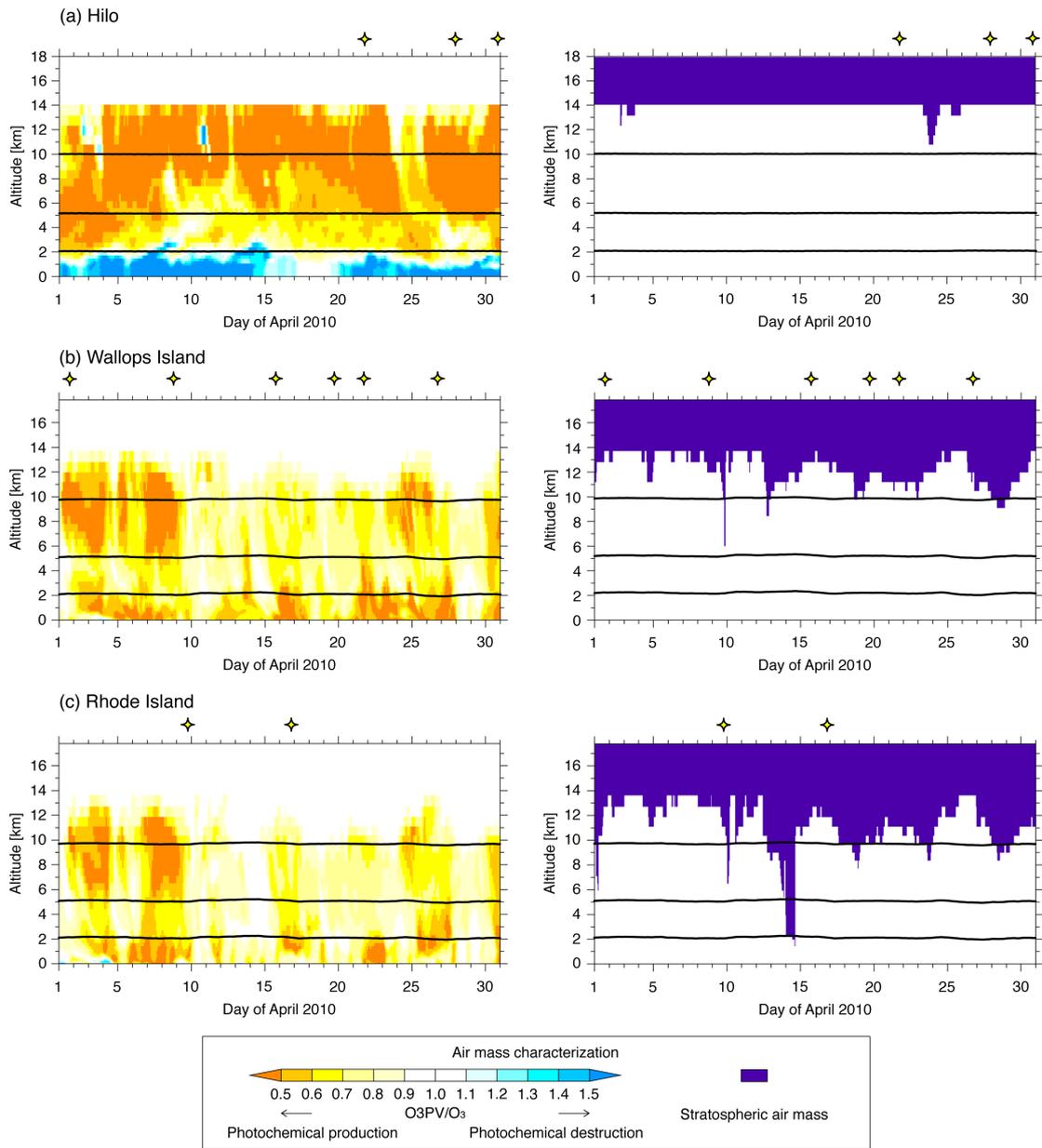
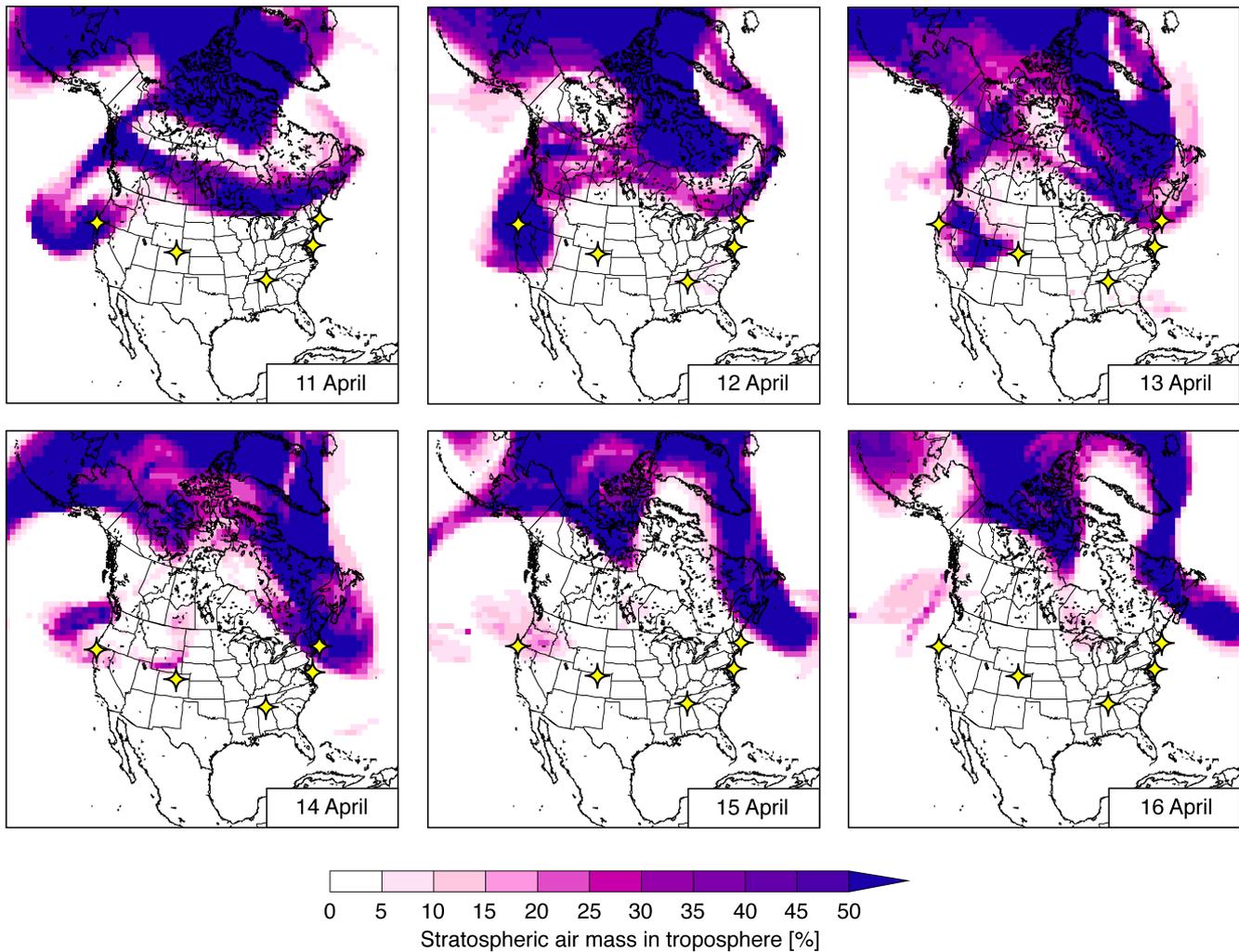


Figure S9. Correspondence of estimated O<sub>3</sub> concentration based on O<sub>3</sub>/PV relation at the uppermost layer as (left) 108 km vs. 36 km and (right) 108 km vs. 12 km at U.S. ozonesonde sites of (a) Trinidad Head (CA), (b) Boulder (CO), (c) Huntsville (AL), (d) Wallops Island (VA), and (e) Rhode Island (RI). Plots are hourly data during April 2010 (total number is 720).



**Figure S10.** Curtin plot of model-diagnosed air mass characterization for (left) O<sub>3</sub>PV/O<sub>3</sub> and (right) stratospheric air mass at U.S. ozonesonde sites of (a) Hilo (HI), (b) Wallops Island (VA), and (c) Rhode Island (RI) during April 2010. See also Figure 9.



**Figure S11.** Spatial distributions of day-to-day variations of stratospheric air mass contributions to total tropospheric O<sub>3</sub> column over the U.S. during middle April 2010. Yellow stars indicate the ozonesonde observational sites. See also Figure 11.

**Table S1. Statistical analysis of modeled O<sub>3</sub> mixing ratios to ozonesonde as latitude dependence.**

|                    | N   | Mean        |         | R                   | NMB    | NME   |
|--------------------|-----|-------------|---------|---------------------|--------|-------|
|                    |     | Observation | Model   |                     |        |       |
| Ozonesonde         |     |             |         |                     |        |       |
| < 40°N sites       |     |             |         |                     |        |       |
| –boundary layer    | 517 | 47.12       | 49.13   | 0.55 <sup>***</sup> | 4.3%   | 21.6% |
| –free troposphere  | 483 | 81.07       | 59.55   | 0.75 <sup>***</sup> | –26.5% | 29.6% |
| –upper model layer | 283 | 538.86      | 461.51  | 0.90 <sup>***</sup> | –14.4% | 33.8% |
| 40°–50°N sites     |     |             |         |                     |        |       |
| –boundary layer    | 907 | 49.20       | 48.95   | 0.37 <sup>***</sup> | –0.5%  | 16.6% |
| –free troposphere  | 831 | 77.97       | 58.12   | 0.76 <sup>***</sup> | –25.5% | 29.0% |
| –upper model layer | 488 | 932.12      | 845.30  | 0.92 <sup>***</sup> | –9.3%  | 26.9% |
| 50°–60°N sites     |     |             |         |                     |        |       |
| –boundary layer    | 966 | 46.64       | 43.37   | 0.55 <sup>***</sup> | –7.0%  | 16.9% |
| –free troposphere  | 776 | 78.39       | 56.28   | 0.78 <sup>***</sup> | –28.2% | 29.3% |
| –upper model layer | 461 | 1026.12     | 861.52  | 0.94 <sup>***</sup> | –16.0% | 25.1% |
| > 60°N sites       |     |             |         |                     |        |       |
| –boundary layer    | 387 | 40.30       | 37.06   | 0.56 <sup>***</sup> | –8.0%  | 20.3% |
| –free troposphere  | 378 | 92.86       | 60.65   | 0.78 <sup>***</sup> | –34.7% | 35.8% |
| –upper model layer | 221 | 1260.21     | 1061.29 | 0.94 <sup>***</sup> | –15.8% | 25.3% |

Note: Corresponded hourly modeled O<sub>3</sub> mixing ratio is used for the comparison with ozonesonde data. Significance levels by Students' t-test for correlation coefficients between observations and simulations are remarked as \*p < 0.05, \*\*p < 0.01, and \*\*\*p < 0.001, and lack of a mark indicates no significance. Ozonesonde observational sites located on < 40°N, 40°–50°N, 50°–60°N, > 60°N, respectively, contain 9, 9, 9, and 6 sites.

**Table S2. Statistical analysis of modeled relative humidity to ozonesonde as latitude dependence.**

|                        | N   | Mean        |       | R       | NMB   | NME    |
|------------------------|-----|-------------|-------|---------|-------|--------|
|                        |     | Observation | Model |         |       |        |
| <b>Ozonesonde</b>      |     |             |       |         |       |        |
| <b>&lt; 40°N sites</b> |     |             |       |         |       |        |
| –boundary layer        | 517 | 61.70       | 70.69 | 0.63*** | 14.6% | 26.0%  |
| –free troposphere      | 469 | 36.38       | 42.22 | 0.72*** | 16.1% | 42.3%  |
| –upper model layer     | 148 | 13.81       | 16.64 | 0.56*** | 20.5% | 77.2%  |
| <b>40°–50°N sites</b>  |     |             |       |         |       |        |
| –boundary layer        | 907 | 59.74       | 70.99 | 0.51*** | 18.8% | 27.5%  |
| –free troposphere      | 810 | 38.73       | 43.47 | 0.77*** | 12.5% | 34.1%  |
| –upper model layer     | 339 | 9.45        | 10.77 | 0.91*** | 13.9% | 44.2%  |
| <b>50°–60°N sites</b>  |     |             |       |         |       |        |
| –boundary layer        | 973 | 60.79       | 65.28 | 0.85*** | 7.4%  | 14.5%  |
| –free troposphere      | 776 | 36.04       | 41.90 | 0.85*** | 16.2% | 27.6%  |
| –upper model layer     | 461 | 6.64        | 9.27  | 0.93*** | 39.7% | 56.8%  |
| <b>&gt; 60°N sites</b> |     |             |       |         |       |        |
| –boundary layer        | 387 | 69.36       | 73.82 | 0.81*** | 6.4%  | 15.4%  |
| –free troposphere      | 378 | 42.46       | 49.42 | 0.80*** | 16.4% | 29.8%  |
| –upper model layer     | 172 | 2.65        | 4.12  | 0.74*** | 55.5% | 102.5% |

Note: Corresponded hourly modeled O<sub>3</sub> mixing ratio is used for the comparison with ozonesonde data. Significance levels by Students' t-test for correlation coefficients between observations and simulations are remarked as \*p < 0.05, \*\*p < 0.01, and \*\*\*p < 0.001, and lack of a mark indicates no significance. Ozonesonde observational sites located on < 40°N, 40°–50°N, 50°–60°N, > 60°N, respectively, contain 9, 9, 9, and 6 sites.

**Table S3. Elevated CASTNET sites in an alphabetical order.**

| ID     | Site name                    | State | Longitude (°) | Latitude (°) | Elevation (m a.s.l.) |
|--------|------------------------------|-------|---------------|--------------|----------------------|
| BBE401 | Big Band NP                  | TX    | -103.178      | 29.303       | 1052                 |
| CAN407 | Canyonlands NP               | UT    | -109.821      | 38.458       | 1809                 |
| CHA467 | Chiricahua NM                | AZ    | -109.389      | 32.009       | 1570                 |
| CNT169 | Centennial                   | WY    | -106.240      | 41.365       | 3175                 |
| CON186 | Converse Station             | CA    | -116.913      | 34.194       | 1718                 |
| GRB411 | Great Basin NP               | NV    | -114.216      | 39.005       | 2060                 |
| GRC474 | Grand Canyon NP              | AZ    | -112.184      | 36.059       | 2073                 |
| GTH161 | Gothic                       | CO    | -106.986      | 38.956       | 2915                 |
| JOT403 | Joshua Tree NP               | CA    | -116.389      | 34.070       | 1244                 |
| LAV410 | Lassen Volcanic NP           | CA    | -121.576      | 40.540       | 1756                 |
| MEV405 | Mesa Verde NP                | CO    | -108.490      | 37.198       | 2165                 |
| PAL190 | Palo Duro                    | TX    | -101.665      | 34.881       | 1053                 |
| PET427 | Petrified Forest             | AZ    | -109.892      | 34.823       | 1723                 |
| PND165 | Pinedake                     | WY    | -109.788      | 42.929       | 2386                 |
| PNF126 | Cranberry                    | NC    | -82.045       | 36.105       | 1216                 |
| ROM206 | Rocky Mountain NP Collocated | CO    | -105.546      | 40.278       | 2742                 |
| ROM406 | Rocky Mountain NP            | CO    | -105.546      | 40.278       | 2743                 |
| SHN418 | Shenandoah NP                | VA    | -78.435       | 38.523       | 1073                 |
| WNC429 | Wind Cave NP                 | SD    | -103.484      | 43.558       | 1292                 |
| YEL408 | Yellowstone NP               | WY    | -110.400      | 44.565       | 2430                 |
| YOS404 | Yosemite NP                  | CA    | -119.706      | 37.713       | 1605                 |

Note: Elevated sites defined as sites with an elevation higher than 1000 m a.s.l. (above sea level). The available sites during April 2010 are listed.