

Modeling Trans-Pacific Transport and Stratospheric Intrusion of Tropospheric Ozone using Hemispheric CMAQ during April 2010: Part 1. Model Evaluation and Air Mass Characterization for Stratosphere-Troposphere Transport

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Abstract.

15 Trans-Pacific transport and stratospheric intrusion has been recognized as a potential source of tropospheric ozone over the U.S.A. The state-of-the-science Community Multiscale Air Quality (CMAQ) Modeling System has recently been extended for hemispheric-scale modeling applications (referred to as H-CMAQ). In this study, H-CMAQ is applied to study the trans-Pacific transport and stratospheric intrusion during April 2010. The results will be presented in two continuous papers. In this part 1 paper, model evaluation for tropospheric ozone (O₃) is presented. Observations at the surface, by ozonesondes and
20 airplane, and by satellite across the northern hemisphere are used to evaluate the model performance for O₃. H-CMAQ is able to capture surface and boundary layer (defined as surface to 750 hPa) O₃ with a normalized mean bias (NMB) of -10%; however, a systematic underestimation with an NMB up to -30% is found in the free troposphere (defined as 750-250 hPa). In addition, a new air mass characterization method is developed to distinguish influences of stratosphere-troposphere transport (STT) from the effects of photochemistry on O₃ levels. This method is developed based on the ratio of O₃ and an inert tracer
25 indicating stratospheric O₃ to examine the importance of photochemistry, and sequential intrusion from upper layer. During April 2010 as monthly average, the relationship between surface O₃ mixing ratios and estimated stratospheric air masses in the troposphere show a slight negative slope, indicating that high surface O₃ values are primarily affected by other factors (i.e., emissions), whereas this relationship shows a slight positive slope at elevated sites, indicating that STT has a possible impact at elevated sites. STT shows large day-to-day variations, and STT impacts can either originate from the same air mass over
30 the entire U.S.A. with an eastward movement found during early April, or stem from different air masses at different locations indicated during late April. Based on this newly established air mass characterization technique, this study can contribute to

understand the role of STT, and also the implied importance of emissions leading to high surface O₃. Further research focused on emissions is discussed in a subsequent part 2 paper.

1 Introduction

Tropospheric ozone (O₃) is a secondary air pollutant produced by a chain of reactions involving photochemical oxidation of volatile organic compounds (VOCs) in the presence of nitrogen oxides (NO_x) (Haagen-Smit and Fox, 1954). Ozone plays a key role in tropospheric chemistry by controlling the oxidizing capacity through the production of hydroxyl (OH) radicals, and is an important greenhouse gas throughout the troposphere (Logan, 1985). Ground level O₃ poses significant risks to human health and therefore many countries regulate it as a criteria pollutant with an ambient air quality standard. In the U.S.A., the National Ambient Air Quality Standard (NAAQS) for O₃ is based on the annual 4th highest maximum daily 8-10 h concentration (MD8O3) averaged over three years and its threshold values have been decreasing from 80 ppbv in 1997 to 75 ppbv in 2008, and 70 ppbv in 2015 (EPA, 2018). Long-term trends of rural O₃ during 1990-2010 revealed significant O₃ decreases in the eastern U.S.A. during spring and summer whereas no significant O₃ decrease was found in the western U.S.A. during spring (Cooper et al., 2012). Analysis of trends in surface O₃ levels between 1998 and 2013 showed that the highest O₃ concentration in the U.S.A. has been reduced in response to substantial decline of precursor (Simon et al., 2015). It was also 15 shown that the O₃ concentration on low O₃ days have increased and led to the narrowing of the O₃ concentration range across the U.S.A.

From the viewpoint of global air quality changes, the dramatic variation of anthropogenic emissions in East Asia (Itahashi et al., 2013, 2014, 2015), may impact atmospheric composition at not only the local and regional scales, but also the global scale. By combining trajectory analysis with detailed chemical and meteorological data, it was suggested that the emissions were lifted into the free troposphere over Asia and then transported to North America in about 5-8 days (Jaffe et al., 1999). Trans-Pacific transport has been studied over the past decade because of its potential impact on rising background O₃ concentrations (Cooper et al., 2010). Asian contributions to surface O₃ levels in the U.S.A. require an additional challenge to meeting more stringent NAAQS for O₃ (Fiore et al., 2002). A typical case of trans-Pacific transport occurred during the so-called “perfect dust storm” during April 2001, transporting Asian dust to North America (Huebert et al., 2003). From an air 25 pollutant perspective, it was reported that the impact of Asian emissions increased background concentration of O₃ by 1 ppbv (2.5%) on a monthly average basis and up to 2.5 ppbv on a daily average basis over the western U.S.A. in April 2001 (Wang et al., 2009). Background O₃ levels entering western North America in spring have increased by approximately 10 ppbv between 1984 and 2002 based on a compilation of observations over the west coast of the U.S.A., and the possible cause for this increase was thought to be Asian emission trends (Jaffe et al., 2003). Asian air pollution can enhance surface O₃ mixing ratios by 5-7 ppbv over western North America in April-May 2006, and the doubled Asian anthropogenic emissions increase 30 during 2000-2006 was estimated to have the impact by 1-2 ppbv (Zhang et al., 2008). The global model simulation assuming the tripling of Asian anthropogenic emissions from 1985 to 2010 indicated an increase O₃ mixing ratios by 2-6 ppbv in the

western U.S.A. and by 1-3 ppb in the eastern U.S.A. on a monthly-mean basis, with the maximum effect occurring in April-June; this increase was suggested to more than offset the benefits of 25% domestic reduction in the western U.S.A. (Jacob et al., 1999). Based on the Emission Database for Global Atmospheric Research (EDGAR) version 4.3.1, anthropogenic emissions of NO_x and VOCs from China are estimated to have increased by 3.2 and 2.1 times during 1985-2010, respectively (Crippa et al., 2016), which is generally consistent with the assumption by Jacob et al. (1999).

The occurrence of trans-Pacific transport can be inferred from variations in the jet stream related to La Niña and El Niño. The springtime Asian outflow may be enhanced following an El Niño winter due to the eastward extension of the atmospheric circulation over the Pacific-North America sector and the southward shift of the subtropical jet stream (Koumoutsaris et al., 2008; Lin et al., 2015). According to the NOAA Climate Prediction Center (CPC), 2009–2010 wintertime was influenced by strong El Niño conditions (NOAA, 2018). Because of the favorable condition for trans-Pacific transport, it was reported that Asian dust reached North America on at least five occasions during April 2010 (Uno et al., 2011). During May-June 2010, the Asian enhancement to MD8O₃ in the western U.S.A. was estimated to reach 8-15 ppbv in high-elevation regions during strong trans-Pacific transport events (Lin et al., 2012a).

Another process affecting tropospheric O₃ is stratosphere-to-troposphere transport (STT), which is known to be a significant contributor to the tropospheric O₃ budget (Lelieveld and Dentener, 2000). The tightening of the O₃ NAAQS and a continuous decrease of anthropogenic emissions have led to an increased focus on STT. On one hand, stratospheric intrusion of O₃ was found to be below 20 ppbv during March-October 2001 over the entire U.S.A (Fiore et al., 2003). On the other hand, a total of thirteen events were identified during April-June 2010 when stratospheric intrusion impacts reached 20-40 ppbv while accounting for 50-60% of total O₃ at fifteen high-elevation (> 1.4 km above mean sea level; m.s.l.) sites in the western U.S.A. (Lin et al., 2012b). From the perspective of interannual variability, springtime stratospheric intrusions may be enhanced following a La Niña winter due to a meandering of the jet stream, and a large variability in terms of magnitude and frequency have been shown from 1990 to 2012 (Lin et al., 2015). The fraction of O₃ in the troposphere that originates from the stratosphere is still uncertain due to its strong dependence on season and location which affect tropopause heights and is therefore still an area of active research (Mathur et al., 2017).

Table 1 summarizes these studies that provided the motivation for evaluating the impacts of both precursor emissions and STT on tropospheric O₃. April 2010 is selected as the study period because enhancement of trans-Pacific transport is expected during the 2009-2010 El Niño winter. Along with the gradual reduction of precursor emissions of NO_x and VOCs in the U.S.A., a gradual decrease of MD8O₃ mixing ratios can be expected and showed a decreasing trend by 0.4%/year; however, mean MD8O₃ mixing ratios in 2010 showed a local maximum and the number of NAAQS threshold exceedances was larger than usual as shown in Fig. S1 in the supplemental material. The variation in monthly mean and percentile distribution of observed MD8O₃ during 2010 are shown in Fig. S2 in the supplemental material. Although high MD8O₃ concentration for 95th percentiles and the number of NAAQS exceedances were found during summer time, it is also apparent that mean MD8O₃ during April 2010 higher than any other month. Lower MD8O₃ concentration for 5th and 25th percentiles were also noted as comparatively high during April 2010, indicating widespread enhancement of low-level O₃ further suggesting the possible

impacts of trans-Pacific transport on O₃ levels across the U.S.A. during this month. This period has already been the subject of other studies (e.g., Uno et al., 2011; Lin et al, 2012a), however, the methods used in this study to investigate the impacts of trans-Pacific transport differ from previous studies. The objective of this study is to better understand the relative contributions of precursor emissions from East Asia and the U.S.A. because the trans-Pacific transport has been recognized as an important factor. Previous studies primarily focused on Asian impacts on the western U.S.A., while this study investigates impacts across the entire U.S.A. In addition, some stratospheric intrusion events have been reported during spring 2010 (Lin et al., 2012b), therefore this period is suitable to examine not only trans-Pacific transport but also stratospheric intrusion, both processes may contribute to the observed high O₃ episodes in the U.S.A. Examination of the impacts of both processes will shed light on the formation mechanisms underlying such high O₃ episodes, thus improving our understanding of their relative importance in leading to these high O₃ episodes. The results of this work will be presented in two parts. Part 1 paper focuses on characterizing the influence of stratosphere-troposphere transport on O₃ distribution in the lower to middle troposphere. A sequential Part 2 manuscript focuses on the contributions of emissions leading to higher O₃ mixing ratio through Trans-Pacific transport. In this part 1 paper, we present the model evaluation and introduce a new method to identify and characterize periods during which surface and lower tropospheric O₃ may be influenced by stratospheric intrusions. This manuscript is organized as follows. In section 2, the modeling system and simulation set up are described, details on the surface, ozonesonde, airplane, and satellite observations used to evaluate the model performance are presented, and evaluation protocols are defined. In section 3, the analysis of model results and comparisons with observations are documented and the newly developed air mass characterization method is introduced and applied to investigate stratospheric intrusions. Finally, the conclusion section includes limitations of this work, future perspectives, and a brief introduction to part 2.

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2 Methodology

2.1 Modeling System and Simulation Set Up

The model used in this work is the Community Multiscale Air Quality (CMAQ) version 5.2 extended for hemispheric applications (H-CMAQ) (Mathur et al., 2017). To investigate the impact of emissions from East Asia, H-CMAQ is configured to cover the entire Northern Hemisphere, utilizing a horizontal discretization of 187×187 grid points with a grid spacing of 108 km. The information of longitude and latitude is presented in Fig. S3 in the supplemental material. While the use of finer horizontal grid spacing can better resolve the STT processes, it will substantially increase computational demands. Cristofanelli et al. (2003) analyzed STT by combining analysis of data from a measurement network and predictions from total of seven model simulations over Europe, and reported that three models with 1°×1° horizontal resolution were able to capture the STT whereas other models with coarser resolutions were not. Another study over Europe investigated the cross-tropopause transport in terms of resolution and diffusion coefficient using horizontal resolutions of 2°×2°, 1°×1°, and 0.5°×0.5° and showed that the simulation with the 2°×2° resolution has difficulty to capture the tracer amount across tropopause (Gery, 2003). Based on

these findings and the model evaluation results (see Section 3.1) in this work, we believe that using a grid resolution of 108 km provides a good compromise between numerical accuracy and computational constraints. The terrain-following vertical coordinate utilizes 44 layers of variable thickness to resolve the model vertical extent between the surface and 50 hPa based on the extension of the previous 35 layers system (Mathur et al., 2017). The revised layer structure using 44 layers with significantly finer resolution above the boundary layer (BL) better represents long-range transport in the free-troposphere (FT) as well as STT processes, and influences from cloud mixing on both the sub-grid and resolved scales. As indicated in Mathur et al. (2017), the 44 layer configuration employed in the H-CMAQ configuration helps better capture dynamics in the vicinity of the tropopause and reduce excessive diffusion relative to coarser vertical resolution configurations. The emission inputs are based on the Hemispheric Transport of Air Pollution version 2 (HTAP2) modeling experiments, and the detailed description can be found in previous studies (Janssens-Maenhout et al., 2015; Pouliot et al., 2015; Galmarini et al., 2017; Hogrefe et al., 2018). The lightning emissions are prescribed using climatological averages as estimated in the Global Emission Inventory Activity (GEIA) as dataset (Price et al., 1997). For gas-phase chemistry, cb05e51 is used (Appel et al., 2017). This gas-phase mechanism includes the condensed halogen chemistry that leads to O₃ loss in marine environments (Sarwar et al., 2015). For aerosol chemistry, aero6 with nonvolatile primary organic aerosol (POA) (Simon and Bhawe, 2012) is adopted. The boundary conditions of H-CMAQ are taken from the clean tropospheric background values with updates to the physical and chemical sinks for organic nitrate species (Mathur et al., 2017).

Potential vorticity (PV) has been shown to be a robust indicator of air mass exchange between the stratosphere and the troposphere. The value of PV itself generally increases with altitude, and previous studies suggested that a value of 2 PVU (1 PVU = 10⁻⁶ m² K kg⁻¹ s⁻¹) is an indicator of stratospheric air (Hoskins et al., 1985; Wernli and Bourqui, 2002; Itoh and Narazaki, 2016). Through this study, the tropopause is diagnosed by 2 PVU. The calculation of the tropopause altitudes using PV (dynamical tropopause) and the traditional approach based on the lapse rate (thermal tropopause) defined by World Meteorological Organization (WMO) (WMO, 1992) have been reported (Hoering et al., 1991).” As shown in Figure S4, estimated tropopause altitudes averaged over April 2010 using PV in this work and the traditional approach of WMO are overall similar with below 10 km over high-latitude region and above 16 km over low-latitude region. PV shows a strong positive correlation with O₃ (Danielsen, 1968), and modeling studies have used this correlation to develop scaling factors that specify O₃ in the modeled upper troposphere/lower stratosphere (UTLS) based on estimated PV. The reported O₃/PV ratios exhibited a wide range from 20 to 100 ppbv/PVU with differences in locations, altitude and season (e.g., Ebel et al, 1991; Carmichael et al, 1998; McCaffery et al, 2004). To account for the seasonal, latitudinal and altitude dependencies in the O₃/PV relationship, a dynamic O₃/PV function was developed to consider latitude, altitude, and time based on 21-year ozonesonde records from the World Ozone and Ultraviolet Radiation Data Centre (WOUDC) and corresponding PV values from WRF-CMAQ simulations across the northern hemisphere from 1990 to 2010 and is used in H-CMAQ (Xing et al., 2016). This parameterization of O₃/PV is constructed at three topmost vertical levels of 58, 76, and 95 hPa fitted as a 5th order polynomial function, and applicable between the range of 50 and 100 hPa. Based on this new parameterization, it was demonstrated that UTLS O₃ agreed much better with observation in terms of its magnitude and seasonality (Xing et al., 2016). Mathur et al.

(2017) further demonstrated improvements in representation of seasonal variations in surface O₃ using the parameterization. To track stratospheric air masses, the O₃ estimated using the O₃/PV relationship in the three layers listed above is also added as a chemically-inert tracer species in the H-CMAQ simulations as O₃PV tracer. The O₃PV tracer undergoes the same transport, scavenging, and deposition processes as O₃, but its mixing ratios are not affected by chemical production or loss processes.

5 The meteorological fields are simulated by the Weather Research and Forecasting (WRF) model version 3.6.1 using the same vertical configuration as H-CMAQ. WRF simulation started from 1 March 2009 with more than one year of spin-up time prior to the analysis period of April 2010. The WRF model is configured to use the rapid radiative transfer model for global climate models (RRTMG) radiation scheme for both longwave and shortwave (Iacono et al., 2008), Morrison double-moment scheme (Morrison et al., 2009) and Grell convective parameterization (Grell 1993; Grell and Devenyi, 2002) for
10 microphysics and cumulus parameterization, and Mellor-Yamada-Janjic scheme for planetary boundary layer (Janjic et al., 1994). Wind, temperature, and water vapor fields are nudged towards NCEP/NCAR final analysis data for all layers, these analysis data have 1 degree spatial and 6 h temporal resolution (NCEP, 2018). The WRF meteorological fields are converted to the format required by H-CMAQ using MCIP version 4.3 (Otte and Pleim, 2010), and then used for the H-CMAQ simulation. Relative humidity (RH) can also be used to diagnose stratospheric air masses because the stratosphere is characterized by dry
15 air. CMAQ used the meteorological fields simulated by WRF and calculated RH based on the improved Magnus form approximation for saturation vapor pressure (Alduchov and Eskridge, 1996), and internally set the maximum value on 99% and minimum value on 0.5%. The CMAQ simulation started from 1 March 2010 and initialized with three-dimensional chemical fields from prior model simulations for 2010 by Hogrefe et al. (2018); March is discarded as a spin-up period and April is used as the analysis period. The O₃PV tracer is also initialized by this prior model simulations of Hogrefe et al. (2018).

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2.2 Observations and Evaluation Protocols

2.2.1 Ground-based Surface O₃ Observations

The northern hemispheric modeling domain and ground-based observations used in this study are shown on the map in Fig. 1. Global ground-based surface O₃ observations were obtained from the World Data Centre for Greenhouse Gases (WDCGG; shown as red circles in Fig. 1). For the study period of April 2010, this dataset contained 52 sites in North America, Europe, and several remote locations with only limited coverage over Asia (WDCGG, 2018). To overcome this limitation, surface O₃ observations are also obtained from the Acid Deposition Monitoring Network in East Asia (EANET) program which provides measurements at 12 sites in Japan, 3 sites in the South Korea, 1 site in Russia, and 4 sites in Thailand. However, the observed data are only available on a daily-mean basis for Russia, and a monthly-mean basis for South Korea and Thailand.
25 Therefore, the only EANET monitors used in this study (EANET, 2018) are those located in Japan; these 9 sites with available data for April 2010 are shown as green triangles in Fig 1. In addition, surface O₃ observations over the U.S.A. were obtained from the Clean Air Status and Trends Network (CASTNET) and are shown as blue squares in Fig. 1. CASTNET monitors (CASTNET, 2018) are located mostly in rural and remote areas, which makes them appropriate for comparison to O₃ fields
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from the coarse resolution H-CMAQ simulations. CASTNET data are available at 81 sites during April 2010. MD8O3 values for April 2010 are calculated from the hourly observations at these WDCGG, EANET, and CASTNET stations.

2.2.2 Ozonesondes

5 An evaluation of simulated vertical O₃ profiles is needed to analyze the model's ability to capture the behavior of aloft O₃. To this end, we obtained ozonesonde data distributed by the WOUDC as well as additional ozonesonde soundings available over the U.S.A. and Greenland that are collected and distributed by the National Oceanic and Atmospheric Administration Earth System Research Laboratory (NOAA-ESRL) (NOAA, 2018a). The total number of available ozonesonde sites during April 2010 was 33 (locations shown as yellow stars in Fig. 1). The data for Hilo and Boulder are available in both
10 the WOUDC and NOAA-ESRL database; the NOAA-ESRL data are used because they include information on uncertainties of the O₃ measurements. Detailed information for each site, including country, site name, latitude (°N), longitude (°E), elevation (m a.s.l.), and the number of launches during April 2010, is provided in Table 2. There are 6 sites located in the U.S.A., 10 sites in Canada, 5 sites over Asia, and 12 sites over the Europe. In addition to measured O₃ mixing ratios, observed RH vertical profiles are used to evaluate the model performance.

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2.2.3 Airplane

In addition to ozonesonde data to evaluate the vertical O₃ distribution, observations from research aircraft for three sites located in the U.S.A. (Cape May, New Jersey; Homer, Illinois; and Southern Great Plains, Oklahoma) are available from NOAA-ESRL (NOAA, 2018b) for April 2010. Because the observations at Cape May and Homer are only available for a single day
20 during April 2010, we only used the NOAA-ESRL aircraft data at Southern Great Plains which is shown as a gray diamond in Fig. 1. A total of seven flights were conducted at this site during April 2010. In addition to O₃ mixing ratios, RH was used to evaluate the model performance.

2.2.4 Satellite

25 Tropospheric column O₃ observed by the Ozone Monitoring Instrument (OMI) onboard the National Aeronautics and Space Administration (NASA) Earth Observing System Aura satellite is used in this study. The methodology to estimate the tropospheric column has been developed (Ziemke et al., 2006) and consists of taking the differences between total column O₃ observed by OMI and stratospheric column O₃ observed by the Microwave Limb Sounder (MLS). The monthly-mean tropospheric O₃ column data are available between 60°S and 60°N (NASA, 2018a). Because this tropospheric column O₃ data
30 are monthly-mean data, in order to take into account the daily missing data by OMI, total column data (OMTO3d) are utilized to obtain the information on daily missing data in order to compare with the model (NASA, 2018b). This total column data

are the products of averaging only good quality flag of level-2 swath data and then gridded into 1×1 degree. Such an approach considering daily deficit data has also been applied in previous study (e.g., Chatani et al., 2014). To diagnose the tropopause in the model, PV with a value of 2 PVU is used as threshold. This diagnosis is applied above the boundary layer to avoid the misdiagnosis near the surface due to the high value of PV caused by turbulence.

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2.2.5 Evaluation Protocol

To evaluate model performance, the Pearson's correlation coefficient (R) with student's *t*-test is used for assessing the statistical significance level. The normalized mean bias (NMB) and the normalized mean error (NME) are calculated using the following equations (e.g., Zhang et al., 2006);

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$$R = \frac{\sum_1^N |(O_i - \bar{O})(M_i - \bar{M})|}{\sqrt{\sum_1^N (O_i - \bar{O})^2} \sqrt{\sum_1^N (M_i - \bar{M})^2}} \quad (1)$$

$$NMB = \frac{\sum_1^N (M_i - O_i)}{\sum_1^N O_i} \quad (2)$$

$$NME = \frac{\sum_1^N |M_i - O_i|}{\sum_1^N O_i} \quad (3)$$

where, N is the total observation number, O_i and M_i represent each individual observation and model value respectively, and \bar{O} and \bar{M} represent the arithmetical mean of observations and model values respectively. Based on a compilation of model evaluation reports, Emery et al. (2017) suggested threshold values of $R > 0.75$, $NMB < \pm 5\%$, and $NME < 15\%$ as performance goal, and threshold values of $R > 0.50$, $\pm 5\% < NMB < \pm 15\%$, and $15\% < NME < 25\%$ as performance criteria for 1-hr O_3 or MD8O3 simulated by regional-scale air quality models. Although these recommendations were developed for regional-scale air quality models and suggested to apply over time-space averaging scales of no longer than one month and no more than 1000 km, these three criteria are applied in this work to judge the performance of the April 2010 H-CMAQ simulations due to the lack of other commonly-accepted model performance criteria for hemispheric or global scale O_3 simulations. Evaluation of surface O_3 simulated by global models indicated a somewhat loose threshold might be required because of the use of a coarse grid resolution (Zhang et al., 2012; He et al., 2015a, 2015b).

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3 Simulation Results and Discussion

3.1 Model Evaluation

A scatterplot of modeled vs. observed MD8O3 at WDCGG, EANET, and CASTNET, sites during April 2010 is shown in Fig. 2 using colors and symbols that are consistent with Fig. 1. A summary of the statistical analysis is provided in Table 3. Almost all of the EANET (green triangles) and CASTNET (blue squares) MD8O3 data pairs were within the 1:2 lines across the entire O₃ mixing ratio range. The comparison of H-CMAQ values with EANET observations over Asia shows an R value of 0.49 which is statistically significant at a level of $p < 0.001$, an NMB of -12.6% and an NME of 20.6% (Table 3). A comparison to CASTNET observations over the U.S.A. shows that the mean observed and modeled values are close with an NMB of -0.9% , and an NME of 12.6% , and that R had a value of 0.61 with $p < 0.001$. This indicates that the H-CMAQ simulations captured the CASTNET observational data within the model criteria performance suggested by Emery et al. (2017). A comparison to WDCGG data across the Northern Hemisphere shows an R of 0.49 with $p < 0.001$, an NMB of -19.3% , and an NME of 23.7% . The mean model value is approximately 10 ppbv less than the mean of the observations. This feature is also evident in the scatter-plot shown in Fig. 2. While observed values reach more than 100 ppbv of MD8O3, the corresponding model values are only about half of these high MD8O3 mixing ratios, indicated by a clustering of WDCGG pairs on the 2:1 line. To investigate this model underestimation further, the spatial distributions of monthly mean modeled and observed O₃ mixing ratios are examined in Fig. 3 which shows observed high mixing ratios over eastern Europe. In particular, the four WDCGG monitors at Kosetice, Czech Republic (15.08°E , 49.58°N , 534 m a.s.l.), K-puszta, Hungary (19.55°E , 46.97°N , 125 m a.s.l.), Rucava, Latvia (21.17°E , 56.16°N , 18 m a.s.l.), and Zoseni, Latvia (25.54°E , 57.08°N , 182 m a.s.l.) measured MD8O3 mixing ratios larger than 100 ppbv by 14, 10, 5, and 4 days, respectively, during April 2010. An analysis of data collected during an airborne measurement campaign during 15-18, April 2010 over Siberia reported enhanced O₃ mixing ratios influenced by long-range transport, biomass burning plumes, and stratospheric intrusion (Berchet et al., 2013). Since the biomass burning emissions used in the current H-CMAQ simulations are based on climatological averages rather than year-specific events, the model underestimation may at least partially be due to the representation of these emissions. From the viewpoint of meteorology, the blocking events over European Russia during spring-summer 2010 were reported, and positive anomaly of O₃ total column over the regions adjacent to the anticyclones (i.e., Europe) were analyzed (Sitnov et al., 2017). Removing data from these four sites from the analysis yields model performance metrics of an R of 0.63 with $p < 0.001$, an NMB of -14.4% , and an NME of 19.5% ; which are comparable to performance at the EANET sites. Aside from the underestimation of high observed MD8O3 mixing ratios at these four European sites, H-CMAQ generally captured the WDCGG observations. Summarizing the model evaluation with surface observations, it is confirmed that model reasonably captures MD8O3 almost within model performance criteria of Emery et al. (2017).

To investigate the vertical profiles of O₃, ozonesonde and airplane data are used in this study. In Fig. 4 and Fig. S5 in the supplemental material, time-height cross-sections (“curtain plots”) of hourly modeled O₃, O3PV, and RH values during April 2010 are shown at the location of the ozonesonde sites in the U.S.A., i.e., at Hilo, HI, Trinidad Head, CA, Boulder, CO,

Huntsville, AL, Wallops Island, VA, and Rhode Island, RI. The plots also show contour lines of modeled PV for PV values of 1.0, 1.5, 2.0, 2.5, and 3.0 PVU with the red thick lines indicating a value of 2.0 PVU that can be used to diagnose the tropopause. Generally, O₃ and O₃PV mixing ratios are very similar in the upper layers, especially above the 2.0 PVU line, indicating that O₃ mixing ratios in these layers are dominated by stratospheric air mass. Below the tropopause as diagnosed by the PV = 2.0 PVU line, O₃ mixing ratios are generally higher than O₃PV mixing ratios, suggesting that O₃ was photochemically produced in the troposphere. On the other hand, instances of O₃ mixing ratios lower than O₃PV mixing ratios are indicative of photochemical loss. One typical example of such photochemical loss can be seen at Hilo (Fig. S5 (a)). At that location, O₃ mixing ratios are less than 30 ppbv below 2 km whereas the O₃PV mixing ratios are larger than 40 ppbv. A likely driver of this strong O₃ loss is the halogen chemistry in marine environments implemented in H-CMAQ (Sarwar et al., 2015) because Hilo site is surrounded by ocean. The impact of photochemical processes is further discussed in Section 3.2. Ozone mixing ratios at the level of the tropopause as diagnosed by the 2 PVU lines generally are around 100 ppbv (light blue colors in Fig. 4). Although high values of PV are typically seen in the upper layer above 10 km, it should be noted that high PV can occasionally also be found in the lower troposphere where could be associated with convection. RH values are below 10% (white colors in Fig. 4) above the tropopause and steeply increased near or below the tropopause as diagnosed by the 2.0 PVU lines. Based on the rough estimation shown in the curtain plots of Fig. 4, RH values at the level of the tropopause are typically on the order of 30-40%. In Fig. 4, the launch times of available ozonesonde measurements are indicated by yellow stars and we discuss below the comparison of model profiles to measurements from these launches.

The vertical profiles of observed and modeled O₃ and RH, as well as modeled O₃PV and PV are shown in Fig. 5 and Fig. S6 in the supplemental material. In this figure, vertical red lines corresponding to a PV value of 2.0 PVU are inserted as index of stratospheric air masses, and the diagnosed stratosphere is colored with purple. A quantitative comparison between simulations and observations is conducted by averaging the observations onto the vertical grid spacing used by the model. The vertical layers are then assigned to three vertical ranges based on typical pressure values, i.e., the boundary layer (surface to approximately 750 hPa), the free troposphere (approximately 750-250 hPa), and the upper model layers (approximately 250-50 hPa) following the same approach used in our previous study (Hogrefe et al., 2018). Furthermore, the statistical analysis is performed separately for the three regions of U.S.A. and Canada, Asia, and Europe and the three layer ranges defined above. As shown in Fig. 4, O₃ and O₃PV show similar variation in the upper model layer; however, O₃ is greater than O₃PV near the tropopause indicated by 2.0 PVU, and this suggests the presence of photochemical production near the tropopause. Results of this statistical analysis for O₃ mixing ratios are shown in Table 3 and reveal that over all three regions the model performed the best for the boundary layer in terms of NMB and NME. The observed mean boundary layer values of around 45 ppbv over the three regions are well captured by the model. Over the U.S.A. and Canada, model performance in the boundary layer satisfies the performance criteria for all three metrics of R, NMB, and NME, and over Asia and Europe, NMB and NME also satisfy the performance criteria whereas R is less than 0.5. Compared to the results for the boundary layer, the model tends to underestimate the observed O₃ mixing ratios in the free troposphere and the upper model layers. In the free troposphere, the mean observed value is around 80 ppbv while the mean model value is below 60 ppbv. As a result, NMB values are greater

than -15% and NME values are greater than 20%. This underestimation is also present in the upper model layers; the mean observed values of 500 – 1000 ppbv are consistently underestimated by about 100 ppbv by the model across the three regions as shown in Table 3. R values tend to increase from the boundary layer to the free troposphere and the upper model layers due to model's ability to capture the increase of O₃ mixing ratios with height. The higher R values in the free troposphere, a region where impacts of photochemistry on O₃ variability is smaller, also suggest greater confidence in the model dynamics, which drive O₃ variations in this part of the atmosphere. Table S1 in the supplemental material shows the statistical results that are obtained when grouping stations into latitude ranges. The results indicate that model performance is similar to that shown in Table 3 and discussed above. These results suggest that although the revision of the dynamic-PV approach described in Xing et al. (2016) led to improved results compared to earlier implementations of the scaling approach, there is a need for further refinement of the approach to better capture high mixing ratios of stratospheric O₃. Using a finer vertical resolution for the upper layers and extending the model top beyond 50 hPa to cover larger portions of the stratosphere could be potential strategies to address this need. This is a hemispheric modeling system but the finer horizontal resolution will be another way to improve this. In addition, the uncertainty of the lightning emissions prescribed as climatological averages in the current simulations may also contribute to the underestimation of O₃ in the free troposphere.

Although RH is a diagnostic variable, it may also provide an indication for stratospheric air mass, it is thus included in the model evaluation. As expected, Fig. 5 shows that RH has higher values and large variations in the troposphere and lower values in the stratosphere. For the analysis of modeled vertical profiles, model results of maximum and minimum values within ± 2 hours from observation time are also shown, and the range of RH showed large variations at lower altitude. Table 4 summarizes the statistical analysis divided into the three regions and three vertical domains for RH. It was found that the model generally overestimates RH over all regions and all three layers ranges. Although the NME value seems high for the upper layers, this is caused by the low absolute values of RH. The mean absolute differences between observed and modeled RH values are 1-2 % over the U.S.A. and Canada and Europe, and 8% over Asia. In Table S2 of the supplemental material, the RH results in Table 4 are presented for different latitude bands in the same fashion as the O₃ results in Table S1. Results show that model performance is similar to that discussed for Table 3. The systematic positive bias of RH occurs despite using analysis nudging for wind, temperature, and water vapor in the WRF simulations. Positive bias in predicted RH is also found in meteorological simulations performed for AQMEII (Vautard et al., 2012).

The tropopause diagnosed by PV = 2.0 PVU is located around 10-12 km at five ozonesonde sites in the U.S.A. except Hilo where it is located around 16 km. Observations in late April show instances of tropopause heights at or below 6 km (e.g., 27 April at Trinidad Head (Fig. 5 (b)), 29 April at Boulder (Fig. 5 (c)), and 27 April at Huntsville (Fig. 5 (d))). These cases illustrate large impacts of episodic STT, with observed O₃ mixing ratios steeply increasing from 100 ppbv at around 6 km to over 500 ppbv at around 8 km. The profiles obtained from the H-CMAQ simulations do not capture this steep increase and only show a gradual increase. This finding further supports a need for further refinement of representing stratospheric high O₃ mixing ratios as discussed above in the context of Table 3. In terms of RH, observed RH values show a sudden decline from

around 60% to near 0% at Trinidad Head and Huntsville, whereas modeled RH values show a gradual decrease with large temporal variations. This contributes to the modeled positive RH bias shown in Table 4.

The comparison of model 3D O₃ structure at Southern Great Plains, Oklahoma, with research aircraft measurements is illustrated in Fig. 6. At this site, the curtain plot of modeled O₃ is shown for the entire month of April 2010 in the top row and zoomed inserts for the seven observational times indicated by gray diamonds above those plots are shown in the second row with each box showing airplane observations overlaid on H-CMAQ values. For O₃, observed and modeled mixing ratios increased from about 30 ppbv at 1 km to about 55 ppbv at 5 km, except for flight #1 which shows persistently high mixing ratios of 50-60 ppbv throughout this altitude range. However, observed high mixing ratios of O₃ over 70 ppbv during flight #5, 6, and 7 are not captured by H-CMAQ. In contrast to O₃, observed and modeled RH generally decreased from 1 km to 5 km as shown in rows 3 and 4. Overestimation in model RH is noted for flights #3, 4, and 6 above 3 km. Considering the profiles of O₃ and RH, flight # 6 might be a case of STT because observed RH is less than 10% and observed O₃ mixing ratios exceed 75 ppbv; however, the model fails to reproduce this behavior, the tropopause as diagnosed by the PV = 2.0 PVU locates near 10 km. The profile data averaged over all airplane ascents and descents are plotted in the bottom panel of Fig. 6, and statistical analysis of these profiles is included in Table 3 for O₃ and Table 4 for RH. Similar to the evaluation results for ozonesondes, the model could reasonably capture observed O₃ and RH profiles, but O₃ mixing ratios are generally underestimated and RH is overestimated.

The observed and modeled tropospheric column O₃ are compared in Fig. 7. The observed latitudinal gradients in tropospheric column O₃ with values greater than 40 D.U. over mid-latitudes, column values around 30 D.U. over high- and low-latitudes, and values below 20 D.U. over the Pacific Ocean near the equator are captured well by H-CMAQ. To illustrate the differences between observations and simulations, the normalized bias is also shown in Fig. 7. This normalized bias map shows model tropospheric column O₃ overestimation over Russia and Africa and a slight underestimation over the Pacific Ocean. While the comparison with surface observations from WDCGG shows model underestimation at four sites over eastern Europe, the model slightly overestimates tropospheric column O₃ in this region. In addition, the model underestimation especially in the free-troposphere is noted through comparison with ozonesonde measurements (Table 3); however, this comparison showed model overestimation. The evaluation of satellite data compared to ozonesonde exhibited scattered correspondence and slight overestimation by satellite derived column O₃. Therefore the model performance could differ from that for column O₃. The results of the statistical analysis for tropospheric column O₃ are also listed in Table 3. The mean of observed and modeled tropospheric column O₃ across Northern Hemisphere is close on average, with an R of 0.65, an NMB of 4.7%, and an NME of 13.5%. The performance of tropospheric column O₃ judged based on the evaluation protocol developed for mixing ratios, suggests that the model satisfies the performance criteria proposed by Emery et al. (2017).

3.2 Air Mass Characterization Method

In order to characterize whether O_3 in a given air mass is dominated by photochemistry or stratospheric intrusion, and further estimate the impacts of STT, a new air mass characterization method is established here. The flowchart the air mass characterization method is illustrated in Fig. 8. The method relies on modeled O_3PV/O_3 ratio to calculate the intensity of photochemistry. Because the top layer is set to 50 hPa in these H-CMAQ simulations, the uppermost layer (layer number is 44) is always regarded as stratospheric air mass in this method. For all layers below (i.e., layer 43 down to the lowermost layer), the importance of photochemistry is determined based on the ratio of the O_3PV and O_3 mixing ratios. As noted in the discussion related to Figs. 4 and 5, if the O_3 mixing ratio is higher than the O_3PV mixing ratio, it implies that photochemical production affected the air mass, and vice versa. Therefore, a O_3PV/O_3 ratio of less (more) than 1.0, is classified as photochemical production (destruction), and a value near 1.0 can be classified as weakly impacted by photochemistry. The O_3PV/O_3 ratio is illustrated in Fig. 9 (left) and Fig. S7 (left) in the supplemental material, wherein locations and times colored as orange (blue) represent air masses influenced by photochemical production (destruction), while ratios near 1.0 (range from 0.9 to 1.1) are colored as white. In Fig. 9 and Fig. S7, horizontal lines indicating 750, 500, and 250 hPa are also shown. The next step in the classification scheme is to determine whether an air mass is of stratospheric origin. The concept of a sequential intrusion from upper layers to lower layers is considered. When the grid cell directly above is also diagnosed as stratospheric air mass, the grid is determined as being dominated by stratospheric air mass. Applying this concept of a sequential stratospheric air mass intrusion is repeatedly used in the air mass characterization scheme to determine whether an air mass is dominated by photochemistry or stratospheric intrusion. It is important to note that characterizing a grid cell as being dominated by a process does not mean that other processes do not impact O_3 mixing ratios as well. For example, O_3 in a grid cell near the tropopause can be dominated by stratospheric air mass, but it can also be affected by photochemical production and destruction. Similarly, although O_3 in a grid cell near the surface layer is often dominated by photochemical processes, it can also be affected by stratospheric air mass.

An illustration of applying this method to illustrate the stratospheric intrusion for the six ozonesonde sites in the U.S.A. is presented in Fig. 9 (right) and Fig. S7 (right) in the supplemental material. Above 750 hPa, the stratospheric air mass is dominated and varied day-to-day with downward movement reach to near 750 hPa. These stratospheric intrusion are clearly found in some cases, and the stratospheric air mass reaches to the surface during early to middle April at Trinidad Head (Fig. 9(a)) and early April at Boulder (Fig. 9(b)). It should be noted that since the classification scheme is based on the most dominant process, a grid cell classified as being dominated by photochemistry can still be influenced by stratospheric air. Therefore, these estimated impacts of stratospheric air masses on the troposphere can be viewed as a lower bound.

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3.3 Investigation of Stratospheric Intrusion

The relationship between the model estimated stratospheric contribution to the total tropospheric O₃ column and observed surface O₃ levels at CASTNET sites is investigated in Fig. 10 on monthly average. The grid classified as stratospheric air mass is used to calculate O₃ column from stratospheric intrusion, and the stratospheric air mass in troposphere is estimated.

5 The troposphere is defined as all layers below 250 hPa for the purpose of this analysis. Using data from all CASTNET locations, the relationship shows a slight negative slope by R value of 0.25 with non-significance ($p > 0.05$), indicating that the influence of stratospheric air decreased with increasing surface MD8O₃ mixing ratios. To further focus on this relationship at elevated sites in the U.S.A., the analysis is repeated using data from sites with an elevation higher than 1000 m as listed in Table S3 in the supplemental material. The result shows a slight positive slope by R value of 0.14 with non-significance ($p > 0.05$), which

10 indicates that at elevated sites, STT has a possible effect on surface mixing ratio values. The finding of a negative slope using the entire dataset over the U.S.A. is consistent with a previous investigation focused on relatively-polluted areas over the western U.S.A. such as the Central Valley, Southern California, and Las Vegas (Lin et al., 2012b). For elevated sites, they also reported a positive slope indicating higher contributions of stratospheric air masses during periods of elevated surface O₃. The reason for the relatively weak relation found in this study seems to stem from differences in simulated stratospheric O₃ mixing

15 ratios. Lin et al. (2012b) used the fully-coupled stratosphere-troposphere chemistry model GFDL AM3 which tended to overestimate O₃ mixing ratios; therefore, they employed a bias correction approach (assuming that when the estimated stratospheric contribution exceeds the model bias, the bias is caused entirely by excessive stratospheric O₃) for estimating the stratospheric impacts on surface O₃. On the other hand, the H-CMAQ simulations analyzed in this study tends to underestimate tropospheric O₃ levels, especially during STT events, which may suggest that its estimates of stratospheric contributions to

20 high surface O₃ events may also be too low.

The monthly-averaged spatial distribution of the stratospheric air mass is shown in Fig. 11. It indicates higher stratospheric impacts over high-latitude region, and 5-25% on monthly-average over western U.S.A. This is the result of monthly-average and it is well known that the stratospheric intrusion is event occurred on short-term and then lead to the high surface concentration. Time series of daily averaged stratospheric air mass in troposphere at five ozonesonde sites at contiguous

25 U.S.A. is also shown in Fig. 11. These time series reveal large temporal variations of stratospheric air mass in troposphere. On a monthly-mean basis during April 2010, air masses classified as being dominated by stratospheric intrusion contribute about 5-25% to the total tropospheric O₃ column mass at five of the ozonesonde sites and 25% at Trinidad Head. However, on specific days, O₃ masses from the stratosphere contribute up to 50-90% of the total tropospheric O₃ column mass.

The previous section introduces an approach to identify cases when stratospheric air masses impact tropospheric O₃.

30 Results of the O₃ column mass analysis identify periods in early, middle, and late April 2010 that are affected by stratospheric intrusions over the contiguous U.S.A., and in this section these events are further analyzed. Daily maps of the spatial patterns of the percentage of tropospheric O₃ column mass diagnosed as being of stratospheric origin during (i) early and (ii) late April are presented in Fig. 12 while maps for mid-April are presented in Fig. S8 in the supplemental material. On 5 April, a large

impact from the stratosphere was seen over the western U.S.A. (indicated as point S_A on the map) and covered Trinidad Head where the contribution of O_3 from the stratosphere to the tropospheric O_3 mass is greater than 50%. This air mass moved eastward on 6 April when the impact at Boulder is also greater than 50%. During 7-8 April, this air mass was located over the central U.S.A., and then moved further to the east, with contributed at Huntsville on 9 April. Finally, this air mass further moved towards the northeast U.S.A. with impact at Wallops Island and Rhode Island, respectively. The stratospheric impacts in early April are associated with an air mass movement from west to east within 5 days; corresponding to an average speed of about 8-9 m/sec. Compared to the early April case, the case in late April is different. On 25 April, large impacts from the stratosphere were found over western (marked as S_{B1}) and eastern (marked as S_{B2}) Canada. The S_{B1} air mass moved into the western U.S.A. on 27 April, and had large impacts from 27 to 30 April at Trinidad Head, affected Boulder on 29 and 30 April, and finally moved southwestern U.S.A. in a U-shaped pattern on 30 April. Another air mass of S_{B2} located over eastern Canada on 26 April moved slowly southward and impacted Wallops Island and Rhode Island, then moved eastward. Thus, for the late April case, stratospheric air was present in different air masses impacting different locations on different days rather than a single air mass simply moving from west to east as in the early April case. Contrasting the early and late April cases illustrates that different synoptic flow scenarios influence how stratospheric air can impact tropospheric O_3 columns over the U.S.A. The impacts of STT during the middle of April are shown in Fig. S8 in the supplemental material. From 12-15 April, tropospheric O_3 columns over the western (i.e., Trinidad Head) and eastern (i.e., Wallops Island and Rhode Island) U.S.A. were dominated by stratospheric intrusion, and these impacts were almost disappeared after 17 April. Previous studies (e.g., Lin et al., 2012b) estimated thirteen STT events during April-June 2010, and April 7, 9, 12-15, 21-23, and 28-29 2010 were reported as STT events. Our investigations based on the air mass characterization method matches with these earlier findings. The impact of STT at Huntsville has been investigated by combining ozonesonde and ozone lidar data (Kuang et al., 2012). In their report, the period of 27-29 April was associated with STT. However, STT estimation was limited above 750 hPa (Fig. 9). This is due to the model difficulty to capture the STT event itself as revealed from the comparison with ozonesonde dataset (Fig. 5). As discussed in model evaluation, finer horizontal and vertical resolution could be potential way to improve STT representation.

25 4 Conclusions

In this study, the regional chemical transport model CMAQ recently extended for hemispheric applications, the H-CMAQ, is applied to investigate trans-Pacific transport during April 2010. This part 1 manuscript presents results from comprehensive evaluation and a new air mass characterization method based on these results and ozonesonde measurements. The comparison of modeled and observed O_3 at the surface shows a very good performance with NMBs around -10%. Comparisons of vertical O_3 distributions against ozonesonde and aircraft-based observations show that the model can capture well O_3 variations within boundary layer similar to those at the surface, although systematic underestimations of free troposphere O_3 occur with NMBs up to -30%, especially during events that are characterized to have strong STT during late April. Modeled RH exhibits a positive bias with NMBs of +10% or greater at all altitudes. Comparisons of modeled

tropospheric O₃ column with satellite observations suggest that the model can represent the general feature with lower column O₃ over the Pacific Ocean near equator and higher column in the mid-latitudes.

Using ozonesonde measurements, the relationship between PV and RH is examined to characterize stratospheric air masses. The PV-RH relation indicates that PV of 2.0 PVU (1 PVU = 10⁻⁶ m² K kg⁻¹ s⁻¹) generally corresponds to RH values of 30-40%. A new air mass characterization method is further developed based on the ratio of modeled O₃ and stratospheric O₃ tracer mixing ratios to examine the relative importance of photochemistry, and PV and RH to examine the stratospheric air mass. The estimated STT impacts show significant day-to-day variations both in the magnitude of the contribution and the origin of the air mass. The relationship between surface O₃ levels and estimated stratospheric air mass in troposphere exhibits a slight negative slope, indicating that at most locations, high surface O₃ mixing ratios typically result from other factors (e.g., emissions). In contrast, at elevated sites the relationship exhibits a slight positive slope, indicating a steady STT contribution to O₃ levels at these locations.

Despite the use of a coarse horizontal grid resolution for H-CMAQ simulations in this work, it was found that the model performance at the surface and in the boundary layer are reasonably within the model performance criteria suggested from regional-scale applications. However, this work also shows that the model has difficulty capturing higher O₃ mixing ratios in the free troposphere through the comparison with ozonesonde data. This result suggests a need for model improvements to accurately represent the STT process. This study focuses on April 2010, and monthly or seasonal behavior of STT, and interannual variations based on the long-term trend analysis should also be considered for future study. The Part 2 paper will focus on other factors that affect surface O₃ mixing ratio, namely emissions, also examine the relative importance of NO_x and VOCs.

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Code availability

Source code for version 5.2 of the CMAQ model can be downloaded from <https://github.com/USEPA/CMAQ/tree/5.2>. For further information, please visit the US Environmental Protection Agency website for the CMAQ system: <https://www.epa.gov/cmaq>.

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Data availability

The observational datasets used in this study are available from their respective websites: <http://ds.data.jma.go.jp/gmd/wdogg/> (WDCGG), <http://www.eanet.asia/index.html> (EANET), and <https://www.epa.gov/castnet> (CASTNET) for surface observation network, <https://woudc.org/home.php> (WOUDC) and <https://www.esrl.noaa.gov/gmd/ozwv/ozsondes/> (NOAA ESRL) for ozonesonde, <https://www.esrl.noaa.gov/gmd/ozwv/aircraft/index.html> (NOAA-ESRL) for airplane, https://acd-ext.gsfc.nasa.gov/Data_services/cloud_slice/index.html (NASA). Last Access: 31 August 2018.

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Competing interests

The authors declare that they have no conflict of interest.

Disclaimer

- 5 The views expressed in this paper are those of the authors and do not necessarily reflect the views or policies of the U.S. Environmental Protection Agency.

Author contributions

- 10 Syuichi Itahashi performed the analysis of observation and model simulation and prepared the manuscript with contributions from all co-authors. Rohit Mathur and Christian Hogrefe contributed to establish the hemispheric modeling application for this study and prepared the emission dataset and initial condition from previous long-term simulation results. Yang Zhang contributed to the literature review of trans-Pacific transport and refined this research through simulation designs, and results interpretation.

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5 **Webpages**

Acid Deposition Monitoring Network in East Asia (EANET): <http://www.eanet.asia/index.html>, Last Access: 19 April 2018.

Clean Air Status and Trends Network (CASTNET), U.S. Environmental Protection Agency Clean Air Markets Division:

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National Centers for Environmental Prediction (NCEP), National Center for Atmospheric Research (NCAR), Final (FNL) Operational Global Analysis data: <https://rda.ucar.edu/datasets/ds083.2/>, Last Access: 15 October 2018.

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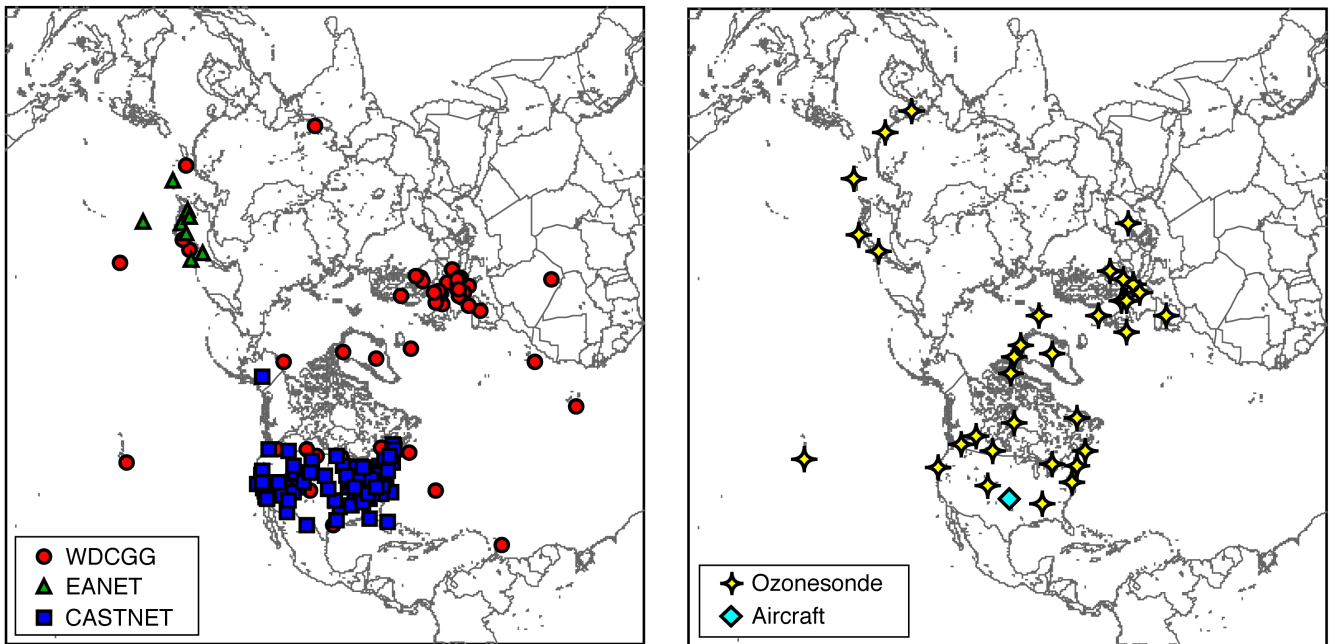


Figure 1. Geographical mapping of the (left) surface and (right) aloft observational sites used in this study. Detailed information about the ozonesonde observation sites is provided in Table 1.

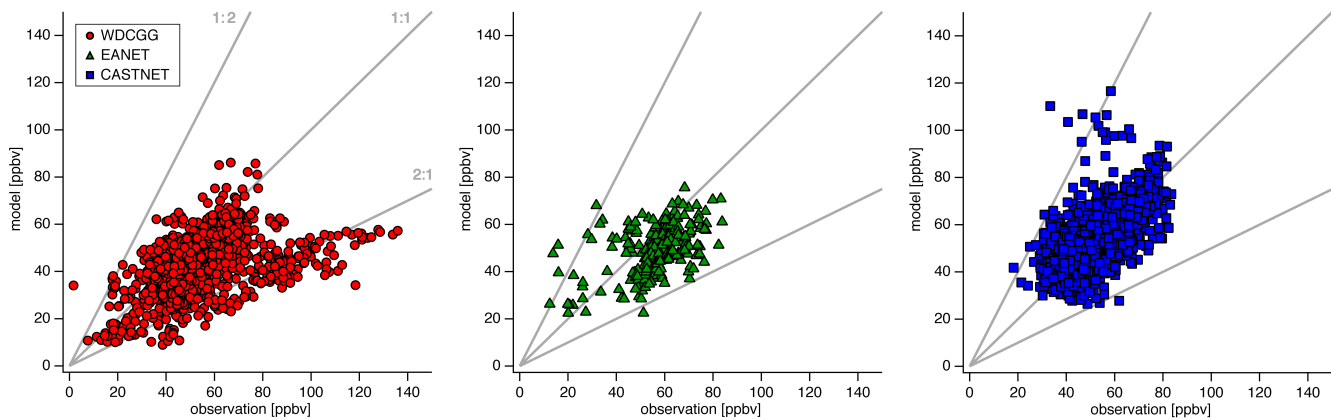


Figure 2. Scatter-plot between observations and H-CMAQ simulations for surface MD8O3 during April 2010. Reference lines are provided at ratios of 2:1, 1:1 and 1:2. The symbols and colors used to represent the different surface observational datasets are consistent with Fig. 1.

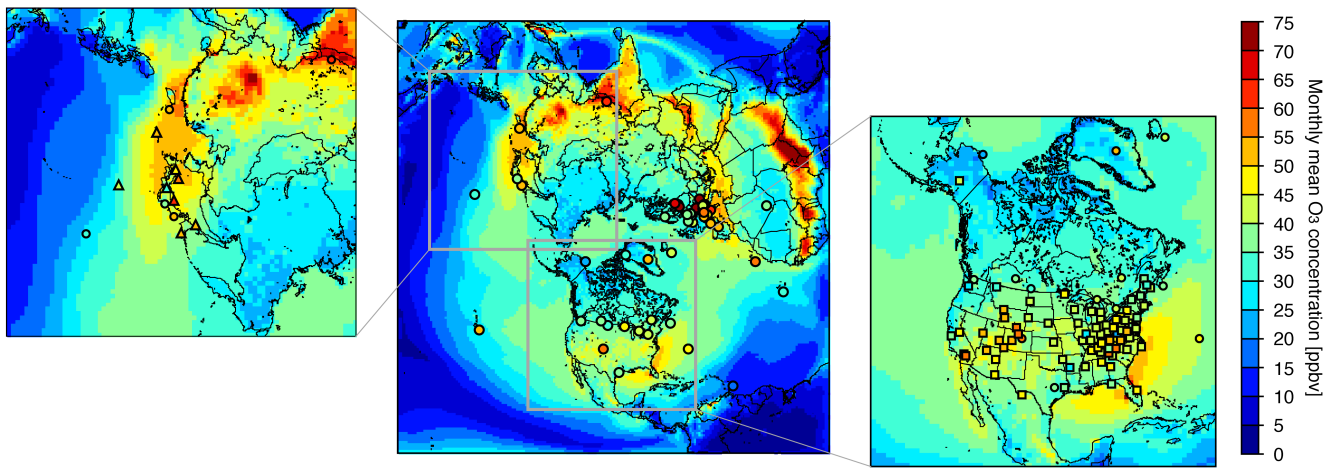


Figure 3. Monthly-mean H-CMAQ O₃ mixing ratios overlaid with WDCGG surface observations, and zoom-in panels over (left) Asia overlaid with EANET surface observations, and (right) U.S. overlaid with CASTNET surface observations.

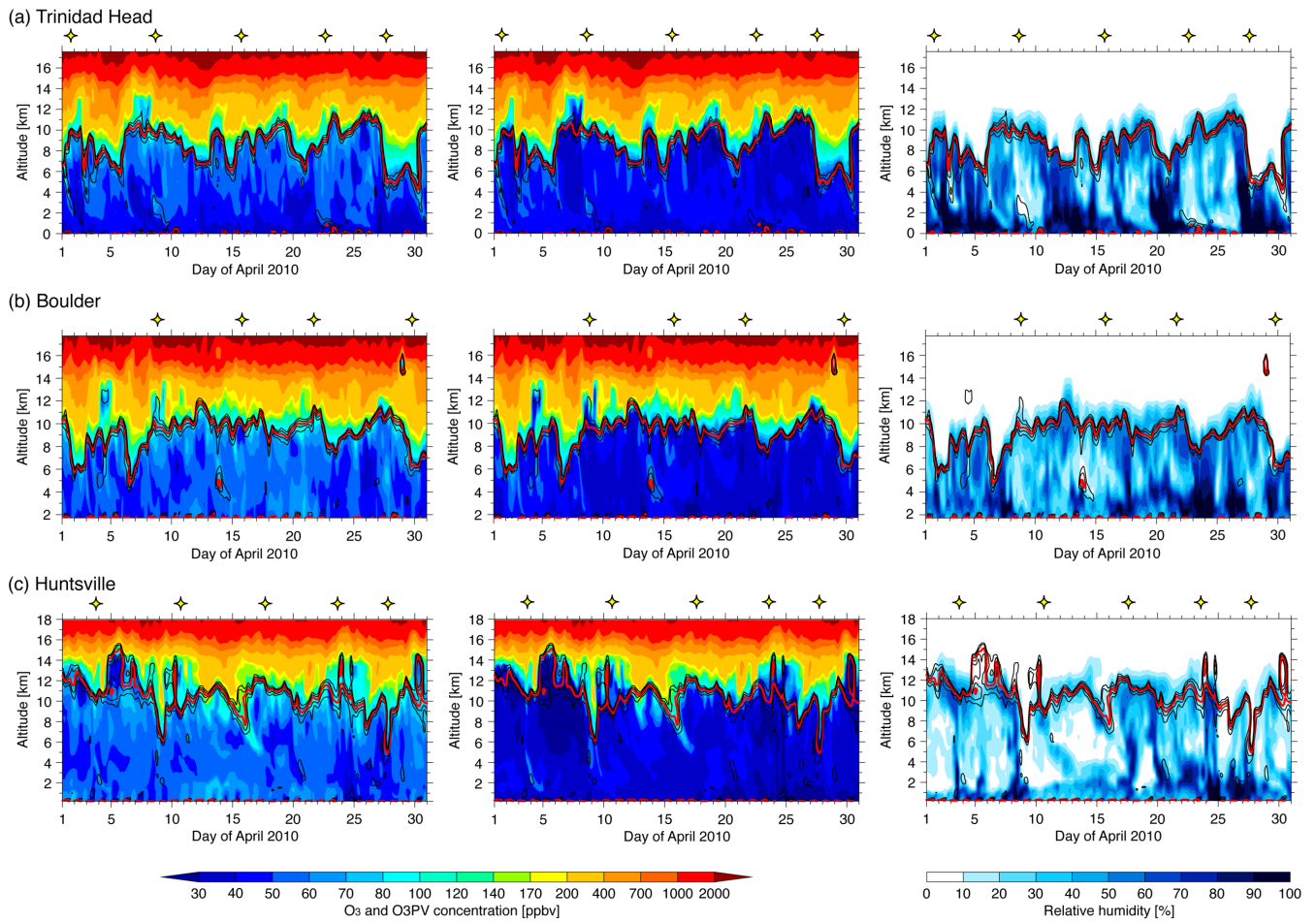


Figure 4. Curtain plots of modeled (left) O_3 , (center) O_3PV , and (right) RH at U.S. ozonesonde sites of (a) Trinidad Head (CA), (b) Boulder (CO), and (c) Huntsville (AL) during April 2010. Yellow stars indicate the time of available ozonesonde measurements. Contour lines of modeled PV are also inserted 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.

5

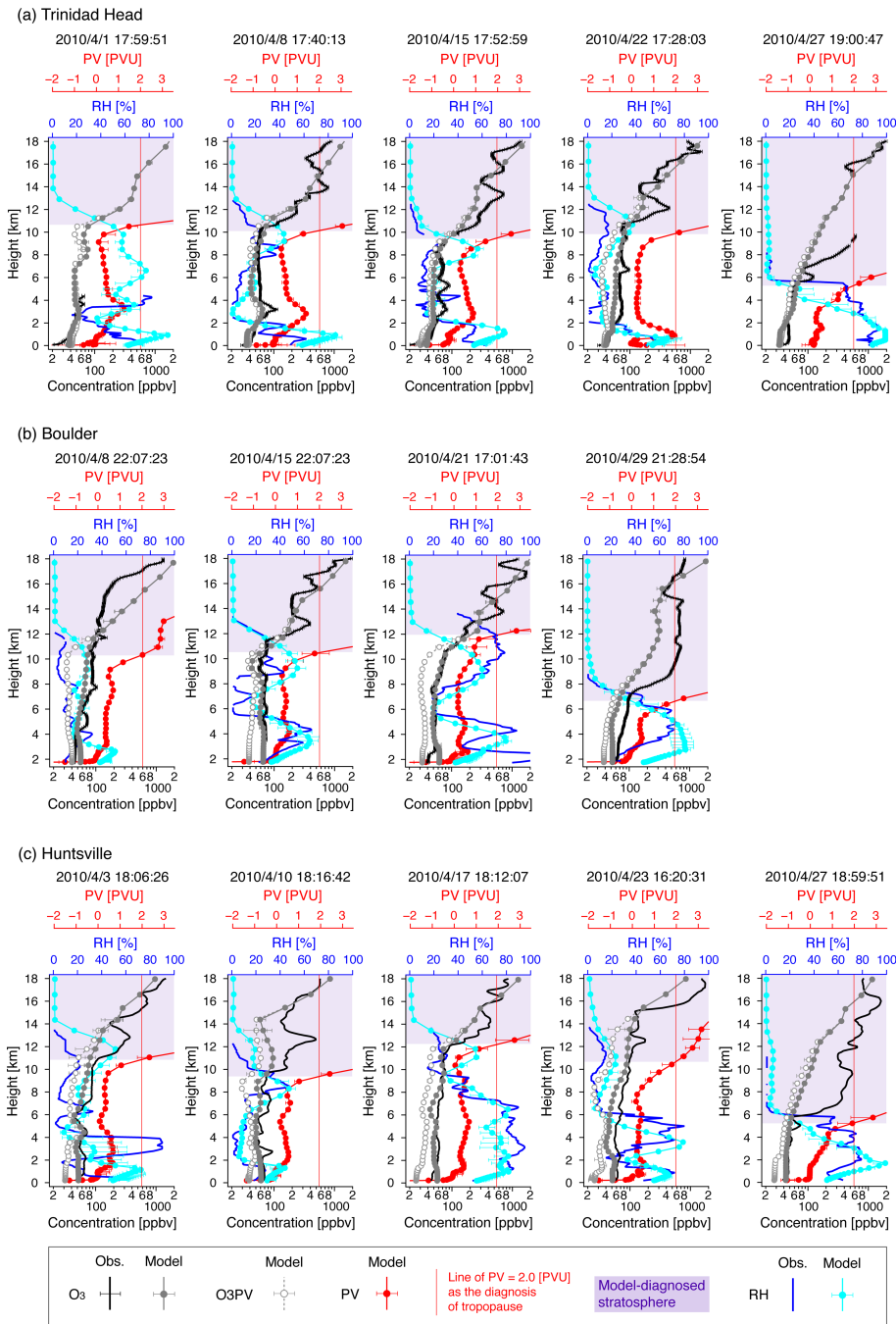


Figure 5. Vertical profiles of observed and modeled O₃ and RH at U.S. ozonesonde sites of (a) Trinidad Head (CA), (b) Boulder (CO), and (c) Huntsville (AL). Also see Figure 4 for ozonesonde measurement times. For modeled O₃ and RH, the hourly result corresponding to the ozonesonde measurement time is shown by circles, and the maximum and minimum model results within ± 2 hours of the measurement time are shown by whiskers. For observed O₃ at Hilo and Boulder, the range of uncertainties of the O₃ observations is shown by whiskers. Modeled O₃PV 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.

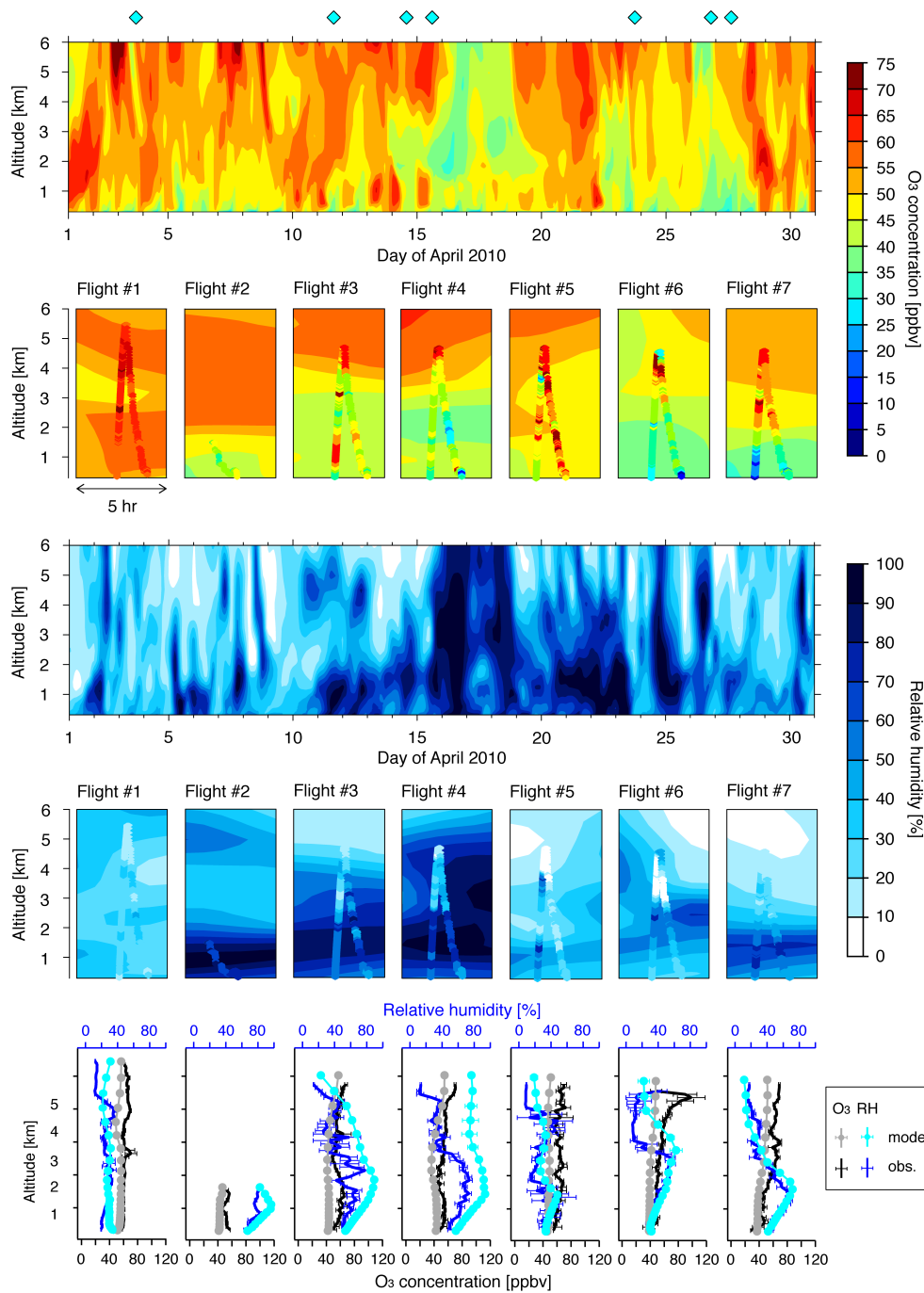


Figure 6. Curtain plot and vertical profiles of O₃ and RH at the aircraft observational site: Southern Great Plains, Oklahoma. Gray diamonds indicate the times of the seven aircraft flights, and results for these flights are shown in the expanded boxes for 5-h time windows overlaid with observations. Observed ascent and descent profile data are averaged into 100-m grid resolution, and profiles of the mean and standard deviations are shown. Modeled ascent and descent data corresponding to the observation times are averaged on original modeled layers, and the mean and standard deviations are shown.

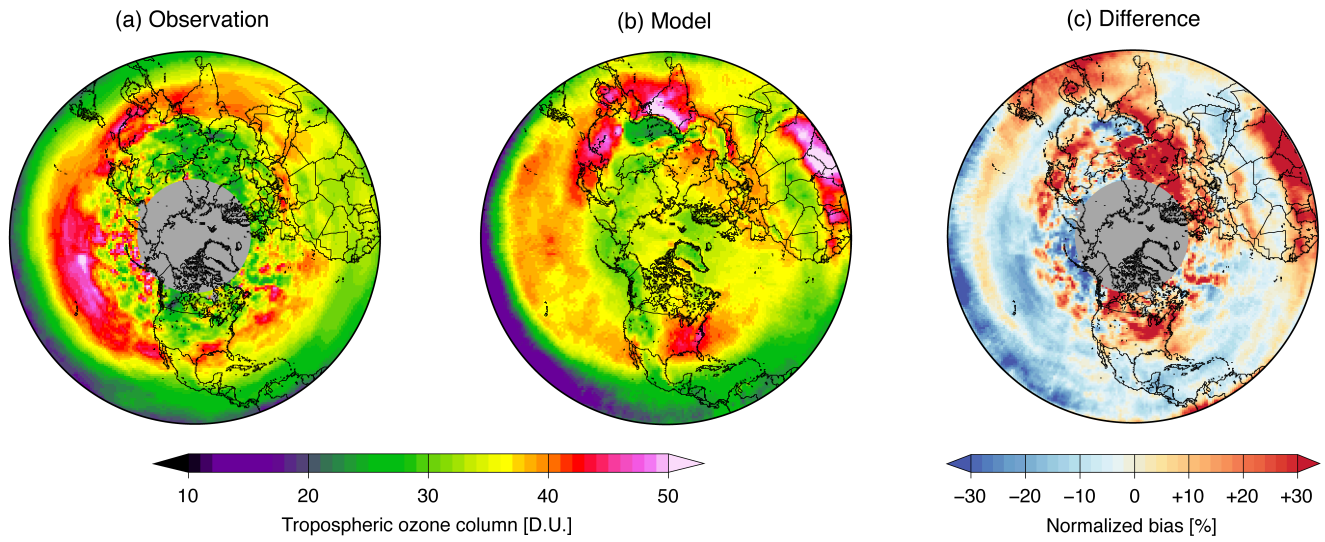


Figure 7. Tropospheric ozone column of (a) satellite observation, (b) H-CMAQ simulation, and (c) their differences shown as normalized bias. Areas filled in gray colored indicate missing observations.

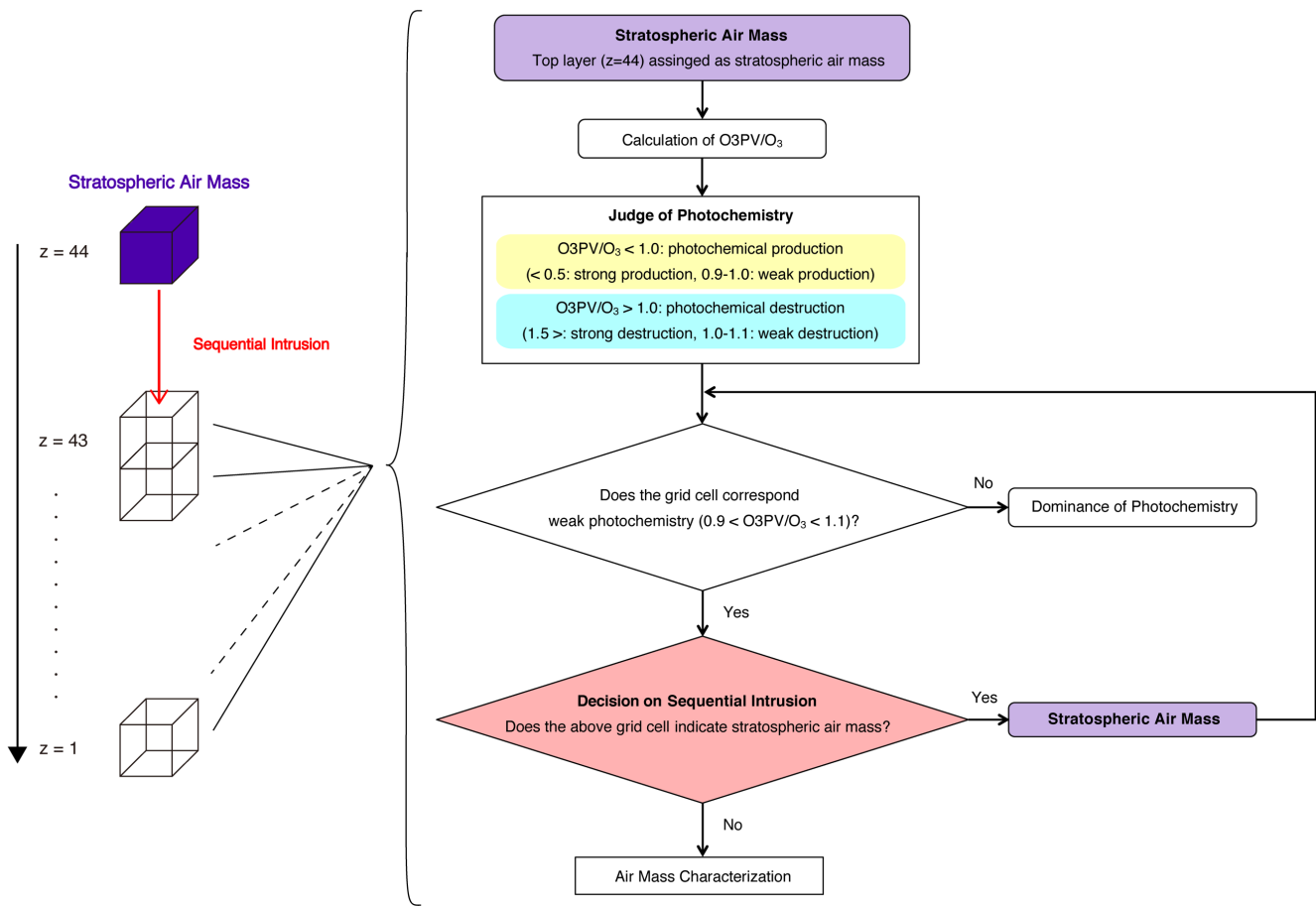


Figure 8. Flowchart of the air mass characterization scheme. See the text for additional details.

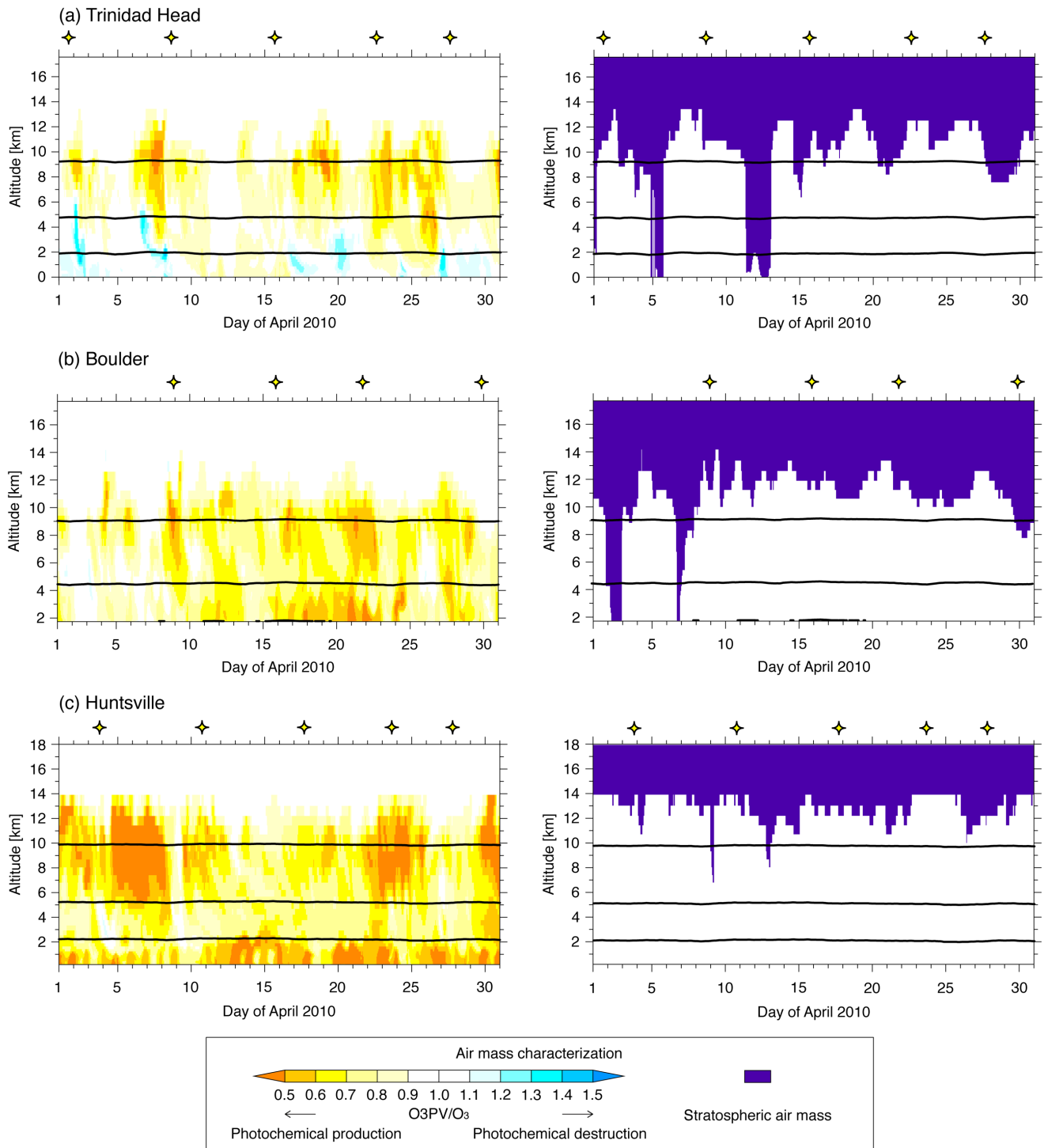
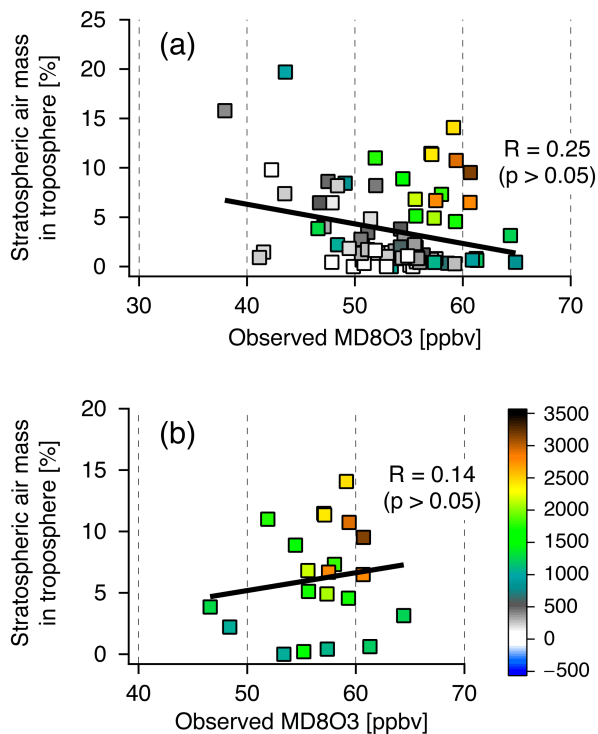


Figure 9. Curtin plot of model-diagnosed air mass characterization for (left) O₃PV/O₃ and (right) stratospheric air mass at U.S. ozonesonde sites of (a) Trinidad Head (CA), (b) Boulder (CO), and (c) Huntsville (AL) during April 2010.



5 **Figure 10. Relationship between observed MD803 at the surface and the estimated stratospheric air mass contributed to total tropospheric O₃ column mass as monthly-average during April 2010. The points are color coded based on elevation of each site. (a) All CASTNET sites (except Alaska), and (b) elevated CASTNET sites defined as having an elevation greater than 1000 m (see also Table S3).**

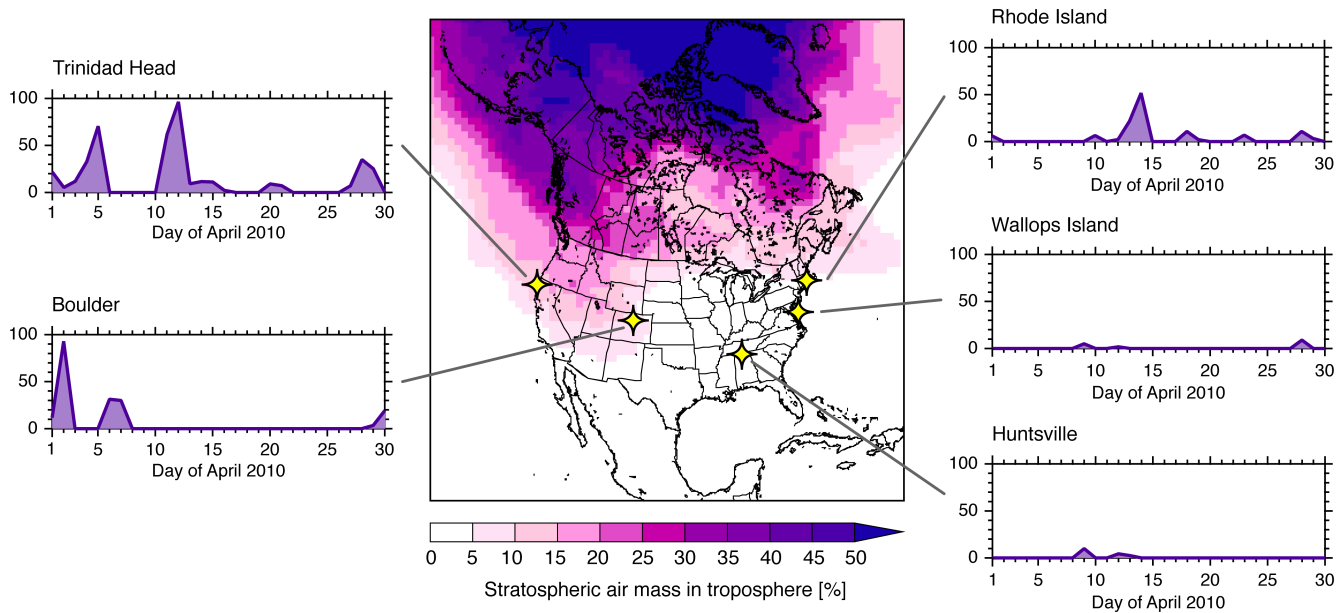
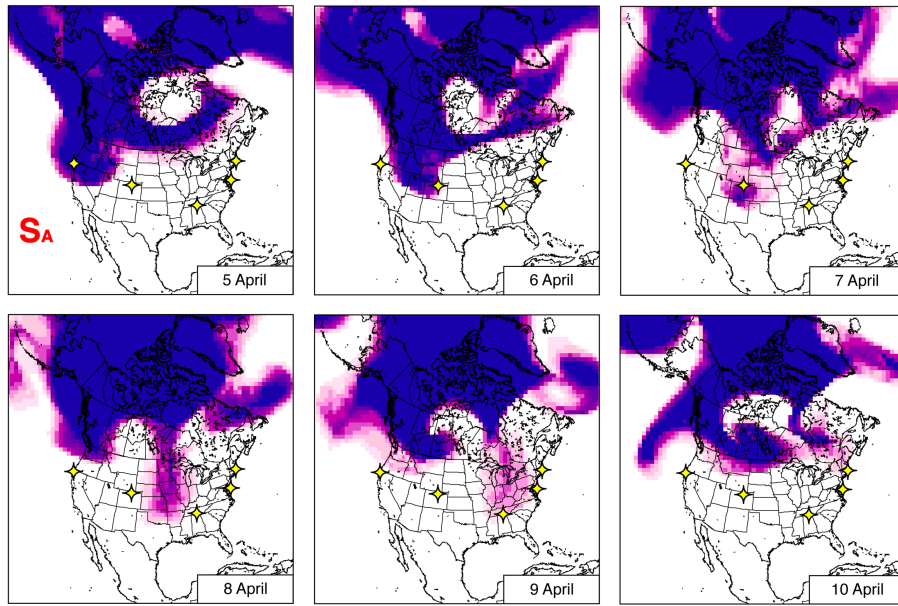


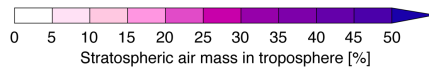
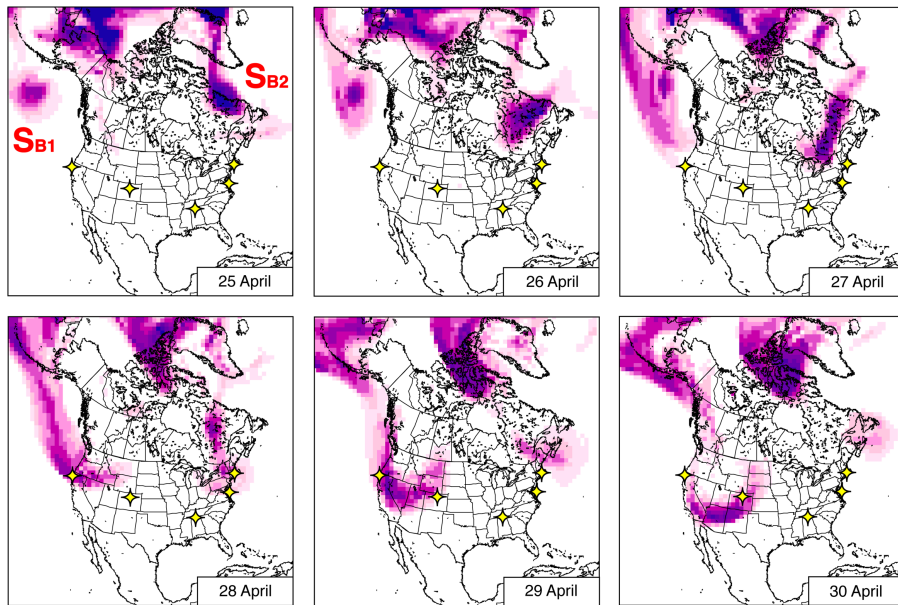
Figure 11. Spatial distributions of monthly-averaged stratospheric air mass contributions to total tropospheric O₃ column mass over the U.S. during April 2010. Yellow stars indicate the ozonesonde observational sites, and the daily variations are plotted.

5

(i) Early April



(ii) Late April



5 **Figure 12.** Spatial distributions of day-to-day variations of stratospheric air mass contributions to total tropospheric O₃ column mass over the U.S. during (i) early, and (ii) late April 2010. Yellow stars indicate the ozonesonde observational sites. Red character strings (S_A and S_{B1}-S_{B2}) denote the different stratospheric air masses discussed in the text.

Table 1. Literature review of previous studies on estimated impacts of (upper) emissions from Asia, and (lower) stratospheric intrusion.

Time Period	Estimated impacts	Location	Method	Reference
April 2001	1 ppbv (monthly mean), up to 2.5 ppbv (daily mean)	western U.S.	model (process analysis)	Wang et al. (2009)
April-May 2002	10 ppbv increase from April-May 1984	5 west coast U.S. sites	observation (linear regression)	Jaffe et al. (2003)
April-May 2006	5-7 ppbv (17 April-15 May 2006; INTEX-B)	western North America	model (zero-out ^a)	Zhang et al. (2008)
	increase the above influence by 1-2 ppbv	western North America	model (doubling NO _x emissions from eastern Asia)	
	2-5 ppbv (17 April-15 May 2006; INTEX-B)	eastern North America	model (zero-out ^a)	
April-June 2010	2-6 ppbv increase from April-June 1985	western U.S.	model (tripling Asian emissions from 1985)	Jacob et al. (1999)
	1-3 ppbv increase from April-June 1985	eastern U.S.	model (tripling Asian emissions from 1985)	
May-June 2010	8-15 ppbv on specific events	high-elevation regions	model (zero-out)	Lin et al. (2012a)
April-June 2010	4.7±2.4 ppbv (three-month mean)	15 high-elevation western U.S. sites	model (zero-out)	Lin et al. (2012b)
March-October 2001	below 20 ppbv	U.S.	tagged O ₃ ^b	Fiore et al. (2003)
April-May 1990-2012	10-25 ppbv	22 high-elevation western U.S. sites	tropopause tracer ^c	Lin et al. (2015)
April-June 2010	22.3±11.5 ppbv (three-month mean)	15 high-elevation western U.S. sites	tropopause tracer ^c	Lin et al. (2012b)

Note: ^a: Estimate the impact from the difference between the standard simulation and a simulation with eastern Asian anthropogenic sources shut off. ^b: This tagged method divides simulated O₃ into individual O₃ tracer to track O₃ produced in different region. ^c: This tracer method account for STT contribution to O₃ using e90 tracer, which differentiates tropospheric air mass based on the globally uniform surface source and 90-day folding lifetime; both have been spun-up for three years.

Table 2. Details of the ozonesonde dataset used in this study.

Country	Site name	Longitude (°)	Latitude (°)	Elevation (m a.s.l.)	Data source	# of launch
USA (HI)	Hilo	-155.05	19.72	10	NOAA ESRL	3
USA (CA)	Trinidad Head	-124.15	41.06	36	NOAA ESRL	5
USA (CO)	Boulder	-105.20	39.95	1743	NOAA ESRL	4
USA (AL)	Huntsville	-86.65	34.73	203	NOAA ESRL	5
USA (VA)	Wallops Island	-75.47	37.93	13	WOUDC	6
USA (RI)	Rhode Island	-71.42	41.49	21	NOAA ESRL	2
Canada (BC)	Kelonwa	-119.4	49.94	456	WOUDC	4
Canada (AB)	Edmonton	-114.1	53.54	766	WOUDC	4
Canada (SK)	Bratt's Lake	-104.7	50.2	580	WOUDC	4
Canada (NU)	Resolute	-94.97	74.71	46	WOUDC	2
Canada (MB)	Churchill	-94.07	58.74	30	WOUDC	4
Canada (NU)	Eureka	-85.94	79.98	10	WOUDC	3
Canada (ON)	Egbert	-79.78	44.23	252	WOUDC	4
Canada (NS)	Yarmouth	-66.11	43.87	9	WOUDC	3
Canada (NU)	Alert	-62.34	82.49	75	WOUDC	3
Canada (NL)	Goose Bay	-60.36	53.3	36	WOUDC	3
Vietnam	Hanoi	105.8	21.01	7	WOUDC	1
China	Hong Kong	114.17	22.31	66	WOUDC	4
Japan	Naha	127.69	26.21	28	WOUDC	3
Japan	Tateno	140.13	36.06	31	WOUDC	3
Japan	Sapporo	141.33	43.06	26	WOUDC	3
Greenland	Summit	-38.46	72.58	3211	NOAA ESRL	4
Ireland	Valentia	-10.25	51.93	14	WOUDC	3
Spain	Madrid	-3.58	40.47	631	WOUDC	3
UK	Lerwick	-1.19	60.14	80	WOUDC	5
Belgium	Uccle	4.35	50.8	100	WOUDC	12
Netherland	De Bilt	5.18	52.1	4	WOUDC	8
Switzerland	Payerne	6.57	46.49	491	WOUDC	12
German	Hohenpeissenberg	11.0	47.8	976	WOUDC	18
Norway	Ny Alesund	11.95	78.93	11	WOUDC	6
Czech	Praha	14.44	50.0	304	WOUDC	10
Poland	Legionowo	20.97	52.4	96	WOUDC	4
Turkey	Ankara	32.86	39.97	890	WOUDC	2

Note: Parenthesis after the country name indicates the state.

Table 3. Statistical analysis of modeled O₃ concentration using surface, ozonesonde, aircraft, and satellite observations.

	N	Mean		R	NMB	NME
		Observation	Model			
Surface						
–WDCGG	1498	53.9	43.5	0.49 ^{***}	–19.3%	23.7%
–CASTNET	2316	53.4	52.9	0.61 ^{***}	–0.9%	12.6%
–EANET	240	56.2	49.1	0.49 ^{***}	–12.6%	20.6%
Ozonesonde						
USA and Canada						
–boundary layer	1016	46.0	42.7	0.70 ^{***}	–7.1%	16.7%
–free troposphere	893	87.7	59.6	0.79 ^{***}	–32.1%	33.8%
–upper model layer	512	905.1	770.3	0.91 ^{***}	–14.9%	30.2%
Asia						
–boundary layer	283	44.2	47.4	0.44 ^{***}	7.1%	24.5%
–free troposphere	207	70.7	59.0	0.43 ^{***}	–16.5%	21.5%
–upper model layer	124	529.8	399.2	0.94 ^{***}	–24.6%	34.4%
Europe						
–boundary layer	1478	47.6	46.8	0.42 ^{***}	–1.6%	17.9%
–free troposphere	1368	78.2	57.2	0.76 ^{***}	–26.8%	29.1%
–upper model layer	817	1015.7	894.3	0.94 ^{***}	–11.9%	24.2%
Aircraft						
–from sufrage upto 6 km	128	55.9	45.3	0.74 ^{***}	–19.0%	19.1%
Satellite						
–Tropospheric column	28020	33.2	34.7	0.65 ^{***}	4.7%	13.5%

Note: The unit of mean for observations and simulations is ppbv except satellite observation expressed as D.U. Maximum daily 8 hour average ozone (MD8O3) is used for surface observational data of WDCGG, CASTNET, and EANET. Corresponded hourly modeled O₃ is used for ozonesonde data. 2-4 hours averaged hourly modeled O₃ is used for aircraft data to fully cover each observation time. 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.

Table 4. Statistical analysis of modeled RH using ozonesonde and aircraft observations.

	N	Mean		R	NMB	NME
		Observation	Model			
Ozonesonde						
USA and Canada						
-boundary layer	1016	57.70	67.07	0.73***	16.2%	24.5%
-free troposphere	881	39.16	43.31	0.83***	10.8%	29.7%
-upper model layer	398	7.81	8.72	0.79***	11.6%	62.3%
Asia						
-boundary layer	283	65.89	79.63	0.45***	20.8%	28.7%
-free troposphere	184	46.26	51.52	0.38***	11.4%	47.7%
-upper model layer	43	18.96	26.47	0.63***	39.6%	67.2%
Europe						
-boundary layer	1485	63.84	68.92	0.73***	8.0%	17.1%
-free troposphere	1368	36.14	42.82	0.80***	18.5%	32.6%
-upper model layer	679	7.13	9.56	0.91***	34.1%	56.1%
Aircraft						
-troposphere	126	41.66	52.04	0.84***	24.9%	28.7%

Note: 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. 5-hour averaged hourly modeled relative humidity is used for ozonesonde data. 2-4 hours averaged hourly modeled relative humidity is used for aircraft data to fully cover each observation time, and original aircraft data are averaged into 100-m resolution to be compared with model.

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