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LETTER

Stratospheric contribution to the summertime high surface ozone events over the western united states

Xinyue Wang\textsuperscript{1,2}*, Yutian Wu\textsuperscript{1}, William Randel\textsuperscript{1} and Simone Tilmes\textsuperscript{2}

\textsuperscript{1} Department of Earth, Atmospheric, and Planetary Sciences, Purdue University, West Lafayette, IN, United States of America

\textsuperscript{2} Atmospheric Chemistry, Observations and Modeling Laboratory, National Center for Atmospheric Research, Boulder, CO, United States of America

\textsuperscript{3} Lamont-Doherty Earth Observatory of Columbia University, Palisades, NY, United States of America

E-mail: wang2807@purdue.edu

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Abstract

The stratospheric influence on summertime high surface ozone (O\textsubscript{3}) events is examined using a twenty-year simulation from the Whole Atmosphere Community Climate Model. We find that O\textsubscript{3} transported from the stratosphere makes a significant contribution to the surface O\textsubscript{3} variability where background surface O\textsubscript{3} exceeds the 95th percentile, especially over western U.S. Maximum covariance analysis is applied to O\textsubscript{3} anomalies paired with stratospheric O\textsubscript{3} tracer anomalies to identify the stratospheric intrusion and the underlying dynamical mechanism. The first leading mode corresponds to deep stratospheric intrusions in the western and northern tier of the U.S., and intensified northeasterlies in the mid-to-lower troposphere along the west coast, which also facilitate the transport to the eastern Pacific Ocean. The second leading mode corresponds to deep intrusions over the Intermountain Regions. Both modes are associated with eastward propagating baroclinic systems, which are amplified near the end of the North Pacific storm tracks, leading to strong descents over the western U.S.

1. Introduction

Surface ozone (O\textsubscript{3}) adversely affects human health and the ecosystem because of its high oxidation capability (U.S. Environmental Protection Agency, 2015). The risk of O\textsubscript{3} pollution on mortality is also significantly raised by high temperatures (Levy and Patz 2015). During summer, surface O\textsubscript{3} level maximizes over the western U.S. (Gaudel et al. 2018), mainly attributed to the combination of active photochemical production and noncontrollable sources, such as intercontinental pollution transport, lightning, and wildfire events (Fiore et al. 2002, Jaffe et al. 2018). Downward transport of O\textsubscript{3} during stratospheric intrusions is also considered to be a contributing factor during summertime (Danielsen 1980, Lefohn et al. 2011, Lefohn et al. 2012, Zanis et al. 2014, Akritidis et al. 2016, Yang et al. 2016, Skerlak et al. 2019). The transport is achieved irreversibly by a tongue-like structure containing high stratospheric O\textsubscript{3} extruding downward, folding into the tropospheric air and descending toward the surface (Danielsen 1968, Johnson and Viezee 1981). When a stratospheric intrusion contributes high O\textsubscript{3} to the surface, in addition to that produced by anthropogenic pollution, it could easily push the surface O\textsubscript{3} values beyond the National Ambient Air Quality Standard threshold 70 ppbv (Langford et al. 2017, Skerlak et al. 2019). Observational and modeling studies have shown that surface O\textsubscript{3} extremes that are directly associated with downward transport from the stratosphere preferentially occur in the western U.S. (Stohl et al. 2003, Lin et al. 2012, Lin et al. 2015, Skerlak et al. 2014). Consequently, the joint effects of chemistry and episodic stratospheric transport make the western U.S. a hot spot of O\textsubscript{3} pollution in summer.

Due to the large dynamic variability of the tropopause, limited temporal and spatial extent of measurements, and mixing with tropospheric air, the observations of transport due to stratospheric intrusions are challenging (Stohl et al. 2003). In addition, most of the previous studies focused on springtime stratospheric influence because tropopause O\textsubscript{3} abundances and downward air mass fluxes maximize...
during that time (Langford 1999, Prather et al 2011, Langford et al 2009, Langford et al 2012, Lin et al 2012, Lin et al 2015, Langford et al 2017, Albers et al 2018). The linkage between summertime stratospheric intrusion and high surface O₃ events over the western U.S. has received less attention (Lefohn et al 2011, Lefohn et al 2012). By analyzing the output of a state-of-the-art chemistry climate model implemented with an artificial stratospheric ozone tracer (O₃S), we aim to 1. estimate the contribution of O₃ reaching the surface associated with summertime stratospheric intrusions, 2. understand the space-time behavior of stratospheric intrusion events, and 3. clarify the underlying dynamical mechanism.

2. Methods

2.1. CESM2(WACCM6) and O₃S Diagnostic

We analyzed daily surface O₃ and stratospheric O₃ tracer (denoted by O₃S) from 1995 to 2014 summer months (June–August, JJA) using the Whole Atmosphere Community Climate Model version 6 (WACCM6) of the Community Earth System Model version 2 (CESM2). It is the high-top version of the Community Atmosphere Model version 6 (CAM6), integrating the atmospheric physics and chemistry from the surface to nearly 140 km. The WACCM6 uses the same atmospheric physics as CAM6. The chemical mechanism includes comprehensive troposphere, stratosphere, mesosphere and lower thermosphere chemistry, described by Emmons et al (2020).

The standard emissions are based on anthropogenic and biomass burning inventories specified for the Coupled Model Intercomparison Project 6 (CMIP6). WACCM6 is coupled to the interactive Community Land Model version 5 (CLM5), which handles dry deposition. The simulations shown here are fully coupled ocean-atmosphere experiment, and feature global monthly climatologies as well as stratospheric deposition. The simulations shown here are fully coupled ocean-atmosphere experiment, and feature global monthly climatologies as well as stratospheric deposition. WACCM6 uses the same atmospheric physics as CAM6. The chemical mechanism includes comprehensive atmosphere, stratosphere, mesosphere and lower thermosphere chemistry, described by Emmons et al (2020).

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2.2. Maximum Covariance Analysis (MCA)

Maximum Covariance Analysis (MCA), known as Singular Value Decomposition (SVD) analysis, is a useful tool for detecting coherent patterns between two different geophysical fields (e.g. (Bretherton et al 1992, Hurrell 1995, Dai 2013)). In this study, we isolate pairs of spatial patterns and corresponding time series by performing the eigenanalysis on the temporal covariance matrix between O₃ anomalies and O₃S anomalies at the lowest model level (detailed calculations are discussed in (Bretherton 2015)). Daily anomalies of O₃ and O₃S at the lowest model level are derived with respect to the twenty-year (1995–2014) mean of that day. The considered domain in this study is 20°–50°N, 70°–140°W.

3. Results

3.1. Evaluation of CESM2 (WACCM6)

Here we evaluate the WACCM6 simulations against observations from the Tropospheric Ozone Assessment Report (TOAR) (Schultz et al 2017). Figure 1(a) is the 1995–2014 mean of ‘all_{mean}’ variables from the TOAR. 5° × 5° (latitude × longitude) monthly median products. High O₃ values (∼45 nmol mol⁻¹) are seen over the western U.S. in TOAR measurements. For WACCM6, we gridded the simulations onto the horizontal grid of TOAR data and calculated median O₃ concentrations using the same metrics to guarantee an apple-to-apple comparison. Generally a good agreement is found between the model and observations over the Central U.S. and most of the West Region, with percentage differences within 15% (see figure S1 (https://stacks.iop.org/ERL/15/1040a6/mmedia)). However, WACCM6 overestimates the surface median O₃ over the southeast U.S. by 55% (∼15 nmol mol⁻¹), which is consistent with the evaluation in Emmons et al (2020). The biased O₃ over the eastern U.S. is a long existing problem that can have various reasons starting with still insufficient complexity of the chemistry, but also the model resolution, deposition scheme, etc (Schwantes et al 2020).

Figure 1(c) and (d) shows the 1995–2014 mean of daily O₃ and O₃S simulations in 0.95° × 1.25° (latitude × longitude) resolution at the lowest model level, respectively. Figure 1(c) is similar to figure 1(b) and shows that high O₃ are concentrated over the western U.S. As shown in 1(d), strong stratospheric impact, ranging from 6 to 10 nmol/mol, is found over the Canada-U.S. border and the Western States, including southern British Columbia.
Our model simulation is in agreement with previous observational studies which reported that deep stratospheric intrusions preferentially occur in the West and the Intermountain West (Brioude et al. 2007, Bourqui and Trépanier 2010, Ambrose et al. 2011, Lefohn et al. 2011, Lefohn et al. 2012, Lin et al. 2012, Langford et al. 2012, Langford et al. 2015, Langford et al. 2017, Clark and Chiao 2019). A minimum stratospheric impact occurs in the Southeast, in good agreement with results in Lin et al. (2012). These features are also consistent with simulations by the Geophysical Fluid Dynamics Laboratory global chemistry-climate model (Clifton et al. 2014). Additionally, the simulated stratospheric impact in late spring (figure S2) agrees well with that shown in figure 11(c) and (d) from Lin et al. (2012).

We now estimate the importance of O$_3$S on surface O$_3$. We calculate the percentage of variance ($r^2$) of surface O$_3$ explained by surface O$_3$S when surface O$_3$ exceeds the 95th percentile (p95, hereinafter) value at each grid point during 1995–2014 summer months using daily average O$_3$ and O$_3$S (see figure 1(c) and (d)). The linear trend has been removed before the calculation. As suggested in figure 1(e), high peaks are located over Wyoming, southern Texas, and the Four Corners area (Colorado, Utah, Arizona, and New Mexico). We see in Arizona, for example, O$_3$S can explain as high as 54% of surface O$_3$ variance during days in which surface O$_3$ exceeds p95. We compare our results with those from prior observational studies. Lefohn et al. (2012) studied stratospheric intrusion events associated with daily maximum hourly O$_3$ in exceedance of 50 nmol mol$^{-1}$ at both rural and urban monitoring sites of the U.S. Statistically significant relations are found over the southern Texas, Arizona, Colorado, Utah, Nevada, California, and Wyoming in summer months during 2006–2008. Overall, our model simulations support observational findings that stratospheric intrusions coincident with O$_3$S preferentially take place in the West and Intermountain West during summertime. We also quantify the stratospheric impact when surface O$_3$ exceeds p95 at each grid cell and find that in regional average over the western U.S. (30°N–50°N, 100°W–125°W), the stratospheric contribution is 18.4% (not shown).
3.2. MCA results and interpretation

We employ MCA to investigate how surface $O_3$ is related to $O_3S$ reaching the surface on daily basis. The first two leading modes are summarized in figure 2. Together, these two modes explain 60% of the covariance pattern over the domain of interest. The first mode shows large positive covariances over the eastern Pacific, western and northern tier of the U.S. (figure 2(a)). The second mode shows dipole structure with $O_3$ and $O_3S$ surplus over the western U.S. and deficit over the east (figure 2(b)). Surface level $O_3S$ anomalies are found to lag the 200–500 hPa $O_3S$ anomalies by two days (not shown), suggesting the downward influence. No significant lead-lag relationships are found either between the expansion coefficient time series (loadings) of $O_3$ and $O_3S$ at the surface or between the first two SVD modes.

We identify ~20 days per month (1404 days in twenty years for the first two leading modes) when both expansion coefficient time series are greater than 0, indicating that surface $O_3$ concentration increase is coincident with $O_3S$ reaching the surface. Lefohn et al (2011), Lefohn et al (2012) investigated the frequency of surface $O_3$ enhancements that are associated with stratosphere-to-troposphere transport down to the surface across the U.S. and reported that the average number of days per month ranges from 16 days to 23 days at monitoring sites in the West and Intermountain West in summer months.

Our results of intrusion frequency are in remarkable agreement with their results.

Next, we define high surface $O_3$ events caused by stratospheric intrusions when both loadings exceed 1.5 standard deviations, as marked by black circles in figure 2. Each event lasts for a few days. Based on this definition, composite analyses according to high surface $O_3$ events are carried out using $O_3S$ daily data. The composited patterns are not sensitive to the thresholds we chose for the analyses. Schematics in figure 3 show the large amplitude composited $O_3S$ structures and near-surface circulations corresponding to both modes. We use neon yellow color to highlight the area with $O_3S$ larger than 120 nmol mol$^{-1}$, which can be treated approximately as the tropopause (Yang et al 2016). During Mode 1, depressed tropopause height followed by enhanced $O_3S$ can be found around 50° N, 120° W (figure 3(a)). Since stratospheric air contains higher values of potential vorticity (PV) and $O_3$, the intrusion of the tropopause tends to replace the tropospheric air by ozone-rich stratospheric air with large PV (Danielsen 1968, Mote et al 1991, Wimmers et al 2003). Transport of $O_3S$ to low levels is tied to stratospheric intrusions and strong subsidence in the troposphere. Figure 4(d) shows a map of composited means of 500 hPa vertical velocity ($\omega$) anomalies on the day of the events corresponding to Mode 1. The intrusion is associated with intensified subsidence on the U.S.-Canada border.
border. Coherently, enhanced O_3S on the northern tier can be seen throughout the troposphere, while the maximum over the Pacific appears below 700 hPa (figure 3(a)). We conduct composited analyses on anomalies of 860 hPa geopotential height as well as surface winds according to events of Mode 1. A quadrupole pattern of low-level geopotential height anomalies is seen over the North American continent. In contrast to the positive correlation between the upper-level cyclonic vorticity and O_3S (figure S3a), low-level geopotential height and O_3S reaching the surface are strongly anti-correlated (figure 3(b)). Dry air (figure S3b) with high PV value (figure S3a) descends on the northern tier of the U.S. as a result of stratosphere-to-troposphere transport by intensified subsidence. Warm and moist tropospheric air is seen (figure S3b–c) downstream (east) of the surface low. In addition to the anomalous descent, we also see a strengthening of northeasterly along the west coast near the surface (figure 3(b)). This intensified easterly associated with stronger anticyclone facilitates the horizontal transport towards the subtropical Pacific in the mid-to-lower troposphere (figure 3(a)).

During Mode 2 (figure 3(c)), the intrusion occurs around 40° N, 115° W. Surface O_3S maximum is concentrated over the Intermountain West, such as Colorado, Arizona, Utah, Nevada. Different from Mode 1, a dipole pattern of the low-level geopotential height anomalies is seen over the continental U.S. Large values of negative geopotential height anomalies are observed above the intermountain regions (figure 3(d)) aligned with cold dry air and positive PV anomalies (figure S4a–c) subsiding west of the surface low pressure while rising warm and moist air occur on the east (figure 4(h) and figure S4b–c).

During both modes, the O_3S reaching the surface can be 10–18 nmol mol⁻¹ (see surface contours of figure 3(a) and 3(c), respectively). In regional average over the western U.S. (30°–50° N, 100° W–125° W, the stratospheric contribution is 11 nmol/mol. The amplitude agrees well with the observational record from the California Baseline Ozone Transport Study (CABOTS), in which they found that stratospheric contribution is 10–20 nmol mol⁻¹ to the surface over Northern California during an intrusion case in August 2016 (Clark and Chiao 2019). We also see ~10 days per summer season when high surface O_3 events associated with the first two SVD modes occur, with variation across years.

3.3. Dynamical mechanism

Next we examine the dynamical mechanism underlying the stratospheric intrusion events associated with
Mode 1 and Mode 2. Figure 4 shows the time evolution of 500 hPa vertical velocity ($\omega$) anomalies and 200 hPa zonal winds for the composites based on high O$_3$ and O$_3$S events in Mode 1 (figures 4(a)–(d)) and Mode 2 (figure 4(e)–(h), respectively. The positions of the upper-level jet axis are marked by dashed lines. The downstream development (Chang 1993), characterized by eastward propagation of the wave systems, can be seen in both modes.

More specifically, for Mode 1, the large descending anomaly is centered at 50° N, 160° W on day −6 (figure 4(a). After 2 days, it shifts eastward following the continuous jet (figure 4(b)). On day −2, the descending center manifests near the west coast (figure 4(c)), consistent with the result that relationship of 500 hPa O$_3$S with the principal component of Mode 1 is maximized about two days ahead of surface O$_3$S peaks (not shown). During day −2 to day 0, the subsidence stays close to the U.S.–Canada border, embedded within the jet stream (figure 4(d)). On day 0, the strong descent on the northern tier of the U.S. continuously facilitates the downward transport of O$_3$S toward the surface (figure 3(a)).

Regarding Mode 2, large disturbance center can be seen on day −6 near the eastern Pacific (figure 4(e)). From day −4 to day −2, the descending center continues to grow near 135° W (figure 4(f)–(g)). The downstream side strengthens remarkably near 125° W where the jet breaks. On day −2, strong anomalous subsidence reaches the western U.S. whereas ascending anomalies are triggered on the east. It is the enhanced subsidence that induces the stratospheric intrusion and downward transport of stratospheric O$_3$ (figures 4(h) and 3(c)).

During boreal summer, the dominant upper-level circulation consists of westerly jet north of 40° N, together with frequent eastward traveling baroclinic waves (White 1982). The baroclinic waves commonly exhibit life cycles of baroclinic growth and barotropic decay along the storm track regions (Simmons and Hoskins 1978, Blackmon et al 1984). Observational and modeling studies have revealed that the baroclinic disturbances leave imprint on atmospheric composition. Aircraft experiments provided evidence that high amounts of stratospheric radioactive debris and ozone were drawn into the troposphere by midlatitude storms (Danielsen 1968). Schoeberl and Krueger (1983) and Mote et al (1991) identified the coherent fluctuations in total O$_3$ and medium scale waves along the wintertime oceanic storm track regions using satellite data. Stone et al (1996) found baroclinic wave features using upper tropospheric water vapor measurement. General circulation model was capable of representing stratosphere–troposphere exchange associated with baroclinic waves in midlatitudes (Mote et al 1994). In our study, baroclinic wave system and its resulting circulation is also found important for the occurrence of deep stratospheric intrusions over the
Figure 5. Days versus longitude diagrams of $\omega$ anomalies (Pa/s) at 500 hPa (a) along the band of 200 hPa jet axis for Mode 1 and (b) along 38° N for Mode 2, respectively.

Our study has examined high surface $O_3$ events associated with downward transport from the stratosphere over the U.S. during summertime, when high temperature could further increase the impact of $O_3$ pollution on mortality. By analyzing a twenty-year (1995-2014) simulation by CESM2 (WACCM6), we have found that the stratospheric $O_3$ can explain as high as 54% of surface $O_3$ variability when surface $O_3$ exceeds 95th percentile, and the regional averaged
stratospheric contribution is ~18% over the western U.S. We have further analyzed the circumstances when stratospheric intrusions of ozone covary with surface O$_3$ anomalies over the region of 20°–50° N and 70°–140° W on daily basis using MCA. The first two leading modes explain 60% of the total covariance pattern. In Mode 1, deep stratospheric intrusions occur preferentially over the northwestern U.S. The associated intensified northeasterly wind anomalies over the west coast further brings continental O$_3$S towards the Pacific Ocean. In Mode 2, deep stratospheric intrusions occur over the Intermountain West. The composited O$_3$S values reaching the surface associated with these two SVD modes range from 10 to 18 nmol/mol. In regional average over the western U.S., the stratospheric contribution is 11 nmol/mol. Both modes are results of eastward propagating wave trains, originating from the central North Pacific and amplifying near the jet exit, with enhanced subsidence over the western U.S.

We have also repeated our MCA analysis to demonstrate the robustness of the dynamical mechanism across seasons. The first leading mode and corresponding anomalous $\omega$ evolution prior to intrusion events for winter, spring, and fall seasons are summarized in figures S5–7. The first leading modes during each of the seasons explain ~40% of covariance between O$_3$ and O$_3$S. Similarly, we find that the strongest disturbance extends from the central North Pacific to the west coast of the U.S. during day $-6$ to day $-4$. During day $-2$ to day 0, the descent remarkably over the west coast while new ascending centers develop on the downstream side. The downward O$_3$ transport is typically regulated by upper-level jet streams across seasons, consistent with previous studies (Langford 1999, Lin et al 2015, Albers et al 2018).

Overall, our study has demonstrated that summertime stratospheric intrusions, though infrequent, can contribute crucially to surface O$_3$ extremes over the Western U.S. These stratospheric intrusion events are caused by strong subsidence in the region, which is a result of eastward propagating baroclinic waves originating from the central North Pacific Ocean. However, a few caveats have to be noted. WACCM6 overestimates tropospheric O$_3$ and thus the exact contribution of stratospheric intrusion to surface should be treated with caution. Additionally, our diagnostic is based on one single climate model because of the use of daily O$_3$S output. It’s worth performing similar analyses in other chemistry climate models to assess the robustness of the conclusions. Future work will also be devoted to 1. studying simulations with high spatiotemporal resolution at certain hot spot areas so that our results can directly benefit air pollution management, and 2. understanding the origination of upstream baroclinic disturbance so that we can improve the predictability of such O$_3$ extremes associated with summertime stratospheric intrusions.

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**Data Availability Statement**

The data that support the findings of this study are available upon request from the authors. All WACCM6 simulations were carried out on the Cheyenne high-performance computing platform (https://www2.cisl.ucar.edu/user-support/acknowledging-nccriscil), and are available at NCAR’s Campaign Storage upon acceptance.

**ORCID iD**

Xinyue Wang @ https://orcid.org/0000-0003-1848-1933

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