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The Evolutions and Large-Scale Mechanisms of Summer Stratospheric Ozone Intrusion Across Global Hotspots

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Key Points:

- There are four hotspots of summer ozone extremes due to stratospheric ozone, North America, Africa, the Mediterranean, and the Middle East
- Summer stratospheric intrusions initiate in the jet axis region near tropopause by isentropic mixing
- Climatological descent drives vertical transport in the lower troposphere and determines the location of the hotspots

Supporting Information:

Supporting Information may be found in the online version of this article.

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Abstract Stratospheric ozone intrusions can have a significant impact on regional near-surface ozone levels. Especially in summer, intrusions can contribute to extreme ozone events because of preexisting high ozone levels near the surface and cause serious health issues. Considering the increasing trend of surface ozone level, an understanding of stratospheric ozone intrusion is necessary. From a 19-year Whole Atmosphere Community Climate Model, version 6 simulation and a stratospheric origin ozone tracer, we identify the global hotspots of stratospheric intrusions based on extreme tracer concentrations near the surface: North America, Africa, the Mediterranean, and the Middle East. We investigate the common underlying large-scale mechanisms of the stratospheric intrusions over the identified hotspots from the lower stratosphere to the lower troposphere. From the trajectory analysis, we find that the upper-level jet drives isentropic mixing near the jet axis and initiates stratospheric ozone intrusion. Subsequently, climatological descent at the lower troposphere brings the ozone down to the surface, which explains the spatial preference of summertime stratospheric intrusion events.

Plain Language Summary High ozone concentration near the surface is harmful to human health. Occasionally, a significant amount of ozone in the stratosphere intrudes deep into the troposphere and increases the surface ozone levels. During summer, as background ozone concentration is high, it is easy for the ozone level to surpass the health threshold with additional contribution from stratospheric ozone intrusion. In this study, we advanced our understanding of the summer stratospheric intrusions, where they happen frequently, and what drives them. We identified four global hotspots of stratospheric ozone intrusion: North America, Africa, the Mediterranean, and the Middle East, which cover areas not well known to be significantly affected in previous studies. We found that upper tropospheric jet dynamics and lower tropospheric descents both play a role in the stratospheric ozone intrusions and determine the locations affected. Based on the mechanisms, we expect to improve our ability to predict when and where summer stratospheric intrusions may occur. Thereby, our findings can also contribute to the establishment of an early warning system for extreme ozone events in summer.

1. Introduction

Ozone is one of the most important chemicals in the atmosphere. The ozone layer in the stratosphere absorbs most of the harmful UV radiation and protects the biosphere at the surface. The energy absorbed by ozone is crucial to the thermal balance in the stratosphere and thereby modifies the stratospheric circulation (e.g., Schoeberl & Hartmann, 1991). On the other hand, ozone in the troposphere is detrimental to the biosphere, particularly to plants (e.g., Heck et al., 1982; Pye, 1988; Reich, 1987; G. Smith et al., 2003). Exposure to high ozone concentrations is harmful to humans also. The World Health Organization (WHO) recommends limiting outdoor activity when 8-hr mean daily maximum ozone levels exceed 50 ppbv (WHO, 2021). Tropospheric ozone is primarily generated by reactions between ozone precursors such as nitrogen oxides (NOx) and Volatile Organic Compounds (VOCs), which originate from both anthropogenic and natural sources (e.g., Finlayson-Pitts & Pitts, 1993). In addition, occasional intrusion of stratospheric ozone into the troposphere can be a major natural source of ozone in certain locations (e.g., Appenzeller & Davies, 1992; Galani et al., 2003; Langford et al., 2015; Lefohn et al., 2011; Lin et al., 2012; Ott et al., 2016; Stohl et al., 2000; Trickl et al., 2014; Wakamatsu et al., 1989; Zanis et al., 2003).

The stratospheric intrusion can happen within multiple phenomena on different temporal and spatial scales, including Rossby wave breaking (RWB) (Holton et al., 1995), tropopause folding (Shapiro, 1980), cut-off low (Price & Vaughan, 1993), and mesoscale convective system (Poulida et al., 1996). The first three types of events can displace the tropopause on the isentropic surface latitudinally and allow a massive amount of stratosphere-troposphere exchange. Then sequentially, the large-scale disturbances and smaller-scale turbulences irreversibly mix the stratospheric air with the troposphere (Holton et al., 1995; Johnson & Viezee, 1981; Mahlman, 1997). Isentropic mixing tends to occur where effective diffusivity (Nakamura, 1996) is high, which coincides with frequent wave breaking (Haynes & Shuckburgh, 2000). Given that wave breaking is frequent on the shear of the jet, it follows that isentropic mixing can also be manifest near the jet. The stratospheric intrusion within the mesoscale convective system is suggested to be initiated from subsidence around the anvil cloud due to mass conservation compensating upwelling tropospheric air. Then, the strong vertical shear can induce differential advection that makes subsidized stratospheric air wrap the anvil cloud (Pan et al., 2014; Phoenix et al., 2020). Given the fast increment of ozone concentration during the intrusion, it is important to understand the nature of stratospheric ozone intrusion to establish an effective policy and ozone warning system for the local community.

Here, we focus on the stratospheric ozone intrusions that intrude deep into the troposphere, which could transport large amounts of ozone to near-surface levels and have substantial potential impact on surface air quality. Škerlak et al. (2015) showed that shallow and medium tropopause folds tend to occur near strong climatological wind in the subtropics, while deep folds follow midlatitude storm tracks. A similar difference is also seen in the stratosphere-to-troposphere transport (STT) patterns between normal STTs and deep STTs (Škerlak et al., 2014). Also, Lin et al. (2015) showed more frequent springtime deep intrusions following La Niña winters, although upper tropospheric ozone peaks after El Niño, underscoring different responses by intrusion depth to climate variabilities. Despite their unique and direct impact on the surface compared to general stratospheric intrusion, deep stratospheric intrusion processes have not yet been studied extensively (Stohl et al., 2003). We will consider deep intrusions that reach 850 hPa due to the relevance to the surface air quality, which is stricter than the 700 hPa criterion used in Sprenger and Wernli (2003).

Moreover, many studies focused on winter or spring intrusions when the intrusion is more frequent and stratospheric ozone is abundant (Breeden et al., 2021; Johnson & Viezee, 1981; Langford et al., 2009; Lin et al., 2012, 2015; Zhao et al., 2021). However, the consequence of summertime intrusions on surface ozone could be more impactful. During summer, photochemical ozone production reaches its peak, and events like thunderstorms, heat waves, and wildfires are more frequent, which could produce ozone precursors (NOx and VOCs) and increase the reaction rate due to high temperature (Gaudel et al., 2018; Jaffe & Wigder, 2012; Lu et al., 2016; Murray, 2016; Solberg et al., 2008). Because the background ozone concentrations are high, surpassing the health threshold with an additional stratospheric contribution can be easy. The model results also show that summer stratospheric origin ozone is not negligible and can potentially trigger extreme ozone events near the surface (Wang et al., 2020). In addition, summer stratospheric intrusions likely favor certain geographical locations (Akritidis et al., 2021; Škerlak et al., 2015). Previous studies have shown that the west coast of the United States (Danielsen, 1980; Lefohn et al., 2011, 2012; Wang et al., 2020) and the eastern Mediterranean (Akritidis et al., 2016; Zanis et al., 2014) are influenced by stratospheric intrusion in summer. However, we lack the general statistics of intrusion frequency that covers the entire globe and commonality in the mechanisms that can unify the events in different locations. As we expect increased STT of ozone and its contribution to tropospheric ozone in the future (Akritidis et al., 2019; Elsbury et al., 2023; Meul et al., 2018), an overall understanding of summer stratospheric intrusion is needed.

The difficulty of intrusion studies is distinguishing the stratospheric contribution in the air, especially once it is mixed with the surroundings. There are some observations from field works and ground observatories, but their spatial coverage or time period is insufficient for a general understanding (Galani et al., 2003; Gerasopoulos et al., 2006; Gronoff et al., 2021; Ott et al., 2016; Trickl et al., 2016, 2020; Wakamatsu et al., 1989; Xiong et al., 2022). To overcome these hardships and cover diverse mechanisms, there are multiple approaches like tropopause folding identification algorithms and back trajectory models on reanalysis and model data (e.g., Li et al., 2015; Škerlak et al., 2015). Here, we will use an artificial tracer called stratospheric origin ozone (O_3S). O_3S is identical to the ozone in the stratosphere. However, once O_3S passes the tropopause and enters the troposphere, O_3S does not have production routes and is removed at the same rate as ozone. Therefore, tracking the stratospheric contribution throughout time and space is very useful through O_3S (Akritidis et al., 2016, 2022; Albers et al., 2022; Bartusek et al., 2023; Elsbury et al., 2023; Lin et al., 2012; Zanis et al., 2014). It also allows us to cover intrusion events regardless of their triggering mechanisms.

In this study, we aim to address three questions about summer stratospheric ozone intrusion using a state-of-the-art chemistry-climate model and a stratospheric origin tracer: (a) Where and how often do we see extreme summer stratospheric ozone intrusion events? (b) What is the pathway for the intrusion? (c) What is the mechanism in common that drives these events across the global hotspots?

2. Methods

2.1. WACCM6 and O_3S

Our study is based on the daily summertime (June-August, JJA) ozone (O_3) and O_3S from the Whole Atmosphere Community Climate Model, version 6 (WACCM6) experiment. WACCM6 is a high-top chemistry-climate model of the Community Earth System Model version 2. The model has a horizontal resolution of $0.95° \times 1.25°$ (latitude × longitude) and 70 hybrid sigma levels in the vertical (~1.1 km resolution near UTLS). It has high reproducibility of sudden stratospheric warmings and variability of physical variables, such as temperature, wind, and chemicals, in the middle atmosphere, leading to a better stratosphere-troposphere coupling than low-top models (Gettelman et al., 2019). WACCM6 shares most of the physical parameterizations as the low-top Community Atmosphere Model version 6 with an additional gravity wave scheme. The model chemistry mechanism covers the troposphere up to the lower thermosphere. WACCM6 has interactive Community Land Model version 5 coupled as default, and our simulation is a fully coupled ocean-atmosphere historical run. In addition to its better performance in stratospheric dynamics, O_3 in both the stratosphere and troposphere has higher fidelity than previous versions (Emmons et al., 2020; Gettelman et al., 2019). Therefore, WACCM6 is suitable for our study on summer stratospheric ozone intrusion (Wang et al., 2020). More information about the model schemes and performance is available in Gettelman et al. (2019) and Emmons et al. (2020). We analyzed the data from 1996 to 2014, during which O_3S is equilibrated. The O_3S tracer is implemented in the model, as Tilmes et al. (2016) demonstrated. This idealized tracer is identical to O_3 above the tropopause and is removed via the same removal process as O_3 in the troposphere. However, unlike O_3, it does not have any production once in the troposphere. The model uses the lapse-rate tropopause as the default, which is defined as the lowest level where the temperature lapse-rate decreases to 2 K/km or less (Reichler et al., 2003; WMO, 1957).

2.2. Maximum Covariance Analysis (MCA)

We applied Maximum Covariance Analysis (MCA) on the daily 850 hPa O_3 and O_3S anomalies during JJA 1996–2014 for each hotspot, which will be defined later. The MCA is a statistical method to identify and analyze relationships between two data sets. It uses Singular Value Decomposition to extract spatial patterns and Principal Component (PC) timeseries of two data sets that maximize the covariance (Bretherton et al., 1992). It can help identify mechanisms that explain the covarying patterns of the two variables and is suitable for our study to analyze the covariability between near-surface O_3 and O_3S. The 850 hPa level is selected to identify the near-surface extreme O_3 intrusion events. Although the high-altitude regions, such as the Tibetan Plateau, potentially have a larger influence from the stratosphere because of their proximity to the stratosphere (Škerlak et al., 2019), we aim to understand the dynamics and impact over the regions in which the distance from the tropopause is similar. We defined daily anomalies after removing the linear trend and seasonal cycle for O_3 and O_3S. We used the Fourier transform on the 19-year average of each day of the year and extracted up to the fourth harmonics to form a seasonal cycle. This way, we could remove some noise from a relatively small number of years.

2.3. TRAJ3D Model

The TRAJ3D model, a three-dimensional Lagrangian trajectory model, operates solely on wind vectors to determine the tracer's location (Bowman, 1993; Bowman & Carrie, 2002). The input daily wind data is obtained from our WACCM6 experiment. The trajectory calculations are performed every hour, and trajectory locations are output daily. The four-dimensional linear interpolation is conducted on the wind vector (Bowman et al., 2013). TRAJ3D is not suitable to analyze trajectory in the lower troposphere since it does not account for convection or turbulent motions. Therefore, tracers are released at 500 hPa, and the backward trajectory is integrated for a period of five days. Given that 5 days is considerably shorter than the typical lifetime of ozone in the free troposphere (a few weeks), we treat O_3S as a passive tracer with an infinite lifetime. The stratospheric intrusion is known to be dominated by isentropic mixing near the tropopause (Holton et al., 1995), which occurs at a finer spatial and

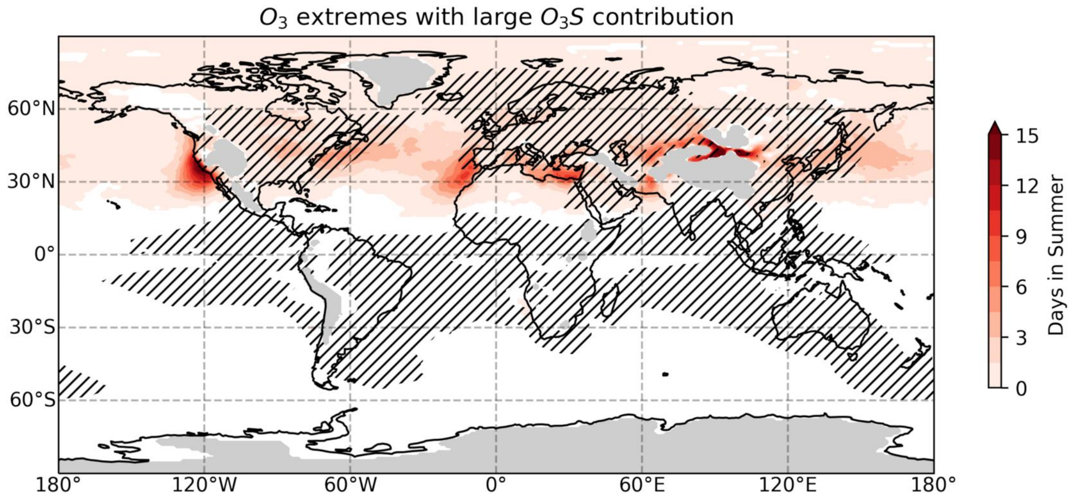

Figure 1. The average number of summer days during which the 850 hPa O_3 exceeds 56.7 ppbv, with 850 hPa O_3S/O_3 ratio exceeding 30%. Red shadings are days per summer, and gray shadings are masked topography. Regions where R-squared values between anomalous O_3 and O_3S below 0.5 are hatched. The summer period is defined as JJA for NH (19 years) and DJF for SH (18 years starting from December 1996).

temporal resolution compared to the given daily vertical velocity. As the pure trajectory model does not incorporate parameterization for convection, mixing, and turbulence, vertical displacement should be considered with potential uncertainties (W. P. Smith et al., 2021).

3. Results

3.1. Global Hotspots of Stratospheric Intrusion Events

Hotspots in our study are defined as where extreme O_3 events are frequent, and the stratospheric contribution to these events is significant. Figure 1 shows the number of days per summer (JJA for NH, DJF for SH) when the 850 hPa O_3 exceeds 56.7 ppbv and the contribution of O_3S to O_3 is greater than 30%. The O_3 threshold is based on the 850 hPa summer median ozone level (56.7 ppbv) of days when the 900–1,000 hPa average O_3 exceeds the health threshold (50 ppbv; WHO). In summer, the boundary layer is elevated, and the mixing processes are vigorous between 900 and 1,000 hPa. In addition to the above two criteria, we further narrowed it to the regions where O_3S accounts for over half of the O_3 variability within the hotspots. Specifically, we considered regions where the correlation between anomalous O_3S and O_3 is significant, with an R-squared larger than 0.5.

Overall, most stratospheric intrusion events occur in the NH midlatitudes, between 20° and 50°N (Figure 1). Conversely, the SH exhibits fewer events mainly due to the low O_3 concentration near the surface. The result reveals four global hotspots for stratospheric intrusions: the West coast of North America (NA), the Northwest coast of Africa (Af), the Eastern Mediterranean (MD), and the Middle East near Iran and Pakistan (ME). Other regions, such as the northern Tibetan Plateau and eastern NA and Asia, are not the focus of this study due to low correlations between anomalous O_3 and O_3S. Interestingly, except for the ME hotspot, the remaining hotspots are the Mediterranean climate regime (Kottek et al., 2006). These hotspots qualitatively align with the NH hotspots for tropopause folding events documented in previous studies (Škerlak et al., 2015; Sprenger et al., 2003). The small discrepancies could arise from the differing altitudes of focus (near-surface vs. near-tropopause). Among the hotspots, the NA region exhibits the highest frequency, with approximately 15 events per summer, and other hotspots also experience a minimum of 6 events per summer. A sensitivity test on the ratio threshold consistently highlights these four hotspots as significant unless an exceptionally large threshold is applied (not shown). However, such high thresholds are deemed inappropriate for our discussion as they result in significantly reduced event frequencies.

We also analyzed the frequencies of events exceeding 99% of all the 850 hPa O_3S across the NH for each season to see where the relative intrusion hotspots are for each season (Figure S1 in Supporting Information S1). Although summer has the lowest 99% O_3S concentration (19.14 ppbv), it clearly shows the four global hotspots we have

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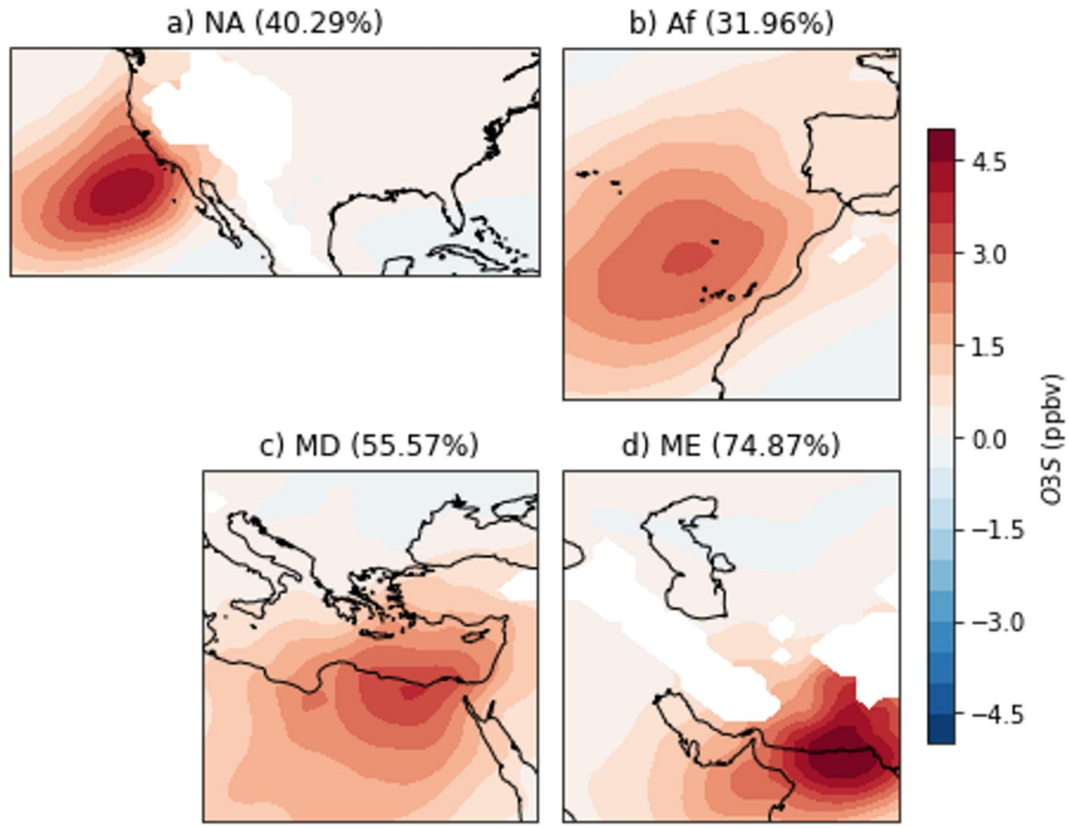

Figure 2. The 850 hPa O_3S spatial patterns from Maximum Covariance Analysis (MCA) leading mode are shown for global hotspots: North America, Af, MD, and ME. The MCA has been conducted on the daily 850 hPa O_3S and O_3 anomalies in JJA 1996–2014. The covariance fraction explained by the leading mode is written in the parenthesis next to each hotspot.

seen in Figure 1. Interestingly, only summer shows such four global hotspots with a strong zonal asymmetry, while other seasons are more zonally symmetric. The NA hotspot always exists throughout the year, but other hotspots disappear in other seasons. Meanwhile, weaker hotspots are seen in different locations in other seasons, for example, over the northern Atlantic and the east coast of NA. This result emphasizes the unique features of summer stratospheric ozone intrusion and its impact on surface ozone extreme events. Also, similar patterns in summer between Figure 1 and Figure S1c in Supporting Information S1 indicate that stratospheric intrusions are indeed important in extreme ozone events near the surface.

3.2. MCA Results

We applied the MCA on each identified hotspot between 850 hPa O_3S and O_3 to determine the major mechanism for the covariability of the two variables (Figure S2 in Supporting Information S1). In this paper, we will study the leading mode of each hotspot since it represents a dominant pattern in the region (Figure 2). The PC timeseries are divided by the standard deviation, and loading vectors are multiplied by that standard deviation. The leading mode outstands the other modes for all the hotspots, especially at the ME hotspot (74.87%). Even the lowest covariance fraction for the leading mode is significantly high (31.96% at the Af hotspot). Thus, the first mode of MCA can represent the large-scale conditions that simultaneously intensify both O_3S and O_3.

The extreme events hereafter are days when both O_3S and O_3 PC timeseries for the leading mode exceed one standard deviation level, and we count the continuous extreme days as a single event. We identified 61, 65, 48, and 51 total events in NA, Af, MD, and ME during 19 years, respectively, which is about 3% of the total days in summer (Figure S3 in Supporting Information S1). Although most events last 1–2 days, long-lived events can last as long as 10 days (not shown). Figure 2 shows that NA and Af patterns peak in the nearby ocean, while MD and ME peaks are on the coast with higher population densities, which appear to be more detrimental to nearby communities. Especially the peak concentration in the ME hotspot is the highest among hotspots (Figure 2). We conducted additional analysis to examine the surface (lowest model level) ozone concentration 1 day after the extreme events at 850 hPa (Figure S4 in Supporting Information S1). Except for Africa, most identified hotspots exhibited elevated absolute ozone concentrations (O_3 > 50 ppbv; red contours) over land areas and corresponding ozone anomalies within part of the extreme ozone areas. As events are identified based on the MCA, coherent patterns between O_3 and O_3S exist in most hotspots. Therefore, identified summer stratospheric intrusion events can contribute to surface ozone extremes. In addition, to verify the dominance of the MCA-identified events among all extreme events, we compared the MCA-identified events with events identified in Figure 1. For each grid over the peaks of hotspot regions, about 30%–50% of the MCA-identified events overlap, and the O_3S averages over event days are similar in magnitude (not shown). Therefore, understanding the mechanism of MCA-identified extreme events could help explain many extreme events in the global hotspots.

Extreme events occur irregularly, and the frequency changes over time. There is no clear increasing or decreasing trend in the number of extreme events in any hotspot during this 19-year (1996–2014) period (not shown). We also analyzed if there is any preference in the timing of events within the summer for each hotspot (Figure S3 in Supporting Information S1). Most locations have a weak intraseasonal variability, except MD, which has a strong intraseasonal variability. The MD hotspot shows a large preference in the early summer and almost no events in August. This is an unexpected result since previous studies emphasized that Etesian wind in the Mediterranean strongly correlates with the intrusion, which peaks in late summer (Dafka et al., 2021; Tyrlis & Lelieveld, 2013). Although a few questions exist in the intraseasonal variability of extreme events, we will focus on the general features of extreme events to understand the commonalities between events.

3.3. Trajectory and Upper-Level Dynamics

The O_3S anomaly composites were analyzed to examine the typical intrusion patterns utilizing the MCA results. To determine the descending process of the intrusion, the box averaged O_3S anomaly was calculated for every level from 8 days prior to the event up until the event days (Figure 3). Based on the O_3S composites, a box region was selected to encompass the potential trajectories associated with each hotspot (Figure S2 in Supporting Information S1). The analysis reveals a descent starting from approximately 4-5 days before the events at about 500 hPa. Above 500 hPa, it is challenging to distinguish a descent due to the high background O_3S concentrations. Consequently, the use of a back trajectory model is necessary to differentiate the intrusion from the background O_3S above 500 hPa.

The TRAJ3D model was employed to estimate the trajectories of summer stratospheric ozone intrusion above 500 hPa. Because TRAJ3D does not account for convection or turbulent motions that are important for the lower troposphere, we release the tracers at 500 hPa to avoid the high uncertainty in the lower troposphere. First, a box region enclosing the statistically significant maximum of the 500 hPa O_3S anomaly three days before the events is designated for each hotspot. This specific date is chosen as the intrusion signal at 500 hPa O_3S anomaly displays the most prominence. Subsequently, tracers are released at the significant area of each intrusion case within the assigned box region, where O_3S exceeds one standard deviation in time. Back trajectories are then calculated for a period of 5 days with a large set of tracers. Hereby, "ensemble" is each extreme stratospheric intrusion case at each hotspot, and "trajectory" is an individual trajectory among a large set of trajectories for each ensemble, which has different initial locations from each other. Within the significant area for each ensemble, 1,000 trajectories are initiated at randomly selected grid points allowing duplication after regridding to 0.5° × 0.5° resolution. For example, the NA hotspot has 61 ensembles, and each ensemble has 1,000 trajectories, total of 61,000 trajectories (61 ensembles × 1,000 trajectories).

The trajectories for each hotspot are presented in the left column of Figure 4. The mean trajectories for each ensemble are depicted as gray lines. The colored line shows the mean trajectory for the hotspot, which is the mean of gray lines, with height represented in color. For instance, in the NA hotspot, gray lines show 61 ensembles, which is the mean of 1,000 trajectories for each ensemble. The colored line is an average of 61 ensembles, which is the mean of 61,000 trajectories. Generally, for all the hotspots, the trajectories exhibit a southeastward descent that crosses the jet axis, which is denoted by the red contours. Consistent with previous studies, the southeastward descending pathway is attributed to the tilted isentropic surfaces and the strong climatological westerlies in the midlatitudes (Akritidis et al., 2016; Škerlak et al., 2014; Sprenger & Wernli, 2003).

Previous studies have highlighted that during boreal summer, NH STT exhibits two latitude maxima: one over the midlatitudes and the other over the subtropics (Hsu et al., 2005; Jing et al., 2004; Tang et al., 2011). In the

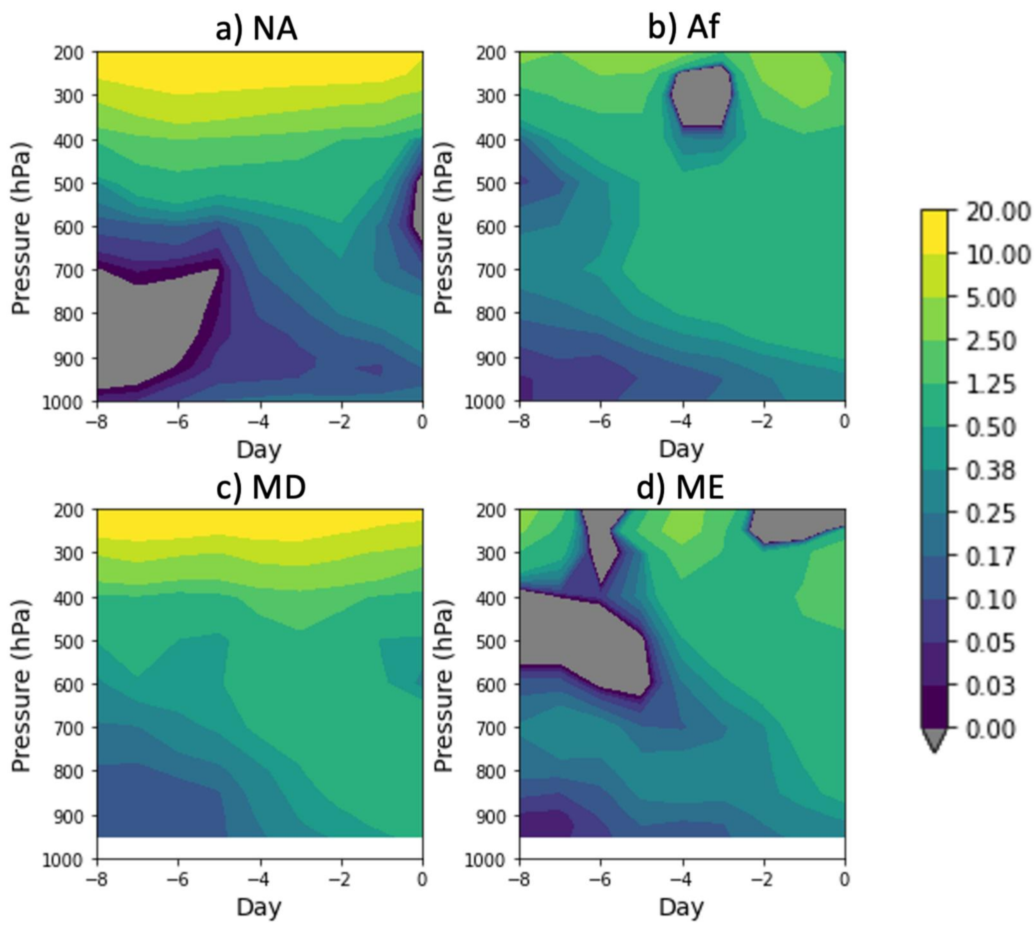

Figure 3. The box averaged O_3S anomaly for each level from 8 days prior to the events to the event days. The box regions are defined in Figure S2 in Supporting Information S1. The negative values are grayed out. The white areas are topography.

midlatitudes, deep convection over continents plays a significant role, whereas, in the subtropics, it is primarily via RWB over the ocean. However, Škerlak et al. (2014) demonstrated that summer deep stratospheric intrusions originate from locations distinct from the mentioned STT maxima. Given the considerable number of ensembles crossing the jet axis, we hypothesize that isentropic mixing near the jet axis is the source of O_3S for deep STT in summer (Holton et al., 1995). We first quantified the possibility of the intruding air parcels passing the jet axis. In other words, whether the estimated trajectories intersect the jet axis. Figure 5 presents an example of a single case with the zonal wind at 200 hPa 8 days before the event. The tracers are denoted by dots, with tracers in the jet axis region depicted in blue. The jet axis region is defined as where the zonal wind exceeds 20 m/s (red shading). The jet cores are identified as wind maxima latitudes for each longitude. We calculated the number of trajectories that crossed the jet axis region regardless of the level and time. In the case study of Figure 5, 38.1% of the trajectories are found to cross the jet axis region at the given level and time. The middle column of Figure 4 summarizes the statistics of all the trajectories and illustrates the trajectory threshold and the corresponding percentage of ensembles that meet the threshold. For instance, a 50% trajectory threshold means that over 50% of the trajectories cross the jet axis. The *y*-axis represents the percentage of ensembles that pass the test at a given trajectory threshold, normalized by the total number of ensembles within each hotspot. The findings reveal a substantially high percentage of ensembles that pass the test for all hotspots. This indicates that intrusion trajectories have a high possibility of crossing the jet axis region and supports the idea that isentropic mixing near the jet axis is the source of O_3S for deep STT in summer.

We further assessed whether the number of trajectories crossing the jet axis region is statistically significant to support the hypothesis that intruding air parcels passed the jet axis. For each ensemble, at multiple pressure levels and dates preceding the event (300 hPa, 250 hPa, and 200 hPa, 4–8 days prior), we computed the area ratio (r) of

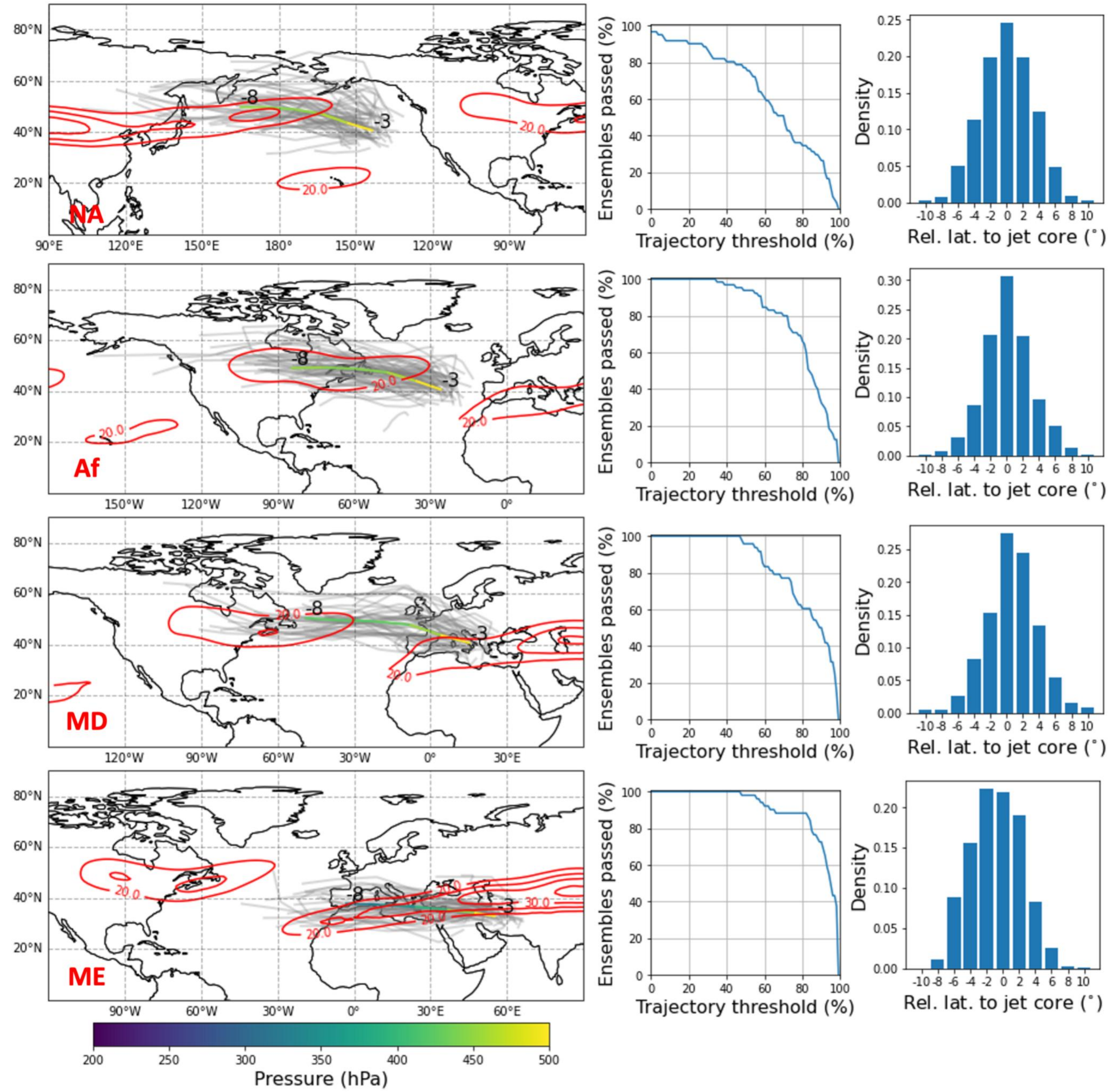

Figure 4. (left) The back trajectories from the TRAJ3D model within each hotspot. The gray lines indicate ensemble mean trajectories. The colored line shows the mean trajectory across all the ensembles, which is the mean of gray lines, with height represented in color. The red contours are 200 hPa zonal wind averaged over 3–8 days before every event. (middle) The percentage of ensembles that pass the jet axis region as a function of trajectory threshold. Any time between 4 and 8 days before the event, if the trajectory passes the jet between 200 and 300 hPa, the trajectory is considered as crossing the jet axis. Details are explained in the text. (right) The histogram of the relative latitude of trajectories to the jet core at 200 hPa. The day with most tracers passing the jet axis from 4 to 8 days prior to the event is considered. Every bin width is 2°, centered on the values at the *x*-axis. Each row exhibits results at each hotspot: North America (first row), Af (second row), MD (third row), and ME (fourth row).

the jet axis region to the box region enclosing the maximum and minimum latitude and longitude of the tracers (Figure 5). Assuming a completely random process, each tracer can be considered to follow a binomial distribution with a sample size (N) of 1,000. Applying the central limit theorem, this distribution can be approximated by a normal distribution with Nr as the mean and $Nr(1-r)$ as the variance. Through standardizing, we found that around 80% of the cases exhibit a significance exceeding two standard deviations for each hotspot (2.5%, one-

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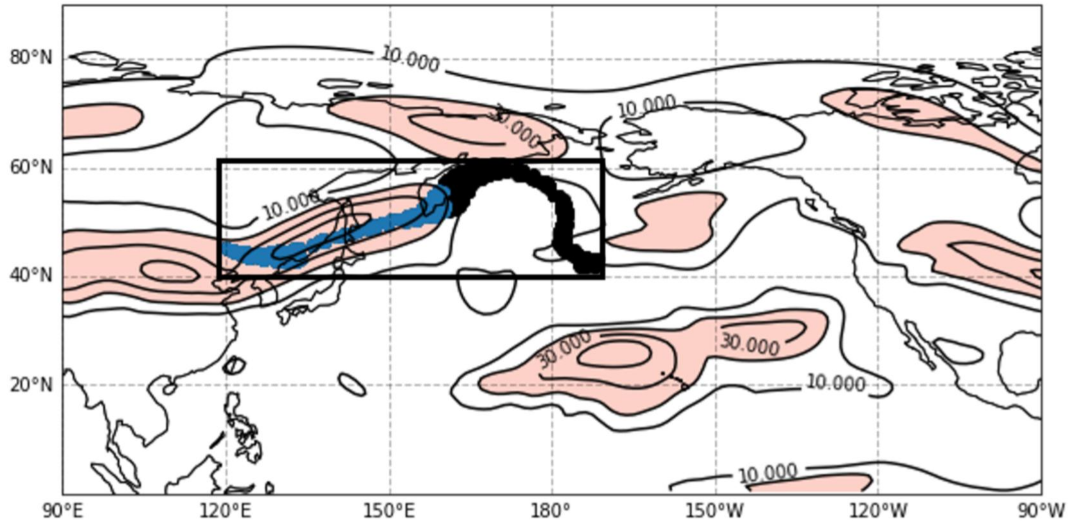

Figure 5. A case study of tracer ensembles 8 days before the event at 200 hPa (30 July 1996). The contours are 200 hPa zonal wind velocity (u). The dots are tracers, and tracers on the jet axis region (red shading; $u > 20$ m/s) are in blue. The box is assigned to encompass all the tracers.

sided). For instance, in the NA hotspot, 79.3% out of 915 cases (61 ensembles × 5 dates × 3 pressure levels) have passed the significance test. The result indicates that, regardless of the time and level, there is a significant likelihood of the trajectory intersecting the jet axis. A sensitivity test was performed by releasing tracers at the maximum within 2 and 4 days before the event, yielding similar substantial probabilities of stratospheric intrusion crossing the jet axis (not shown).

Furthermore, we investigated the preferred crossing location of trajectories relative to the jet core in terms of latitude at 200 hPa. The location on a day with a maximum number of tracers on the jet axis between 4 and 8 days before the event is examined. As shown in the right column of Figure 4, the distribution is centered near the core region, with a slight poleward tilt except for the ME hotspot. A sensitivity test on different pressure levels shows a slight shift to the poleward flank on lower pressure levels. Still, it does not affect the general feature of the relative locations (not shown). This finding is consistent with Yang et al. (2016), which noted that summer intrusions exhibit a unique characteristic wherein the peak ozone flux into the troposphere occurs near the core region, while other seasons prefer the poleward flank. The reason for the ME hotspot's preference for the equatorward flank of the jet remains not understood, but it could be attributed to a distinct dynamical mechanism associated with the Asian summer monsoon (Tyrlis et al., 2014; Wu et al., 2018).

Next, we examine the transport process below 500 hPa using the WACCM O_3S as the intrusion process becomes eminent due to the low background O_3S level. Also, because TRAJ3D does not account for convection or turbulent motions that are important for the lower troposphere, it is not suitable for the lower troposphere. Three hotspots, that is, NA, Af, and MD, continue their southeastward or southward gradual descent to 850 hPa following the large-scale circulation (Figure S5 in Supporting Information S1). However, ME trajectories experience a rapid descent closer to the event days into the lower troposphere. Then, the northerly wind transports O_3S toward the 850 hPa hotspot (not shown). We will briefly discuss how the ME hotspot trajectories and governing mechanisms differ from others in the next section.

3.4. Vertical Transport in the Lower Troposphere

Now that we know the pathway, we further address the question of what contributes to the vertical descent in the lower troposphere, where intrusion departs from isentropic motion and crosses the isentropic surface (not shown). In other words, what is the dynamics that is in common in bringing the O_3S down to the near-surface level? To answer the question, we conducted a budget analysis on the tendency of the O_3S anomaly transport at 850 hPa (600 hPa for ME). The tendency can be decomposed into the following terms:

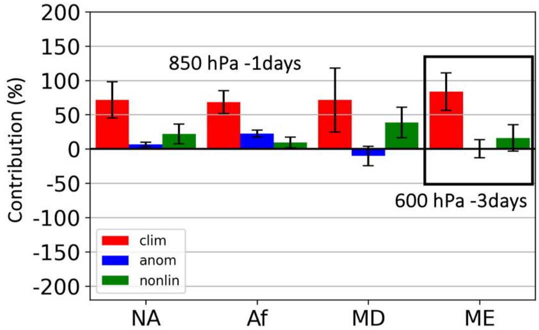

Figure 6. The relative importance of vertical transport decomposition to the total vertical transport. Red, blue, and green indicate climatological wind-driven, anomalous wind-driven, and nonlinear terms, respectively. The total vertical transport is calculated in the tendency equation on the maximum tendency at a given time and pressure level. To compare with other hotspots, each term in vertical transport is divided by the total vertical transport from the corresponding hotspot. Thus, three terms percentages add up to 100% for each hotspot. Three hotspots (North America, Af, and MD) are calculated on day −1 at 850 hPa, while ME is calculated on day −3 at 600 hPa. The terminations of each term and their formula follow the equation in Section 3.4, and a detailed explanation of the time and level selection is given there.

$$\frac{\partial O_3S_a}{\partial t} = -\omega_c \frac{\partial O_3S_a}{\partial p} - \omega_a \frac{\partial O_3S_c}{\partial p} - \left(\omega_a \frac{\partial O_3S_a}{\partial p}\right)_a + (\text{Zonal}) + (\text{Meridional}) + (\text{Residuals}),$$

where subscript a indicates anomaly and c indicates climatological seasonal cycle. We first decomposed the tendency into zonal, meridional, vertical transport, and residual terms. Then, we further separated vertical transport into contributions from climatological wind ($-\omega_c \frac{\partial O_3S_a}{\partial p}$), anomalous wind ($-\omega_a \frac{\partial O_3S_c}{\partial p}$), and nonlinear ($-\left(\omega_a \frac{\partial O_3S_a}{\partial p}\right)_a$) terms, as the focus is on the mechanism that brings air down. We smoothed the data by taking a 5° × 5° moving box mean for each term. Then, we examined the maximum tendency for each pressure level near the hotspots and 10 to 0 days before the events. The analysis reveals a greater magnitude of horizontal transport compared to vertical transport (not shown), which is expected due to larger horizontal wind velocities. However, since our question is on the mechanism of vertical transport to the near-surface level, we focus on the common factors that contribute to the vertical transport across the global hotspots.

The role of climatological wind-driven vertical transport is substantial in all hotspots (red bars in Figure 6). This figure illustrates the vertical transport in the maximum tendency for a day before events at 850 hPa and its decomposition for each hotspot (NA, Af, and MD). Below 500 hPa, we see a gradual descent from the O_3S anomaly composites in three hotspots except ME. The ME hotspot shows a strong descent a few days ahead of the event and then horizontal transport to the event region. As we focus on the driver of vertical transport, we selected the level and time where the major descent happens at the ME hotspot, which is −3 days at 600 hPa (not shown). The climatological vertical transport dominates in the lower troposphere from 600 to 850 hPa near the hotspots, even out of the maximum tendency region. This climatological dominance explains the co-location of climatological descent regions (mainly the Mediterranean climate regime) and global hotspots of stratospheric intrusion. In addition, it's worth noting that the climatological wind-driven vertical transport also depends on the vertical gradient of the O_3S anomaly. Our understanding is that the upper-level system induces this anomaly, as demonstrated in Section 3.3. Once the gradient is established, the climatological descent transports the anomaly to the near-surface level. The upper-level dynamics that initiate the intrusion is attributable to jet dynamics, as our trajectory results and previous studies suggested (Figure 3, Wang et al., 2020). Although climatological descent in the lower troposphere does not elucidate all the intrusion processes and determines the location, it has a considerable contribution. This also explains the distinct geographical locations of O_3S extremes in the summer compared to other seasons. The climatological descent is prominent during the summer due to the anticyclone formation in the ocean (NA and Af, Rodwell & Hoskins, 2001) and the Asian summer monsoon (MD and ME, Wu et al., 2018) (Figure 7).

The ME hotspot is notable as its strong descending region is far apart from the hotspot region. The O_3S hotspot is located near the coast of Pakistan, while strong descent happens about 10°N. This is an example of horizontal transport moving the descended ozone from one location to another and setting the location of O_3S extremes. Once we focus on the descending period (day −3 at 600 hPa), the climatological descent dominates the vertical transport, as mentioned earlier. The northerly wind that transports O_3S to the hotspot shows a similar pattern to the Asian summer monsoon circulation. In addition, anomalous high precipitation is also observed in the Bay of Bengal 2 days before the extreme events (not shown). These results are consistent with the large-scale descent and tropopause folds in the Middle East occurring as a result of monsoon dynamics discussed in previous studies (Rodwell & Hoskins, 2001; Wu et al., 2018).

4. Conclusion and Discussions

We identify summertime stratospheric intrusion hotspots using a state-of-the-art chemistry climate model and a stratospheric origin tracer, and investigate the pathway and mechanism of these intrusion events. MCA (Figure 2)

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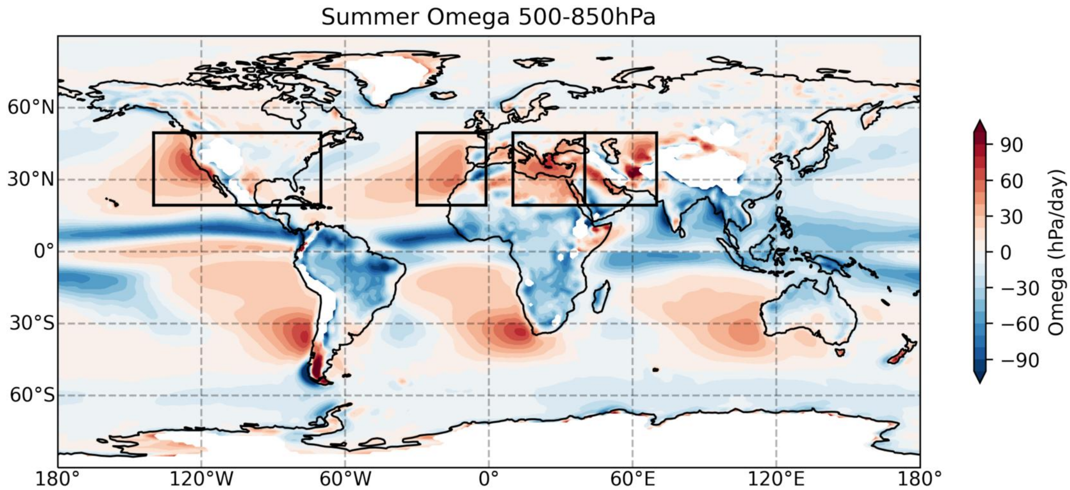

Figure 7. Summer climatological omega averaged from 500 to 850 hPa. Positive omega indicates descent. Each box region corresponds to each hotspot. The summer period is defined as JJA for NH (19 years) and DJF for SH (18 years starting from December 1996).

demonstrates that there are four global hotspots with frequent near-surface summer ozone extreme events due to stratospheric intrusion: the West coast of NA, the Northwest coast of Africa (Af), the Eastern Mediterranean (MD), and the Middle East near Iran and Pakistan (ME). Figure 8 summarizes the dynamical mechanism underlying the intrusion process from the lower stratosphere to the lower troposphere. To elucidate the trajectory and underlying mechanisms of each hotspot, we employ the Lagrangian pure transport model (TRAJ3D). The stratospheric intrusions above 500 hPa generally follow a southeastward descent and traverse the jet axis as they enter the troposphere (Figure 8a). The estimated trajectories align well with previous studies and are potentially driven by isentropic mixing near the tropopause (Škerlak et al., 2014; Yang et al., 2016). Furthermore, budget analysis shows that the climatological descent-driven vertical transport is the governing mechanism for descent from the mid- to the lower-troposphere (below 500 hPa) over all hotspots (Figure 8b). This explains the global hotspots being located in the strong climatological descent regions, mostly in the Mediterranean climate regime (Figure 7).

Our study differs from previous works in that we identified hotspots of stratospheric intrusion with the frequency of extreme ozone events, which is directly relevant to the air quality. Most previous studies focus on a monthly or

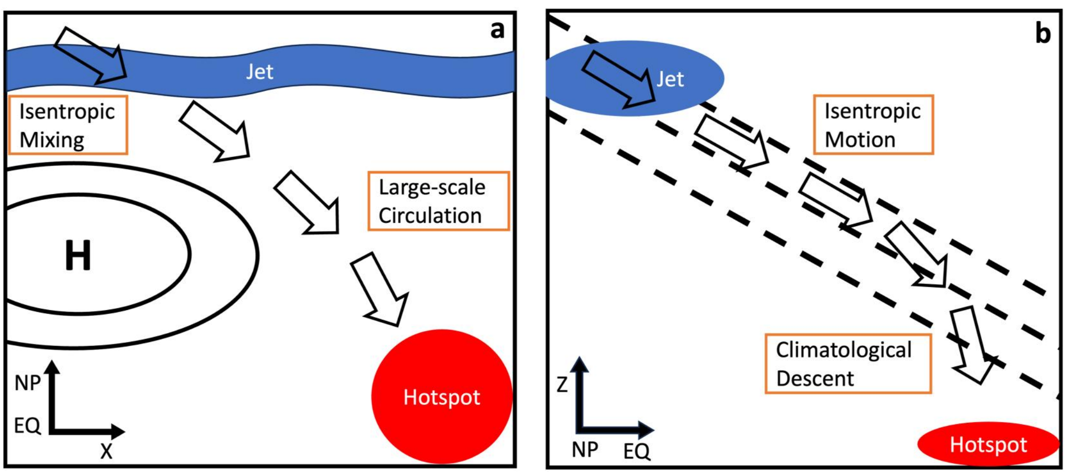

Figure 8. Schematic illustrating horizontal (left) and vertical (right) cross-section of the stratospheric intrusion. Arrows show the movement of O_3S, and red oval areas indicate a stratospheric intrusion hotspot. The blue curve and oval area on top of the panels are jet stream axis regions. Black solid contours and the "H" on the left panel denote an anticyclone that induces large-scale circulation. Dashed lines on the right panel indicate isentropic surfaces.

seasonal average amount of mass or ozone transported from the stratosphere to the troposphere (e.g., Albers et al., 2022; Škerlak et al., 2014; Sprenger & Wernli, 2003). Anomalously high amounts of ozone intrusion on a monthly scale are commonly associated with frequent extreme events. However, the frequency of extremes also depends on the variation of the distribution, resulting in deviation from the average. In addition, our results provided a comprehensive analysis of the continuous transport process from the lower stratosphere all the way down to the lower troposphere, while many previous studies focused on origin and/or destinations (e.g., Škerlak et al., 2014; Sprenger & Wernli, 2003). We also identified the common dynamical drivers governing summer hotspots, including the initiation of intrusion through isentropic mixing near the jet and large-scale climatological descent over the hotspot.

Furthermore, we have shown that deep summer stratospheric intrusion has unique characteristics and affects the regions not much considered earlier, such as the northwest coast of Africa and near Iran and Pakistan (ME hotspot). Especially the Pakistan region shows extremely high frequency of stratospheric intrusion. Our analyses suggest the Asian summer monsoon as a possible precursor. Therefore, studies examining the linkage between the Asian summer monsoon and the summer stratospheric intrusion in Pakistan are needed considering their poor background air quality and high population (Anjum et al., 2021; Mehmood et al., 2020).

There are still several unresolved issues regarding summer stratospheric intrusions. The Rossby waves near the upper tropospheric jet and persistent climatological descent cannot explain the rareness of the intrusion events. We propose that these two pieces must be connected with a suitable horizontal wind, which is a potential third key factor, for extreme events to occur. If the ozone flux in the upper troposphere does not reach the climatological descent region, it will not be able to reach the near-surface level. Another possibility is that either the upper-tropospheric wave activities or the climatological descent is extreme during these events. However, the intra-seasonal variability of the climatological descent is likely too weak to explain the occurrence of extreme events. A mechanistic study on the upper tropospheric dynamics in summer could fill the gap in our understanding of the summer stratospheric intrusion. The intraseasonal and interannual variability of the summer stratospheric intrusion also needs further study. For example, a strong intraseasonal variability exists in the MD hotspot, with no events in August in this model simulation. Also, the interannual variability of the summer intrusion at the NA hotspot does not show a connection to ENSO (not shown), whereas the spring deep STT increases during La Ninas (Albers et al., 2022; Lin et al., 2015).

Although this study is based on a single chemistry climate model output, it provides a comprehensive analysis of the global hotspots of summertime stratospheric intrusions and their underlying dynamical mechanism. It is worthwhile conducting further studies with a different model and data set to test the robustness. Also, our analysis was able to track the stratospheric intrusion signal a week before the event. Although we need further study on the unresolved issues to address precursors precisely, our findings can potentially contribute to forecasting extreme ozone events in summer and benefit policymakers in establishing an early warning system.

Data Availability Statement

The WACCM6 processed data used for the analysis in the study are available at Columbia University Academic Commons (Lee, 2024).

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