The Evolutions and Large-scale Mechanisms of Summer Stratospheric Ozone Intrusion across Global Hotspots

Jae Won Lee¹, Yutian Wu¹, and Xinyue Wang²

¹Lamont-Doherty Earth Observatory of Columbia University ²University of colorado boulder

September 11, 2023

Abstract

Stratospheric ozone intrusions have a significant impact on 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. Previous studies mainly focused on the case studies, and general knowledge of the spatial structure and large-scale dynamics underlying these intrusions is lacking. Thus, based on the Whole Atmosphere Community Climate Model, version 6 (WACCM6) simulation and a stratospheric origin ozone tracer, we identify the global hotspots of stratospheric intrusions: North America, Africa, the Mediterranean, and the Middle East, and investigate the underlying large-scale mechanisms. 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. Apart from others, the Middle East shows a relatively fast descent, likely related to the Asian summer monsoon circulation.

Hosted file

972206_0_art_file_11317671_rzccbc.docx available at https://authorea.com/users/656158/ articles/661649-the-evolutions-and-large-scale-mechanisms-of-summer-stratospheric-ozoneintrusion-across-global-hotspots

1 2 3 4	The Evolutions and Large-scale Mechanisms of Summer Stratospheric Ozone Intrusion across Global Hotspots
5	J. Lee ¹ , Y. Wu ¹ , and X. Wang ²
6	
7	¹ Lamont-Doherty Earth Observatory of Columbia University, New York, NY
8	² National Center for Atmospheric Research, Boulder, CO
9	
10	Corresponding author: Jaewon Lee (jaewonl@ldeo.columbia.edu)
11	
12	Key Points:
13 14	• There are 4 hotspots of summer ozone extremes due to stratospheric ozone, North America, Africa, the Mediterranean, and the Middle East.
15 16	• Summer stratospheric intrusions initiate in the jet axis region near tropopause by isentropic mixing.
17 18	• Climatological descent drives vertical transport in the lower troposphere and determines the location of the hotspots.

19 Abstract

Stratospheric ozone intrusions have a significant impact on surface ozone levels. 20 Especially in summer, intrusions can contribute to extreme ozone events because of preexisting 21 high ozone levels near the surface and cause serious health issues. Considering the increasing 22 trend of surface ozone level, an understanding of stratospheric ozone intrusion is necessary. 23 Previous studies mainly focused on the case studies, and general knowledge of the spatial 24 25 structure and large-scale dynamics underlying these intrusions is lacking. Thus, based on the Whole Atmosphere Community Climate Model, version 6 (WACCM6) simulation and a 26 stratospheric origin ozone tracer, we identify the global hotspots of stratospheric intrusions: 27 28 North America, Africa, the Mediterranean, and the Middle East, and investigate the underlying large-scale mechanisms. From the trajectory analysis, we find that the upper-level jet drives 29 isentropic mixing near the jet axis and initiates stratospheric ozone intrusion. Subsequently, 30 climatological descent at the lower troposphere brings the ozone down to the surface, which 31 explains the spatial preference of summertime stratospheric intrusion events. Apart from others, 32 the Middle East shows a relatively fast descent, likely related to the Asian summer monsoon 33 circulation. 34

35 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, because 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.

49 **1 Introduction**

Ozone is one of the most important chemicals in the atmosphere. The ozone layer in the 50 stratosphere absorbs most of the harmful UV radiation and protects the biosphere at the surface. 51 52 The energy absorbed by ozone is crucial to the thermal balance in the stratosphere and thereby 53 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., 54 1982; Pye, 1988; Reich, 1987; Smith et al., 2003). Exposure to high ozone concentrations is 55 56 harmful to humans also. The World Health Organization (WHO) recommends limiting outdoor activity when 8-hour mean daily maximum ozone levels exceed 50 ppbv (WHO, 2021). 57 Tropospheric ozone is primarily generated by reactions between ozone precursors such as 58 59 nitrogen oxides (NOx) and Volatile Organic Compounds (VOCs), which originate from both anthropogenic and natural sources (e.g., Finlayson-Pitts & Pitts Jr, 1993). In addition, occasional 60 intrusion of stratospheric ozone into the troposphere can be a major natural source of ozone in 61 certain locations (e.g., Appenzeller & Davies, 1992; Galani et al., 2003; Langford et al., 2015; 62 Lefohn et al., 2011; Lin et al., 2012; Ott et al., 2016; Stohl et al., 2000; Trickl et al., 2014; 63 Wakamatsu et al., 1989; Zanis et al., 2003). 64

65	The stratospheric intrusion can happen within multiple phenomena on different temporal
66	and spatial scales, including Rossby wave breaking (Holton et al., 1995), tropopause folding
67	(Shapiro, 1980), cut-off low (Price & Vaughan, 1993), and mesoscale convective system
68	(Poulida et al., 1996). The first three types of events can displace the tropopause on the
69	isentropic surface latitudinally and allow a massive amount of stratosphere-troposphere
70	exchange. Then sequentially, the large-scale disturbances and smaller-scale turbulences
71	irreversibly mix the stratospheric air with the troposphere (Holton et al., 1995; Johnson &
72	Viezee, 1981; Mahlman, 1997). The stratospheric intrusion within the mesoscale convective
73	system is suggested to be initiated from subsidence around the anvil cloud due to mass
74	conservation compensating upwelling tropospheric air. Then, the strong vertical shear can induce
75	differential advection that can wrap the subsidized stratospheric air (Pan et al., 2014; Phoenix et
76	al., 2020). Given the relatively high ozone level in the stratosphere and the fast increment of
77	ozone concentration during the intrusion, it is important to understand the nature of stratospheric
78	ozone intrusion to establish an effective policy and ozone warning system for the local
79	community.

Here, we focus on the stratospheric ozone intrusions that intrude deep into the 80 troposphere, transporting substantial amounts of ozone to near-surface levels and have 81 substantial potential impact on surface air quality. Škerlak et al. (2015) showed that shallow and 82 medium tropopause folds tend to occur near strong climatological wind in the subtropics, while 83 deep folds follow midlatitude storm tracks. A similar difference is also seen in the stratosphere-84 to-troposphere transport (STT) patterns between normal STTs and deep STTs (Škerlak et al., 85 2014). Also, Lin et al. (2015) showed more frequent springtime deep intrusions following La 86 87 Niña winters, although upper tropospheric ozone peaks after El Niño, underscoring different

88	responses by intrusion depth to climate variabilities. Despite their unique and direct impact on
89	the surface compared to general stratospheric intrusion, deep stratospheric intrusion processes
90	have not yet been studied extensively (Stohl et al., 2003).
91	Moreover, most studies focused on winter or spring intrusions when the intrusion is more
92	frequent and stratospheric ozone is abundant (Breeden et al., 2021; Johnson & Viezee, 1981;
93	Langford et al., 2009; Lin et al., 2012, 2015; Zhao et al., 2021). However, the consequence of
94	summertime intrusions on surface ozone could be more impactful. During summer,
95	photochemical ozone production reaches its peak, and events like thunderstorms, heat waves,
96	and wildfires are more frequent, which could produce ozone precursors (NOx and VOCs) and
97	increase the reaction rate due to high temperature (Gaudel et al., 2018; Jaffe & Wigder, 2012; Lu
98	et al., 2016; Murray, 2016; Solberg et al., 2008). Because the background ozone concentrations
99	are high, surpassing the health threshold with an additional stratospheric contribution is easy.
100	The model results also show that summer stratospheric origin ozone is not negligible and can
101	potentially trigger extreme ozone events near the surface (Wang et al., 2020). In addition,
102	summer stratospheric intrusions likely favor certain geographical locations (Škerlak et al., 2015).
103	Previous studies have shown that the west coast of the United States (Danielsen, 1980; Lefohn et
104	al., 2011, 2012; Wang et al., 2020) and the eastern Mediterranean (Akritidis et al., 2016; Zanis et
105	al., 2014) are influenced by stratospheric intrusion in summer. However, we lack the general
106	statistics that cover the entire globe and mechanisms that unify the events in different locations.
107	As we face an increasing trend of surface ozone (Gaudel et al., 2018), an overall understanding
108	of summer stratospheric intrusion is needed.
109	The difficulty of intrusion studies is distinguishing the stratospheric contribution in the

110 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 111 general understanding (Galani et al., 2003; Gronoff et al., 2021; Ott et al., 2016; Trickl et al., 112 2014; Wakamatsu et al., 1989; Xiong et al., 2022). To overcome these hardships and cover 113 diverse mechanisms, there are multiple approaches like tropopause folding identification 114 algorithms and back trajectory models on reanalysis and model data (e.g., Li et al., 2015; Škerlak 115 116 et al., 2015). Here, we will use an artificial tracer called stratospheric origin ozone (O₃S). O₃S is identical to the ozone in the stratosphere. However, once O₃S passes the tropopause and enters 117 the troposphere, O₃S does not have production routes and is removed at the same rate as ozone. 118 119 Therefore, tracking the stratospheric contribution throughout time and space is very useful through O₃S (Lin et al., 2012). It also allows us to cover intrusion events regardless of their 120 triggering mechanisms. 121

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: 1) Where and how often do we see extreme summer stratospheric ozone intrusion events? 2) What is the pathway for the intrusion? 3) What is the mechanism in common that drives these events across the global hotspots?

127 2 Methods

128 2.1 WACCM6 & O₃S

Our study is based on the daily summertime (June-August, JJA) ozone (O₃) and O₃S from the WACCM6 experiment. WACCM6 is a high-top chemistry-climate model of the Community Earth System Model version 2 (CESM2). The model has a horizontal resolution of 0.95°x1.25° (latitude x longitude) and 70 hybrid sigma levels in the vertical (~1.1 km resolution near UTLS).

It has high reproducibility on variabilities like sudden stratospheric warmings (SSWs) and 133 physical variables, such as temperature, wind, and chemicals, in the middle atmosphere, leading 134 to a better stratosphere-troposphere coupling than low-top models (Gettelman et al., 2019). 135 WACCM6 shares most of the physical parameterizations as the low-top Community Atmosphere 136 Model version 6 (CAM6) with an additional gravity wave scheme. The model chemistry 137 138 mechanism covers the troposphere up to the lower thermosphere. WACCM6 has interactive Community Land Model version 5 (CLM5) coupled as default, and our simulation is a fully 139 coupled ocean-atmosphere historical run (BWHIST). In addition to its better performance in 140 141 stratospheric dynamics, O₃ in both the stratosphere and troposphere has higher fidelity than previous versions (Emmons et al., 2020; Gettelman et al., 2019). Therefore, WACCM6 is 142 suitable for our study on summer stratospheric ozone intrusion (Wang et al., 2020). More 143 information about the model schemes and performance is available in Gettelman et al. (2019) 144 and Emmons et al. (2020). The available data period is from 1995 to 2014, but due to the spin-up 145 146 time of O_3S , we analyzed it from 1996. The O_3S tracer is implemented in the model, as Tilmes et al. (2016) demonstrated. This idealized tracer is identical to O₃ above the tropopause and is 147 removed via the same removal process as O_3 in the troposphere. However, unlike O_3 , it does not 148 149 have any production once in the troposphere. The tropopause here is defined following Reichler et al. (2003), which applied the lapse-rate tropopause definition, the lowest level where the 150 151 temperature lapse-rate decreases to 2 K/km or less (WMO, 1957).

152

2.2 Maximum Covariance Analysis (MCA)

We applied MCA on the daily 850 hPa O₃ and O₃S 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 multivariate datasets. It uses Singular Value Decomposition

(SVD) to extract spatial patterns and Principal Component (PC) timeseries of two datasets that 156 maximize the covariance (Bretherton et al., 1992). It can identify mechanisms that explain the 157 covarying patterns of the two variables and is suitable for our study to analyze the covariability 158 between near-surface O₃ and O₃S. The 850 hPa level is selected to identify the near-surface 159 extreme O₃ intrusion events. Although the high-altitude regions, such as the Tibetan Plateau, 160 161 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 162 163 regions in which the distance from the tropopause is similar. We defined daily anomalies after removing the linear trend and seasonal cycle for O₃ and O₃S. We used the Fourier transform on 164 the 19-year average of each day of the year and extracted up to the 4th harmonics to form a 165 seasonal cycle. This way, we could remove some noises from a relatively small number of years. 166

167 2.3 TRAJ3D model

The TRAJ3D model, a three-dimensional Lagrangian trajectory model, operates solely on 168 wind vectors to determine the tracer's location (Bowman, 1993; Bowman & Carrie, 2002). The 169 170 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 171 interpolation is conducted on the wind vector (Bowman et al., 2013). A large number of tracers 172 173 are released at 500 hPa, and the backward trajectory is integrated for a period of five days. Given that five days is considerably shorter than the typical lifetime of ozone in the free troposphere (a 174 few weeks), we treat O₃S as a passive tracer with an infinite lifetime. The stratospheric intrusion 175 is known to be dominated by isentropic mixing near the tropopause (Holton et al., 1995), which 176 occurs at a finer spatial and temporal resolution compared to the given daily vertical velocity. As 177 178 the pure trajectory model does not incorporate parameterization for convection, mixing, and

turbulence, vertical displacement should be considered with potential uncertainties (Smith et al.,2021).

181 **3 Results**

182 3.1 Global Hotspots of Stratospheric Intrusion Events

183 Hotspots in our study are defined as where extreme O₃ events are frequent, and the stratospheric contribution to these events is significant. Figure 1 shows the number of days per 184 summer (JJA for NH, DJF for SH) when the 850 hPa O₃ exceeds 56.7 ppbv and the contribution 185 of O₃S to O₃ is greater than 30%. The 850 hPa O₃ threshold is based on the summer median 186 187 (56.7 ppbv) of days when the 900-1000 hPa average O₃ exceeds the surface health threshold (50 ppbv; WHO). In summer, the boundary layer is elevated, and the mixing processes are vigorous 188 between 900 and 1000 hPa. In addition to the above two criteria, we further narrowed it to the 189 190 regions where O₃S accounts for over half of the O₃ variability within the hotspots. Specifically, we considered regions where the correlation between anomalous O₃S and O₃ is significant, with 191 an R-squared larger than 0.5. 192

Overall, most stratospheric intrusion events occur in the NH midlatitudes, between 20° 193 and 50°N (Fig. 1). Conversely, the SH exhibits fewer events mainly due to the low O₃ 194 concentration near the surface. The result reveals four global hotspots for stratospheric 195 intrusions: the West coast of North America (NA), the Northwest coast of Africa (Af), the 196 Eastern Mediterranean (MD), and the Middle East near Iran and Pakistan (ME). Other regions, 197 198 such as the northern Tibetan Plateau and eastern North America and Asia, are not the focus of this study due to low correlations between anomalous O₃ and O₃S. Interestingly, except for the 199 ME hotspot, the remaining hotspots are the Mediterranean climate regime. These hotspots 200

qualitatively align with the NH hotspots for tropopause folding events documented in previous 201 studies (Škerlak et al., 2015; Sprenger et al., 2003). The small discrepancies could arise from the 202 differing altitudes of focus (near-surface vs. near-tropopause). Among the hotspots, the NA 203 region exhibits the highest frequency, with approximately 15 events per summer, and other 204 hotspots also experience a minimum of six events per summer. A sensitivity test on the ratio 205 206 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 207 our discussion as they result in significantly reduced event frequencies. 208







215

We also analyzed the frequencies of events exceeding 99% of all the 850 hPa O₃S across the NH for each season to see where the relative intrusion hotspots are for each season (Fig. S1).

218	Although summer has the lowest 99% O ₃ S concentration (19.14 ppbv), it clearly shows the four
219	global hotspots we have seen in Fig. 1. Interestingly, only summer shows such four global
220	hotspots with a strong zonal asymmetry, while other seasons are more zonally symmetric. The
221	NA hotspot always exists throughout the year, but other hotspots disappear in other seasons.
222	Meanwhile, weaker hotspots are seen in different locations in other seasons, for example, over
223	the northern Atlantic and the east coast of North America. This result emphasizes the unique
224	features of summer stratospheric ozone intrusion and its impact on surface ozone extreme events.
225	Also, similar patterns in summer between Fig. 1 and Fig. S1c indicate that stratospheric
226	intrusions are indeed important in extreme ozone events near the surface.
227	3.2 MCA results
228	We applied the MCA on each identified hotspot between 850 hPa O_3S and O_3 to
229	determine the major mechanism for the covariability of the two variables (Fig. S2). In this paper,
230	we will study the leading mode of each hotspot since it is the dominant mode and represents an
231	overall intensification in the region (Fig. 2). The PC timeseries are divided by the standard
232	deviation, and loading vectors are multiplied by that standard deviation. The leading mode
233	outstands the other modes for all the hotspots, especially at the ME hotspot (74.87%). Even the
234	lowest covariance fraction for the leading mode is significantly high (31.96% at the Af hotspot).
235	Thus, the first mode of MCA can represent the large-scale conditions that simultaneously
236	intensify both O ₃ S and O ₃ .



Figure 2. The 850 hPa O₃S spatial patterns from MCA leading mode are shown for global hotspots: NA, Af, MD, and ME. The MCA has been conducted on the daily 850 hPa O₃S and O₃ anomalies in JJA 1996-2014. The covariance fraction explained by the leading mode is written in the parenthesis next to each hotspot.

242

237

The extreme events hereafter are days when both O₃S and O₃ PC timeseries for the 243 leading mode exceed one standard deviation level, and we count the continuous extreme days as 244 a single event. We identified 61, 65, 48, and 51 total events in NA, Af, MD, and ME during 19 245 246 years, respectively, which is about 3% of the total days in summer (Fig. S3). Although most events last 1-2 days, long-lived events can last as long as 10 days (not shown). Figure 2 shows 247 that NA and Af patterns peak in the nearby ocean, while MD and ME peaks are on the coast with 248 249 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 (Fig. 2). To 250 determine the dominance of the leading mode among all extreme events, we compared the above 251 252 extreme events associated with the leading mode with events identified in Fig. 1. For each grid

over the peaks of hotspot regions, about 30-50% of the MCA-identified events overlap, and the
 O₃S 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 257 258 increasing or decreasing trend in the number of extreme events in any hotspot (not shown). We also analyzed if there is any preference in the timing of events within the summer for each 259 hotspot (Fig. S3). Most locations have a weak intraseasonal variability, except MD, which has a 260 strong intraseasonal variability. The MD hotspot shows a large preference in the early summer 261 and almost no events in August. This is an unexpected result since previous studies emphasized 262 that Etesian wind in the Mediterranean strongly correlates with the intrusion, which peaks in late 263 summer (Dafka et al., 2021; Tyrlis & Lelieveld, 2013). Although a few questions exist in the 264 intraseasonal variability of extreme events, we will focus on the general features of extreme 265 266 events to understand the commonalities between events.

267 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 (Fig. 3). Based on the O_3S composites, a box region was selected to encompass the potential trajectories associated with each hotspot (Fig. S2). 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. 275 Consequently, the use of a back trajectory model is necessary to differentiate the intrusion from



the background O_3S above 500 hPa.



Figure 3. The box averaged O₃S anomaly for each level from 8 days prior to the events to the event days. The box regions are defined in Fig. S2. The negative values are grayed out. The white areas are topography.

281

The TRAJ3D model was employed to estimate the trajectories of summer stratospheric ozone intrusion above 500 hPa. First, a box region enclosing the statistically significant maximum of the 500 hPa O₃S anomaly three days before the events is designated for each hotspot. This specific date is chosen as the intrusion signal at 500 hPa O₃S anomaly displays the most prominence. Subsequently, tracers are released at the significant area of each intrusion case within the assigned box region, where O₃S 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 289 individual trajectory among a large set of trajectories for each ensemble, which has different 290 initial locations from each other. Within the significant area for each ensemble, 1000 trajectories 291 are initiated at randomly selected grid points allowing duplication after regridding to 0.5°x0.5° 292 resolution. For example, the NA hotspot has 61 ensembles, and each ensemble has 1000 293 294 trajectories, total of 61000 trajectories (61 ensembles x 1000 trajectories). In different ensembles, both the initial tracer locations and the wind field can change, as significant area and event date 295 differ by the ensemble. 296

The trajectories for each hotspot are presented in the left column of Fig. 4. The mean 297 trajectories for each ensemble are depicted as gray lines. The colored line shows the mean 298 trajectory for the hotspot, which is the mean of gray lines, with height represented in color. For 299 instance, in the NA hotspot, gray lines show 61 ensembles, which is the mean of 1000 300 trajectories for each ensemble. The colored line is an average of 61 ensembles, which is the mean 301 of 61000 trajectories. Generally, for all the hotspots, the trajectories exhibit a southeastward 302 descent that crosses the jet axis, which is denoted by the red contours. Consistent with previous 303 studies, the southeastward descending pathway is attributed to the tilted isentropic surfaces and 304 305 the strong climatological westerlies in the midlatitudes (Akritidis et al., 2016; B. Škerlak et al., 2014; Sprenger & Wernli, 2003). Three hotspots, i.e., NA, Af, and MD, continue their 306 307 southeastward descent to 850 hPa. However, ME trajectories experience a rapid descent near the 308 500 hPa into the lower troposphere. Then, the northerly wind transports O₃S toward the 850 hPa hotspot (not shown). We will briefly discuss how the ME hotspot trajectories and governing 309 310 mechanisms differ from others and how they relate to the Asian summer monsoon in the next 311 section.





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 to 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 days to 8 days before the event, if the trajectory

passes the jet between 200 to 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: NA (1st row), Af (2nd row), MD (3rd

- 324 row), and ME (4^{th} row).
- 325

319

320

321

322

323

Previous studies have highlighted that during boreal summer, NH STT exhibits two 326 latitude maxima: one over the midlatitudes and the other over the subtropics (Hsu et al., 2005; 327 Jing et al., 2004; Tang et al., 2011). In the midlatitudes, deep convection over continents plays a 328 significant role, whereas, in the subtropics, it is primarily via Rossby wave breaking (RWB) over 329 the ocean. However, Škerlak et al. (2014) demonstrated that summer deep stratospheric 330 intrusions originate from locations distinct from the mentioned STT maxima. Given the 331 considerable number of ensembles crossing the jet axis, we hypothesize that isentropic mixing 332 near the jet axis is the source of O₃S for deep STT in summer (Holton et al., 1995). We 333 conducted a quantitative analysis to ascertain whether the estimated trajectories intersect the jet 334 335 axis. Figure 5 presents an example of a single case with the zonal wind at 200 hPa eight days before the event. The tracers are denoted by dots, with tracers in the jet axis region depicted in 336 337 blue. The jet axis region is defined as where the zonal wind exceeds 20 m/s (red shading). The jet 338 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 339 Fig. 5, 38.1% of the trajectories are found to cross the jet axis region at the given level and time. 340 341 The middle column of Fig. 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 O₃S for deep STT in summer.



349

Figure 5. A case study of tracer ensembles 8 days before the event at 200 hPa. 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.

353

We further assessed whether trajectories crossing the jet axis region are statistically significant. For each ensemble, at multiple pressure levels and dates preceding the event (300 hPa, 250hPa, and 200 hPa, 4 to 8 days prior), we computed the area ratio (r) of the jet axis region to the box region enclosing the maximum and minimum latitude and longitude of the tracers (Fig. 5). Assuming a completely random process, each tracer can be considered to follow a binomial distribution with a sample size (N) of 1000. Applying the central limit theorem, this

360	distribution can be approximated by a normal distribution with Nr as the mean and $Nr(1-r)$ as the
361	variance. Through standardizing, we found that around 80% of the cases exhibit a significance
362	exceeding two standard deviations for each hotspot (2.5%, one-sided). For instance, in the NA
363	hotspot, 79.3% out of 915 cases (61 ensembles x 5 dates x 3 pressure levels) have passed the
364	significance test. The result indicates that, regardless of the time and level, there is a significant
365	likelihood of the trajectory intersecting the jet axis. A sensitivity test was performed by releasing
366	tracers at the maximum within 2 and 4 days before the event, yielding similar substantial
367	probabilities of stratospheric intrusion crossing the jet axis (not shown).
368	We also examined the potential relationship between the stratospheric intrusion events
369	and RWB, a major driver of isentropic mixing, to determine if any association exists. However,
370	our results did not reveal a significant relationship between the two (not shown). While this
371	finding may differ from previous studies (Holton et al., 1995), we do not consider it
372	contradictory. The events we analyzed may differ from those examined in previous studies,
373	which could lead to variations in the observed relationship.
374	Furthermore, we investigated the preferred crossing location of trajectories relative to the
375	jet core in terms of latitude at 200 hPa. The location on a day with a maximum number of tracers
376	on the jet axis between 4 to 8 days before the event is examined. As shown in the right column of
377	Fig. 4, the distribution is centered near the core region, with a slight poleward tilt except for the
378	ME hotspot. A sensitivity test on different pressure levels shows a slight shift to the poleward
379	flank on lower pressure levels. Still, it does not affect the general feature of the relative locations
380	(not shown). This finding is consistent with Yang et al. (2016), which noted that summer
381	intrusions exhibit a unique characteristic wherein the peak ozone flux into the troposphere occurs
382	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.

385 3.4 Vertical Transport in the Lower Troposphere

Now that we know the pathway to the mid-troposphere, we further address the question of what contributes to the vertical descent in the lower troposphere. 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:

391
$$\frac{\partial O3S_a}{\partial t} = -\omega_c \frac{\partial O3S_a}{\partial p} - \omega_a \frac{\partial O3S_c}{\partial p} - \left(\omega_a \frac{\partial O3S_a}{\partial p}\right)_a + (Zonal) + (Meridional) + (Residuals),$$

where subscript a indicates anomaly and c indicates climatological seasonal cycle. We first 392 decomposed the tendency into zonal, meridional, vertical transport, and residual terms. Then, we 393 further separated vertical transport into contributions from climatological wind $(-\omega_c \frac{\partial O3S_a}{\partial p})$, 394 anomalous wind $(-\omega_a \frac{\partial O3S_c}{\partial p})$, and nonlinear $(-(\omega_a \frac{\partial O3S_a}{\partial p})_a)$ terms, as the focus is on the 395 mechanism that brings air down. We smoothed the data by taking a 5°x5° moving box mean for 396 each term. Then, we examined the maximum tendency for each pressure level near the hotspots 397 and 10 to 0 days before the events. The analysis reveals a greater magnitude of horizontal 398 transport compared to vertical transport (not shown), which is expected due to larger horizontal 399 wind velocities. However, since our question is on the mechanism of vertical transport to the 400 near-surface level, we focus on the common factors that contribute to the vertical transport 401 across the global hotspots. 402



Figure 6. The relative importance of vertical transport decomposition to the total 404 vertical transport. Red, blue, and green indicate climatological wind-driven, anomalous 405 wind-driven, and nonlinear terms, respectively. The total vertical transport is calculated in 406 the tendency equation on the maximum tendency at a given time and pressure level. To 407 408 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% 409 for each hotspot. Three hotspots (NA, Af, and MD) are calculated on day -1 at 850 hPa, 410 while ME is calculated on day -3 at 600 hPa. The terminations of each term and their 411 formula follow the equation in section 3.4, and a detailed explanation of the time and level 412 selection is given there. 413

414

403

The role of climatological wind-driven vertical transport is substantial in all hotspots (red bars in Fig. 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). Based on the trajectory, the ME hotspot tendency is calculated for -3 days at 600 hPa. We see a gradual

descent from the trajectory in three hotspots except ME, where strong descent happens a few 419 days ahead of the event and then horizontally shifts to the event region. The climatological 420 vertical transport dominates in the lower troposphere from 600 to 850 hPa near the hotspots, 421 even out of the maximum tendency region. This climatological dominance explains the co-422 location of climatological descent regions (mainly the Mediterranean climate regime) and global 423 424 hotspots of stratospheric intrusion. In addition, it's worth noting that the climatological winddriven vertical transport also depends on the vertical gradient of the O₃S anomaly. Our 425 understanding is that the upper-level system induces this anomaly, as demonstrated in Section 426 3.3. Once the gradient is established, the climatological descent transports the anomaly to the 427 near-surface level. The upper-level dynamics that initiate the intrusion is attributable to jet 428 dynamics, as our trajectory results and previous studies suggested (Fig. 3; Wang et al., 2020). 429 Although climatological descent in the lower troposphere does not elucidate all the intrusion 430 processes and determines the location, it has a considerable contribution. This also explains the 431 distinct geographical locations of O₃S extremes in the summer compared to other seasons. The 432 climatological descent is prominent during the summer due to the anticyclone formation in the 433 ocean (NA and Af; Rodwell & Hoskins, 2001) and the Asian summer monsoon (MD and ME; 434 435 Wu et al., 2018).

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° north. 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 two 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).

447 **4 Conclusion and discussions**

We identify summertime stratospheric intrusion hotspots using a state-of-the-art 448 449 chemistry climate model and a stratospheric origin tracer, and investigate the pathway and mechanism of these intrusion events. Maximum covariance analysis demonstrates that there are 450 four global hotspots with frequent near-surface summer ozone extreme events due to 451 stratospheric intrusion: the West coast of North America (NA), the Northwest coast of Africa 452 453 (Af), the Eastern Mediterranean (MD), and the Middle East near Iran and Pakistan (ME). To elucidate the trajectory and underlying mechanisms of each hotspot, we employ the Lagrangian 454 pure transport model (TRAJ3D). The stratospheric intrusions above 500 hPa generally follow a 455 456 southeastward descent and traverse the jet axis as they enter the troposphere. The estimated trajectories align well with previous studies and are potentially driven by isentropic mixing near 457 the tropopause (Škerlak et al., 2014; Yang et al., 2016). Furthermore, budget analysis shows that 458 459 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. This explains the global 460 hotspots being located in the strong climatological descent regions, mostly in the Mediterranean 461 climate regime. 462

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 values 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).

470 There are still several unresolved issues regarding summer stratospheric intrusions. The wave dynamics near the upper tropospheric jet and persistent climatological descent cannot 471 explain the rareness of the intrusion events. We propose that these two pieces must be connected 472 with a suitable horizontal wind, which is a potential third key factor, for extreme events to occur. 473 If the ozone flux in the upper troposphere does not reach the climatological descent region, it 474 will not be able to reach the near-surface level. Another possibility is that either the upper-475 tropospheric wave activities or the climatological descent is extreme during these events. 476 However, the intraseasonal variability of the climatological descent is likely too weak to explain 477 the occurrence of extreme events. A mechanistic study on the upper tropospheric dynamics in 478 summer could fill the gap in our understanding of the summer stratospheric intrusion. The 479 intraseasonal and interannual variability of the summer stratospheric intrusion also needs further 480 481 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 482 483 NA hotspot does not show a connection to ENSO (not shown), whereas the spring deep STT 484 increases during La Ninas (Lin et al., 2015; Albers et al., 2022).

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 488 model and data set to test the robustness. Our findings can potentially contribute to the

forecasting of extreme ozone events in summer and benefit policymakers in establishing an earlywarning system.

491

492 Acknowledgments

493 We thank S. Tilmes of the National Center for Atmospheric Research for providing the

494 WACCM6 model results. We also thank K. P. Bowman of Texas A&M University for the

495 TRAJ3D code, W. P. Smith of the National Center for Atmospheric Research for great help with

the trajectory analysis, and W. Randel of the National Center for Atmospheric Research for

497 helpful discussion on dynamics. J. L. and Y. W. are supported by the National Science

498 Foundation award AGS-1802248.

499

500 Data Availability Statement

501 The processed data used for the analysis in the study are available at Columbia University 502 Academic Commons via (Lee, 2023) All WACCM6 simulations were carried out on the 503 Cheyenne high-performance computing platform and are available at NCAR's Campaign

504 Storage upon acceptance.

505

506 **References**

507 Akritidis, D., Pozzer, A., Zanis, P., Tyrlis, E., Škerlak, B., Sprenger, M., & Lelieveld, J. (2016).

508 On the role of tropopause folds in summertime tropospheric ozone over the eastern

509 Mediterranean and the Middle East. *Atmospheric Chemistry and Physics*, *16*(21), 14025–14039.

510 <u>https://doi.org/10.5194/acp-16-14025-2016</u>

- Albers, J. R., Butler, A. H., Langford, A. O., Elsbury, D., & Breeden, M. L. (2022). Dynamics of
- 512 ENSO-driven stratosphere-to-troposphere transport of ozone over North America. *Atmospheric*
- 513 *Chemistry and Physics*, 22(19), 13035–13048. https://doi.org/10.5194/acp-22-13035-2022
- Anjum, M. S., Ali, S. M., Imad-ud-din, M., Subhani, M. A., Anwar, M. N., Nizami, A.-S., et al.
- 515 (2021). An Emerged Challenge of Air Pollution and Ever-Increasing Particulate Matter in
- 516 Pakistan; A Critical Review. Journal of Hazardous Materials, 402, 123943.
- 517 https://doi.org/10.1016/j.jhazmat.2020.123943
- 518 Appenzeller, C., & Davies, H. C. (1992). Structure of stratospheric intrusions into the
- 519 troposphere. *Nature*, *358*(6387), 570–572.
- 520 Bowman, K. P. (1993). Large-scale isentropic mixing properties of the Antarctic polar vortex
- from analyzed winds. *Journal of Geophysical Research: Atmospheres*, 98(D12), 23013–23027.
- 522 Bowman, K. P., & Carrie, G. D. (2002). The mean-meridional transport circulation of the
- troposphere in an idealized GCM. Journal of the Atmospheric Sciences, 59(9), 1502–1514.
- 524 Bowman, K. P., Lin, J. C., Stohl, A., Draxler, R., Konopka, P., Andrews, A., & Brunner, D.
- 525 (2013). Input data requirements for Lagrangian trajectory models. *Bulletin of the American*
- 526 *Meteorological Society*, 94(7), 1051–1058.
- 527 Breeden, M. L., Butler, A. H., Albers, J. R., Sprenger, M., & Langford, A. O. (2021). The spring
- transition of the North Pacific jet and its relation to deep stratosphere-to-troposphere mass
- transport over western North America. *Atmospheric Chemistry and Physics*, 21(4), 2781–2794.
- 530 https://doi.org/10.5194/acp-21-2781-2021
- 531 Bretherton, C. S., Smith, C., & Wallace, J. M. (1992). An Intercomparison of Methods for
- 532 Finding Coupled Patterns in Climate Data. *Journal of Climate*, 5(6), 541–560.
- 533 https://doi.org/10.1175/1520-0442(1992)005<0541:AIOMFF>2.0.CO;2

- 534 Dafka, S., Akritidis, D., Zanis, P., Pozzer, A., Xoplaki, E., Luterbacher, J., & Zerefos, C. (2021).
- On the link between the Etesian winds, tropopause folds and tropospheric ozone over the Eastern
- 536 Mediterranean during summer. *Atmospheric Research*, 248, 105161.
- 537 Danielsen, E. F. (1980). Stratospheric source for unexpectedly large values of ozone measured
- over the Pacific Ocean during Gametag, August 1977. Journal of Geophysical Research:
- 539 *Oceans*, 85(C1), 401–412.
- 540 Emmons, L. K., Schwantes, R. H., Orlando, J. J., Tyndall, G., Kinnison, D., Lamarque, J.-F., et
- al. (2020). The chemistry mechanism in the community earth system model version 2 (CESM2).
- 542 Journal of Advances in Modeling Earth Systems, 12(4).
- 543 Finlayson-Pitts, B. J., & Pitts Jr, J. N. (1993). Atmospheric chemistry of tropospheric ozone
- formation: scientific and regulatory implications. *Air & Waste*, 43(8), 1091–1100.
- 545 Galani, E., Balis, D., Zanis, P., Zerefos, C., Papayannis, A., Wernli, H., & Gerasopoulos, E.
- 546 (2003). Observations of stratosphere-to-troposphere transport events over the eastern
- 547 Mediterranean using a ground-based lidar system. *Journal of Geophysical Research:*
- 548 *Atmospheres*, *108*(D12).
- 549 Gaudel, A., Cooper, O. R., Ancellet, G., Barret, B., Boynard, A., Burrows, J. P., et al. (2018).
- 550 Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric
- ozone relevant to climate and global atmospheric chemistry model evaluation. *Elementa: Science*
- 552 of the Anthropocene, 6.
- 553 Gettelman, A., Mills, M. J., Kinnison, D. E., Garcia, R. R., Smith, A. K., Marsh, D. R., et al.
- 554 (2019). The whole atmosphere community climate model version 6 (WACCM6). Journal of
- 555 *Geophysical Research: Atmospheres*, *124*(23), 12380–12403.

- 556 Gronoff, G., Berkoff, T., Knowland, K. E., Lei, L., Shook, M., Fabbri, B., et al. (2021). Case
- 557 study of stratospheric Intrusion above Hampton, Virginia: lidar-observation and modeling
- analysis. *Atmospheric Environment*, 259, 118498.
- Heck, W. W., Taylor, O. C., Adams, R., Bingham, G., Miller, J., Preston, E., & Weinstein, L.
- 560 (1982). Assessment of Crop Loss from Ozone. Journal of the Air Pollution Control Association,
- 561 *32*(4), 353–361. https://doi.org/10.1080/00022470.1982.10465408
- 562 Holton, J. R., Haynes, P. H., McIntyre, M. E., Douglass, A. R., Rood, R. B., & Pfister, L. (1995).
- 563 Stratosphere-troposphere exchange. *Reviews of Geophysics*, *33*(4), 403.
- 564 https://doi.org/10.1029/95RG02097
- 565 Hsu, J., Prather, M. J., & Wild, O. (2005). Diagnosing the stratosphere-to-troposphere flux of

ozone in a chemistry transport model. Journal of Geophysical Research: Atmospheres,

- 567 *110*(D19).
- Jaffe, D. A., & Wigder, N. L. (2012). Ozone production from wildfires: A critical review.
- 569 *Atmospheric Environment*, 51, 1–10.
- Jing, P., Cunnold, D. M., Wang, H. J., & Yang, E. S. (2004). Isentropic cross-tropopause ozone
- transport in the Northern Hemisphere. *Journal of the Atmospheric Sciences*, *61*(9), 1068–1078.
- Johnson, W. B., & Viezee, W. (1981). Stratospheric ozone in the lower troposphere —I.
- 573 Presentation and interpretation of aircraft measurements. Atmospheric Environment (1967),
- 574 15(7), 1309–1323. https://doi.org/10.1016/0004-6981(81)90325-5
- 575 Langford, A. O., Aikin, K. C., Eubank, C. S., & Williams, E. J. (2009). Stratospheric
- 576 contribution to high surface ozone in Colorado during springtime. *Geophysical Research Letters*,
- 577 *36*(12).

- 578 Langford, A. O., Senff, C. J., Alvarez Ii, R. J., Brioude, J., Cooper, O. R., Holloway, J. S., et al.
- 579 (2015). An overview of the 2013 Las Vegas Ozone Study (LVOS): Impact of stratospheric
- intrusions and long-range transport on surface air quality. Atmospheric Environment, 109, 305-
- 581 322.
- 582 Lee, J. (2023). Data: The Evolutions and Large-scale Mechanisms of Summer Stratospheric
- 583 *Ozone Intrusion across Global Hotspots*. Columbia University Academic Commons.
- 584 <u>https://doi.org/10.7916/5pcm-er89</u>
- Lefohn, A. S., Wernli, H., Shadwick, D., Limbach, S., Oltmans, S. J., & Shapiro, M. (2011). The
- importance of stratospheric-tropospheric transport in affecting surface ozone concentrations in
- the western and northern tier of the United States. *Atmospheric Environment*, 45(28), 4845–
- 588 **4857**.
- Lefohn, A. S., Wernli, H., Shadwick, D., Oltmans, S. J., & Shapiro, M. (2012). Quantifying the
- 590 importance of stratospheric-tropospheric transport on surface ozone concentrations at high-and
- ⁵⁹¹ low-elevation monitoring sites in the United States. *Atmospheric Environment*, 62, 646–656.
- Li, D., Bian, J., & Fan, Q. (2015). A deep stratospheric intrusion associated with an intense cut-
- ⁵⁹³ off low event over East Asia. *Science China Earth Sciences*, *58*, 116–128.
- Lin, M., Fiore, A. M., Cooper, O. R., Horowitz, L. W., Langford, A. O., Levy, H., et al. (2012).
- 595 Springtime high surface ozone events over the western United States: Quantifying the role of
- 596 stratospheric intrusions. Journal of Geophysical Research: Atmospheres, 117(D21).
- 597 Lin, M., Fiore, A. M., Horowitz, L. W., Langford, A. O., Oltmans, S. J., Tarasick, D., & Rieder,
- 598 H. E. (2015). Climate variability modulates western US ozone air quality in spring via deep
- 599 stratospheric intrusions. *Nature Communications*, 6(1), 7105.
- 600 https://doi.org/10.1038/ncomms8105

- Lu, X., Zhang, L., Yue, X., Zhang, J., Jaffe, D. A., Stohl, A., et al. (2016). Wildfire influences on
- the variability and trend of summer surface ozone in the mountainous western United States.
- Atmospheric Chemistry and Physics, 16(22), 14687–14702.
- Mahlman, J. D. (1997). Dynamics of transport processes in the upper troposphere. *Science*,
 276(5315), 1079–1083.
- Mehmood, T., Ahmad, I., Bibi, S., Mustafa, B., & Ali, I. (2020). Insight into monsoon for
- shaping the air quality of Islamabad, Pakistan: Comparing the magnitude of health risk
- associated with PM 10 and PM 2.5 exposure. Journal of the Air & Waste Management
- 609 Association, 70(12), 1340–1355. https://doi.org/10.1080/10962247.2020.1813838
- Murray, L. T. (2016). Lightning NO x and impacts on air quality. *Current Pollution Reports*, *2*,
 115–133.
- 612 Ott, L. E., Duncan, B. N., Thompson, A. M., Diskin, G., Fasnacht, Z., Langford, A. O., et al.
- 613 (2016). Frequency and impact of summertime stratospheric intrusions over Maryland during
- 614 DISCOVER-AQ (2011): New evidence from NASA's GEOS-5 simulations. Journal of
- 615 *Geophysical Research: Atmospheres*, *121*(7), 3687–3706.
- Pan, L. L., Homeyer, C. R., Honomichl, S., Ridley, B. A., Weisman, M., Barth, M. C., et al.
- 617 (2014). Thunderstorms enhance tropospheric ozone by wrapping and shedding stratospheric air.
- 618 *Geophysical Research Letters*, *41*(22), 7785–7790.
- 619 Phoenix, D. B., Homeyer, C. R., Barth, M. C., & Trier, S. B. (2020). Mechanisms Responsible
- 620 for Stratosphere-to-Troposphere Transport Around a Mesoscale Convective System Anvil.
- *Journal of Geophysical Research: Atmospheres*, *125*(10). https://doi.org/10.1029/2019JD032016

- 622 Poulida, O., Dickerson, R. R., & Heymsfield, A. (1996). Stratosphere-troposphere exchange in a
- 623 midlatitude mesoscale convective complex: 1. Observations. *Journal of Geophysical Research:*
- 624 Atmospheres, 101(D3), 6823–6836. https://doi.org/10.1029/95JD03523
- Price, J. D., & Vaughan, G. (1993). The potential for stratosphere-troposphere exchange in cut-
- off-low systems. *Quarterly Journal of the Royal Meteorological Society*, *119*(510), 343–365.
- 627 Pye, J. M. (1988). Impact of Ozone on the Growth and Yield of Trees: A Review. Journal of
- 628 *Environmental Quality*, *17*(3), 347–360.
- 629 https://doi.org/10.2134/jeq1988.00472425001700030003x
- 630 Reich, P. B. (1987). Quantifying plant response to ozone: a unifying theory. *Tree Physiology*,
- 631 3(1), 63–91. https://doi.org/10.1093/treephys/3.1.63
- Reichler, T., Dameris, M., & Sausen, R. (2003). Determining the tropopause height from gridded
 data. *Geophysical Research Letters*, *30*(20).
- Rodwell, M. J., & Hoskins, B. J. (2001). Subtropical anticyclones and summer monsoons.
- 635 *Journal of Climate*, *14*(15), 3192–3211.
- 636 Schoeberl, M. R., & Hartmann, D. L. (1991). The Dynamics of the Stratospheric Polar Vortex
- and Its Relation to Springtime Ozone Depletions. *Science*, *251*(4989), 46–52.
- 638 https://doi.org/10.1126/science.251.4989.46
- 639 Shapiro, M. A. (1980). Turbulent mixing within tropopause folds as a mechanism for the
- exchange of chemical constituents between the stratosphere and troposphere. *Journal of*
- 641 *Atmospheric Sciences*, *37*(5), 994–1004.
- 642 Škerlak, B., Sprenger, M., & Wernli, H. (2014). A global climatology of stratosphere–
- troposphere exchange using the ERA-Interim data set from 1979 to 2011. Atmospheric
- 644 *Chemistry and Physics*, 14(2), 913–937. https://doi.org/10.5194/acp-14-913-2014

- 645 Škerlak, B., Sprenger, M., Pfahl, S., Tyrlis, E., & Wernli, H. (2015). Tropopause folds in ERA-
- 646 Interim: Global climatology and relation to extreme weather events. *Journal of Geophysical*
- 647 *Research: Atmospheres*, *120*(10), 4860–4877.
- 648 Škerlak, B., Pfahl, S., Sprenger, M., & Wernli, H. (2019). A numerical process study on the rapid
- transport of stratospheric air down to the surface over western North America and the Tibetan
- 650 Plateau. Atmospheric Chemistry and Physics, 19(9), 6535–6549. https://doi.org/10.5194/acp-19-
- 651 6535-2019
- 652 Smith, G., Coulston, J., Jepsen, E., & Prichard, T. (2003). A national ozone biomonitoring
- program–results from field surveys of ozone sensitive plants in northeastern forests (1994–2000).
- *Environmental Monitoring and Assessment*, 87, 271-291.
- 655 Smith, W. P., Pan, L. L., Honomichl, S. B., Chelpon, S. M., Ueyama, R., & Pfister, L. (2021).
- 656 Diagnostics of Convective Transport Over the Tropical Western Pacific From Trajectory
- 657 Analyses. Journal of Geophysical Research: Atmospheres, 126(17).
- 658 https://doi.org/10.1029/2020JD034341
- 659 Solberg, S., Hov, Ø., Søvde, A., Isaksen, I. S. A., Coddeville, P., De Backer, H., et al. (2008).
- European surface ozone in the extreme summer 2003. *Journal of Geophysical Research:*
- 661 *Atmospheres*, *113*(D7).
- 662 Sprenger, M., & Wernli, H. (2003). A northern hemispheric climatology of cross-tropopause
- exchange for the ERA15 time period (1979–1993). Journal of Geophysical Research:
- 664 *Atmospheres*, *108*(D12).
- 665 Sprenger, M., Croci Maspoli, M., & Wernli, H. (2003). Tropopause folds and cross-tropopause
- exchange: A global investigation based upon ECMWF analyses for the time period March 2000
- to February 2001. Journal of Geophysical Research: Atmospheres, 108(D12).

- 668 Stohl, A., Spichtinger-Rakowsky, N., Bonasoni, P., Feldmann, H., Memmesheimer, M., Scheel,
- 669 H. E., et al. (2000). The influence of stratospheric intrusions on alpine ozone concentrations.
- 670 *Atmospheric Environment*, *34*(9), 1323–1354.
- 671 Stohl, A., Wernli, H., James, P., Bourqui, M., Forster, C., Liniger, M. A., et al. (2003). A New
- 672 Perspective of Stratosphere–Troposphere Exchange. Bulletin of the American Meteorological
- 673 Society, 84(11), 1565–1574. https://doi.org/10.1175/BAMS-84-11-1565
- Tang, Q., Prather, M. J., & Hsu, J. (2011). Stratosphere-troposphere exchange ozone flux related
 to deep convection. *Geophysical Research Letters*, *38*(3).
- Tilmes, S., Lamarque, J.-F., Emmons, L. K., Kinnison, D. E., Marsh, D., Garcia, R. R., et al.
- (2016). Representation of the community earth system model (CESM1) CAM4-chem within the
- chemistry-climate model initiative (CCMI). *Geoscientific Model Development*, 9(5), 1853–1890.
- 679 Trickl, T., Vogelmann, H., Giehl, H., Scheel, H.-E., Sprenger, M., & Stohl, A. (2014). How
- stratospheric are deep stratospheric intrusions? *Atmospheric Chemistry and Physics*, *14*(18),
 9941–9961.
- Tyrlis, E., & Lelieveld, J. (2013). Climatology and dynamics of the summer Etesian winds over
- the eastern Mediterranean. *Journal of the Atmospheric Sciences*, 70(11), 3374–3396.
- Wakamatsu, S., Uno, I., Ueda, H., Uehara, K., & Tateishi, H. (1989). Observational study of
- 685 stratospheric ozone intrusions into the lower troposphere. Atmospheric Environment (1967),
- 686 *23*(8), 1815–1826.
- 687 Wang, X., Wu, Y., Randel, W., & Tilmes, S. (2020). Stratospheric contribution to the
- summertime high surface ozone events over the western united states. *Environmental Research*
- 689 Letters, 15(10), 1040a6. <u>https://doi.org/10.1088/1748-9326/abba53</u>

- 690 WHO (2021). WHO global air quality guidelines: particulate matter (PM2. 5 and PM10), ozone,
- nitrogen dioxide, sulfur dioxide and carbon monoxide: executive summary.
- 692 WMO (1957). Meteorology—A three-dimensional science: Second session of the commission
- 693 for aerology. *WMO Bull.*, *4*(4), 134–138.
- Wu, Y., Chen, G., Taylor, L., & Zhang, P. (2018). On the linkage between the Asian summer
- monsoon and tropopause folds. *Journal of Geophysical Research: Atmospheres*, *123*(4), 2037–
 2049.
- 697 Xiong, X., Liu, X., Wu, W., Knowland, K. E., Yang, Q., Welsh, J., & Zhou, D. K. (2022).
- 698 Satellite observation of stratospheric intrusions and ozone transport using CrIS on SNPP.
- 699 Atmospheric Environment, 273, 118956.
- Yang, H., Chen, G., Tang, Q., & Hess, P. (2016). Quantifying isentropic stratosphere-
- troposphere exchange of ozone. *Journal of Geophysical Research: Atmospheres*, *121*(7), 3372–
 3387.
- Zanis, P., Trickl, T., Stohl, A., Wernli, H., Cooper, O., Zerefos, C., et al. (2003). Forecast,
- observation and modelling of a deep stratospheric intrusion event over Europe. *Atmospheric*
- 705 *Chemistry and Physics*, *3*(3), 763–777.
- Zanis, P., Hadjinicolaou, P., Pozzer, A., Tyrlis, E., Dafka, S., Mihalopoulos, N., & Lelieveld, J.
- 707 (2014). Summertime free-tropospheric ozone pool over the eastern Mediterranean/Middle East.
- 708 *Atmospheric Chemistry and Physics*, 14(1), 115–132.
- Zhao, K., Huang, J., Wu, Y., Yuan, Z., Wang, Y., Li, Y., et al. (2021). Impact of stratospheric
- intrusions on ozone enhancement in the lower troposphere and implication to air quality in Hong
- Kong and other South China regions. *Journal of Geophysical Research: Atmospheres*, 126(18),
- 712 e2020JD033955.



JGR Atmosphere

Supporting Information for

The Evolutions and Large-scale Mechanisms of Summer Stratospheric Ozone Intrusion across Global Hotspots

J. Lee¹, Y. Wu¹, and X. Wang²

¹Lamont-Doherty Earth Observatory of Columbia University, New York, NY

²National Center for Atmospheric Research, Boulder, CO

Contents of this file

Figures S1 to S3



Figure S1. The average days per season when 850 hPa O_3S exceeds 99% of NH O_3S each season. The 99% O_3S threshold across the entire NH for each season is written in parentheses. Red shadings are days per season, and gray shadings are masked topography. Regions where R-squared values between anomalous O_3 and O_3S below 0.5 are hatched.



Figure S2. Boxes are drawn on top of Fig. 1 for MCA and composite analysis. Black boxes show where MCA is conducted for each hotspot. They all have the same latitude range of $20^{\circ}-50^{\circ}$ N. The ranges of longitude for each hotspot are NA ($140^{\circ}-70^{\circ}$ W), Af (30° W- 0°), MD ($10^{\circ}-40^{\circ}$ E), and ME ($40^{\circ}-70^{\circ}$ E). Blue boxes show where stratospheric ozone intrusion speed has been estimated from box-averaged O₃S anomaly. The latitude range for NA is $20^{\circ}-80^{\circ}$ N, while others all have the same latitude range of $20^{\circ}-60^{\circ}$ N. The ranges of longitude for each hotspot are NA ($180^{\circ}-120^{\circ}$ W), Af (60° W- 0°), MD (15° W- 40° E), and ME ($15^{\circ}-70^{\circ}$ E).



Figure S3. The density histogram of the start date of extreme events for every global hotspot. The extreme events are defined following section 3.2 and calculated for the MCA's leading mode. Each bin is 10 days long except for the last bin, which is 12 days. Each hotspot's total number of events is written in the parenthesis next to the hotspot name.