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Quantifying the importance of stratospheric-tropospheric transport on surface ozone concentrations at high- and low-elevation monitoring sites in the United States

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HIGHLIGHTS

- The study quantifies the frequency of STT with enhanced surface O_3.
- Trajectories were used to identify coincidences between enhanced surface O_3 and STT.
- Many US sites exhibit frequent coincidences between enhanced surface O_3 and STT.
- STT plays an important role in affecting background O_3 concentrations across the US.

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ABSTRACT

In this study, we quantify the frequency of stratosphere-troposphere exchange (STE) events that result in ozone (O_3) concentration enhancements (i.e., hourly average concentrations $\geq$50 ppb) observed at 39 high- and low-elevation monitoring sites in the US during the years 2007–2009. We employ a refined forward trajectory-based approach to address the relationship between stratospheric intrusions and enhancements in hourly average O_3 concentrations. The model is applied to high-resolution European Center for Medium-Range Weather Forecasting (ECMWF) analyses to identify specific days when the potential for stratosphere-to-troposphere transport (STT) exists to affect surface O_3 levels. Our results indicate that STT down to the surface (STT-S) frequently contributes to enhanced surface O_3 hourly averaged concentrations at sites across the US, with substantial year-to-year variability. The O_3 concentrations associated with the STT-S events appear to be large enough to enhance the measured O_3 concentrations during specific months of the year. Months with a statistically significant coincidence between enhanced O_3 concentrations and STT-S occur most frequently at the high-elevation sites in the Intermountain West, as well as at the high-elevation sites in the West and East. These sites exhibit a preference for coincidences during the springtime and in some cases, the summer, fall, and late winter. Besides the high-elevation monitoring sites, low-elevation monitoring sites across the entire US experience enhanced O_3 concentrations coincident with STT-S events.

1. Introduction

Background ozone (O_3) is defined differently by various researchers, and several activities have attempted to distinguish between the terms baseline and background (McDonald-Buller et al., 2011). The Task Force on Hemispheric Transport of Air Pollution (2010) defined baseline concentrations as "…an observation made at a site when it is not influenced by recent, locally

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emitted or produced pollution." Zhang et al. (2011) estimated natural background by setting methane emissions to pre-industrial levels and eliminating anthropogenic emissions worldwide. Natural background sources of O_3 are associated with: (1) transport from the stratosphere; and (2) chemical production associated with lightning, the biosphere, and open fires (Zhang et al., 2011).

Recent research results corroborate that the contribution of stratosphere–troposphere exchange (STE) plays an important role in affecting natural background O_3 concentrations at high- and low-elevation monitoring sites across the US (Ambrose et al., 2011; Cooper et al., 2011; Lefohn et al., 2011; Emery et al., 2012; Langford et al., 2012). The relevance of STE for affecting low-tropospheric O_3 concentrations has been well documented (e.g., Reed, 1955; Junge, 1962; Danielsen, 1968; Danielsen, 1974; Danielsen and Mohnen, 1977; Ludwig et al., 1977; Shapiro, 1980; Haagenson et al., 1981; Davies and Schuepbach, 1994; Lamarque and Hess, 1994; Schuepbach et al., 1999; Stohl et al., 2000; Lefohn et al., 2001; Cooper et al., 2005; Cristofanelli et al., 2006; Hocking et al., 2007; Ordóñez et al., 2007; Langford et al., 2009; Akriditis et al., 2010; Cristofanelli et al., 2010). Estimations of the contribution from STE on surface O_3 concentrations in the US were described by Ludwig et al. (1977). Examining the behavior of radioactive debris injected into the stratosphere during nuclear weapons testing in the 1960s, they estimated that the greatest contribution to surface O_3 concentration would be along the eastern slope of the Rocky Mountains, Upper Midwest, and in a band stretching from Texas into the northeastern US. Buzzi et al. (1984), Davies and Schuepbach (1994), Stohl et al. (2000), and Gerasopoulos et al. (2001) have commented on the importance of STE affecting O_3 concentrations at high-elevation sites in close proximity to the leeward side of mountain ranges.

Ambrose et al. (2011) investigated the importance of STE versus long-range transport affecting lower tropospheric O_3 concentrations at the Mt. Bachelor Observatory (2763 m) in central Oregon. They reported that enhanced O_3 levels in the free troposphere were transported to the high-elevation site and were driven mostly by subsidence of O_3-rich air masses from the upper troposphere/lower stratosphere (~52%), Asian long-range transport (~13%), and a combination of Asian long-range transport and upper troposphere/lower stratosphere influences (~36%). Cooper et al. (2011) investigated the contribution to baseline O_3 concentrations in California over a 6-week period in May/June 2010. The authors indicated that descending stratospheric intrusions and Asian pollution plumes had an important influence on the O_3 concentration distributions along the California coast. Over the 6-week investigation, the greatest enhancements to surface O_3 from baseline sources occurred over the Los Angeles Basin (32–63%). Langford et al. (2012) found that surface O_3 measurements from 41 sampling stations indicated that ~13% of the variance in the maximum daily 8-h average (MDA8) O_3 between May 10 and June 19, 2010 was associated with the passage of upper-level troughs.

Lefohn et al. (2001) attributed STE processes to the observation that hourly average O_3 concentrations ≥50 ppb occurred frequently during the photochemically quiescent months in the winter and spring at several rural sites across southern Canada and the northern US. Lefohn et al. (2011), using trajectory calculations, investigated the frequency of STE events and their associated enhancements on 12 surface O_3 monitoring sites in the western and northern tier of the US. For most of the sites analyzed, they indicated that contributions from stratosphere-to-troposphere transport to the surface (STT-S) were frequent during specific months and appeared to enhance the surface O_3 concentrations at both high- and low-elevation monitoring sites.

This study expands the analysis of Lefohn et al. (2011) by investigating the frequency of STT-S events in enhancing hourly averaged O_3 concentrations ≥50 ppb at 39 urban and rural high- and low-elevation O_3 monitoring sites located in the West, Intermountain West, Midwest, and Eastern US for 2007–2009. In contrast to the study by Lefohn et al. (2011), the investigation here also includes sites that are heavily influenced by anthropogenic emissions. The seasonal and spatial aspects of the importance of STT-S are described and conclusions reached concerning the frequency of STT processes affecting enhanced surface O_3 concentrations across the US.

2. Approach

The selection of sites, the identification of stratospheric intrusions, and the statistical methodology used to relate enhanced O_3 concentrations with STT-S events are discussed in detail.

2.1. The selection of surface ozone monitoring sites for the study

Hourly averaged O_3 concentration data were downloaded for 37 monitoring sites across the US for 2007, 2008, and 2009 from the US EPA Air Quality System (AQS) and Clean Air Status and Trends Network (CASTNet) databases. Data from Trinidad Head, a NOAA experimental research site on California's north coast, and Mount Washington in New Hampshire, an Atmospheric Investigation, Regional Modeling, Analysis and Prediction (AIRMAP) research site (http://airmap.unh.edu/about/), were included with the AQS and CASTNet data. Tables 1 and 2 summarize the characteristics for the 16 high-elevation (>1.3 km) and the 23 low-elevation monitoring sites used in the analysis, respectively. We preferred to select monitoring sites that collected data over a 12-month period during each year. However, 5 of the 23 EPA AQS database sites collected data for less than 12 months (see Table 2) because of the EPA-defined O_3 season. For all other sites, data were collected over a 12-month period during each year. The mean across all 39 sites for the percentage of hourly average concentrations reported for 2007, 2008, and 2009 was 95%, 96%, and 96%, respectively. Ninety-seven percent of all site-months included in the study experienced at least 27 days within each month when a 1-h average concentration was recorded. Although infrequent, there were days in which all hourly average concentrations were missing.

2.2. Identification of stratospheric intrusions

As in Lefohn et al. (2011), a Lagrangian method, based on the approach introduced by Wernli and Bourqui (2002), is used to identify STT events down to the surface (i.e., STT-S events). The trajectory model introduced by Wernli and Davies (1997) is used to identify days of high probability for STT trajectories to enhance surface O_3 at specific monitoring sites. The model provides information along trajectories with a temporal resolution of 6 h. As input data, the model uses 6-hourly meteorological fields from the operational European Center for Medium-Range Weather Forecasting (ECMWF) analyses. They have a spectral truncation of T799, corresponding to a horizontal resolution of about 40 km and 91 vertical layers. The analysis fields (i.e., temperature and three wind components) were interpolated onto a regular latitude-longitude grid with a resolution of 0.6° and used to calculate potential temperature and potential vorticity (PV) that were used in the trajectory calculation.

The STT events were identified with the methodology described by Lefohn et al. (2011) and detailed in the Supplement Appendix. The 2-pvu isosurface of PV is used to define the tropopause (1 PV unit (pvu) corresponds to 10^{-6} K kg^{-1} m^2 s^{-1}). It is assumed that STT trajectories are originally associated with high stratospheric O_3 values and that a high percentage of the enhanced O_3

Table 1
Characteristics of the high-elevation (>1.3 km) O_3 monitoring sites used in the study.

Site name	Site ID	LU[a]	Latitude	Longitude	Elevation (m)	Months of data
West						
Lassen Volcanic NP, California	060893003	RF	40°32′ N	121°34′ W	1788	January–December
Yosemite NP (Turtleback Dome), CA	060430003	RF	37°42′ N	119°42′ W	1605	January–December
Crestline, California	060710005	RR	34°15′ N	117°16′ W	1384	January–December
Intermountain West						
Yellowstone NP, Wyoming	560391011	RF	44°34′ N	110°24′ W	2468	January–December
Pinedale, Wyoming	PND165	RF	42°55′ N	109°47′ W	2388	January–December
Centennial, Wyoming	CNT169	RF	41°22′ N	106°14′ W	3178	January–December
Gothic, Colorado	GTH161	R	38°57′ N	106°59′ W	2926	January–December
Rocky Mountain NP, Colorado	080690007	RF	40°17′ N	105°33′ W	2743	January–December
Chiricahua NM, Arizona	CHA467	R	32°01′ N	109°23′ W	1570	January–December
Grand Canyon NP, Arizona	GRC474	RF	36°03′ N	112°11′ W	2073	January–December
Great Basin NP, Nevada	GRB411	RF	39°00′ N	114°13′ W	2060	January–December
Canyonlands NP, Utah	CAN407	RD	38°27′ N	109°49′ W	1809	January–December
Jefferson County, Colorado	080590006	RI	39°54′ N	105°11′ W	1802	January–December
Mesa Verde NP, Colorado	080830101	RF	37°12′ N	108°29′ W	2165	January–December
East						
Mount Washington, New Hampshire	AIRMAP	RF	44°16′ N	71°18′ W	1917	January–December
Whiteface Mountain, New York	360310002	RF	44°22′ N	73°54′ W	1483	January–December

NP, National Park; NM, National Monument.
[a] LU, land use; R, rural; RA, rural agricultural; RD, rural desert; RF, rural forest; RI, rural industrial; RR, rural residential; SR, suburban residential; UI, urban industrial; UR, urban residential.

concentrations is transferred to the low troposphere over the US during the descent along the 10-day STT trajectory without significant mixing with the environment. The 10-day time period is motivated by the studies of Bithell et al. (2000) and Bourqui and Trépanier (2010), who reported elevated O_3 values in mid-tropospheric air parcels 10 days after they descended from the stratosphere. As in Lefohn et al. (2011), the turbulent processes that transport O_3 from the top of the boundary layer to the surface are not explicitly investigated. Instead, the vertical gradient of potential temperature, $\Delta\theta$, between the surface and the altitude of the stratospheric intrusion, is analyzed and regarded as a measure for the static stability of the atmosphere in the layer between the surface and the arrival height of the stratospheric intrusion. Small values of the potential temperature difference (i.e., $\Delta\theta < 5$ K) indicate that it is likely that the stratospheric air, which contains high levels of O_3, is mixed down to the surface due to boundary

Table 2
Characteristics of the low-elevation O_3 monitoring sites used in the study.

Site name	Site ID	LU[a]	Latitude	Longitude	Elevation (m)	Months of data
West						
Cheeka Peak, Washington	530090013	RF	48°17′ N	124°37′ W	466	January–December
King County, Washington	530330080	UR	47°34′ N	122°18′ W	105	January–December
Mount Rainier NP, Washington	530531010	RF	46°45′ N	122°07′ W	415	January–December
Trinidad Head, California	NOAA Site	R	41°06′ N	124°06′ W	107	January–December
El Dorado County, California	060170010	SR	38°43′ N	120°49′ W	585	January–December
Intermountain West						
Glacier National Park, Montana	300298001	RA	48°30′ N	113°59′ W	968	January–December
Big Bend NP, Texas	BBE401	RF	29°18′ N	103°10′ W	1052	January–December
Midwest						
Theodore Roosevelt National Park, ND	380070002	RA	46°53′ N	103°22′ W	850	January–December
Voyageurs National Park, Minnesota	271370034	RF	48°24′ N	92°49′ W	429	January–December
Ann Arbor, Michigan	ANA115	RF	42°25′ N	83°54′ W	267	January–December
Cook County, Illinois	170310001	SR	41°40′ N	87°44′ W	188	April–October
Stockton, Illinois	STK138	RA	42°17′ N	90°00′ W	274	January–December
Alhambra, Illinois	ALH157	RA	38°52′ N	89°37′ W	164	January–December
Harris County, Texas	482010066	UI	29°43′ N	95°30′ W	21	January–December
East						
Georgia Station, Georgia	GAS153	RA	33°10′ N	84°24′ W	270	January–December
Rockdale, Georgia	132470001	RA	33°35′ N	84°04′ W	219	March–October
Cuyahoga County, Ohio	390355002	SR	41°32′ N	81°27′ W	328	April–October
Bucks County, Pennsylvania	420170012	SR	40°06′ N	74°53′ W	12	April–October
Shenandoah National Park, Virginia	511130003	RF	38°31′ N	78°26′ W	1073	January–December
Blackwater NWR, Maryland	BWR139	RF	38°26′ N	76°06′ W	4	January–December
Abington, Connecticut	ABT147	RA	41°50′ N	72°00′ W	209	January–December
Fairfield County, Connecticut	090013007	SR	41°09′ N	73°06′ W	3	April–September
Chittenden County, Vermont	500070007	RF	44°31′ N	72°52′ W	392	January–December

NP, National Park; NWR, National Wildlife Refuge.
[a] LU, land use; R, rural; RA, rural agricultural; RD, rural desert; RF, rural forest; RI, rural industrial; RR, rural residential; SR, suburban residential; UI, urban industrial; UR, urban residential.

layer turbulence. In contrast, a larger value of $\Delta\theta$ makes it unlikely that the stratospheric air mass will affect surface O_3 concentrations. The total number of STT events with $\Delta\theta < 5$ K (i.e., the most likely STT events affecting the surface O_3 concentrations) is referred to in our study as STT-S (stratosphere-to-troposphere transport to surface). STT-S is regarded as a combined long-distance laminar transport typically along isentropes from the stratosphere to the low troposphere (that can be assessed with trajectory calculations) and a comparatively small-scale mainly vertical turbulent transport in the atmospheric boundary layer (that is not represented by the trajectories and is coarsely estimated in our study with the aid of the parameter $\Delta\theta$).

In this study, we applied a slight refinement of the methodology described in Lefohn et al. (2011). Once the trajectory comes close to the surface (i.e., $\Delta\theta < 5$ K), we only regard the subsequent 18 h as potentially relevant for increasing surface O_3 concentrations. The rationale behind this criterion is that an originally stratospheric air mass likely loses its stratospheric O_3 signature within less than one day due to turbulent mixing in the boundary layer. We postulated that 18 h is an appropriate length of time because the boundary layer mixing is strongly driven by the diurnal cycle of solar radiation. The rationale for modifying our previous methodology is summarized in Fig. 1. The top panels illustrate a horizontal view of the trajectories as they approach the measurement site, with red coloring indicating a potential temperature difference between the trajectory and the surface of less than 5 K. Along these trajectory segments, turbulent boundary layer mixing is likely to occur, leading to a dilution of the high stratospheric O_3 values. The idealized temporal evolution of the O_3 concentration along these trajectories is shown in the bottom panels. For the STT-S events (left) associated with a boundary layer residence time of 18 h or less (referred to as "direct"), the turbulent dilution begins when the trajectory reaches the surface site (i.e., green dot). It is anticipated that the O_3 concentration is still elevated. After 18 h, we refer to the STT-S events as "indirect" (right). For these events, one option is that due to the ongoing turbulent mixing the trajectory reaches the site with low (i.e., no longer stratospheric) O_3 values (blue line). Another option to consider is that trajectories for which $\Delta\theta < 5$ K is fulfilled during more than 18 h may travel in the boundary layer over fairly large distances (e.g., up to 1000 km) before reaching the site. It may be possible that they mix with O_3 concentrations associated with anthropogenic emissions. In this case (orange line), the trajectory reaches the site with elevated O_3 values that are associated with anthropogenic sources. In contrast, we assume that if the "direct" STT-S trajectories (left) cross a region with anthropogenic emissions, they will be unaffected by the emissions because the trajectories are still separated from the surface by a strong vertical potential temperature difference (i.e., $\Delta\theta > 5$ K). For the statistical evaluations in this study, only the direct STT-S events have been considered, whereas Lefohn et al. (2011) considered all STT-S events without separating direct and indirect events.

2.3. Statistical treatment

For each 24-h period for each of the 39 monitoring sites, the number of STT-S events was calculated using the procedure described in Section 2.2. As described in Lefohn et al. (2011), the STT-S events information was related to the maximum hourly averaged O_3 concentration that occurred during the same 24-h period. In our analysis, N50 refers to the number of days during a particular month when the daily maximum hourly average O_3 concentrations were $\geq$50 ppb, which is above the level that the EPA estimates for background (US EPA, 2006).

A 2 $\times$ 2 coincidence table was constructed that summarized the 24-h values by month. The columns are labeled STT-S = 0 and STT-S > 0. The rows were labeled Max O_3 < 50 ppb (referred to N50 = 0) and Max O_3 $\geq$50 ppb (referred to as N50 $\geq$ 1). The value in the top left box (referred to as element "a") in the table represents the number of days during the month when both N50 and STT-S were equal to 0. The value in the bottom right box represents the number of days during the month when both N50 and STT-S were greater than 0 (referred to as element "d"). The values in the top left box and bottom right box are referred to as the on-diagonal elements. The value in the top right box represents the number of days during the month when N50 = 0 and STT-S $\geq$ 0 (referred to as element "b"). The magnitude of this element represents the number of days within the month when air parcels associated with STT-S events did not contain sufficiently enhanced O_3 levels to provide enhancement to the surface O_3 concentrations. The value in the bottom left box represents the number of days during the month when N50 $\geq$ 0

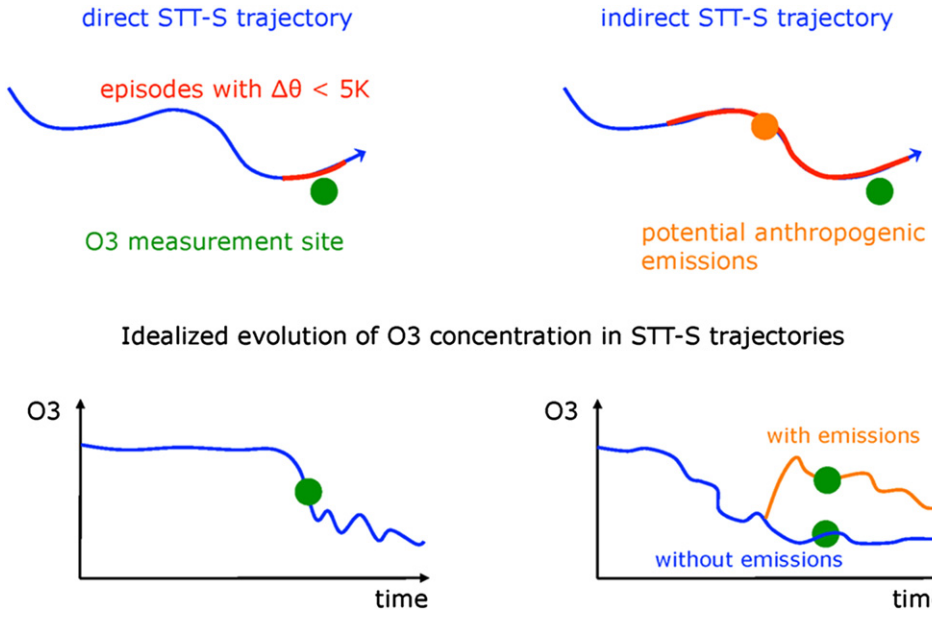

Fig. 1. Schematic illustration of "direct" and "indirect" STT-S events. See Section 2.2 for explanation.

and STT-S = 0 (referred to as element "c"). The magnitude of this element reflects the number of days within the month when surface O_3 concentration enhancements occurred and the enhancements were not associated with STT-S events but rather with anthropogenic emissions, long-range transport outside of North America, lightning, or wildfires. Elements "b" and "c" are referred to as the off-diagonal elements. The on-diagonal elements provide an indication of the strength of the coincidental relationship between the daily occurrences during the month of the N50 and STT-S values. For each month for each site, we summed the on-diagonal elements and divided this sum by the number of days with data within the month. The resulting value we refer to as the coincidence value, which ranges from 0 (i.e., no relationship) to 1 (i.e., strong relationship).

As described in Lefohn et al. (2011), we applied a level of 0.75 to indicate when a coincidence value indicated that particular events conform to the hypothesis that there is a relationship between N50 and STT-S values. As described in Lefohn et al. (2011), if the coincidence value was equal to or greater than the lower limit, it was considered to be statistically significant. In our analysis, we form coincidence tables and then calculate coincidence values and lower limit values for each of the 39 monitoring sites by month and determine the statistical significance for the monthly coincidence value. We do not include in our analysis a month in which the "d" element is 0 and the month is significant (based on the sum of the "a" and "d" elements). Table 3 provides an example of the application of the methodology. The description of the use of the data contained within Table 3 is discussed in the Supplement Appendix.

Unlike our previous analysis (Lefohn et al., 2011), our current study includes some sites that are heavily influenced by anthropogenic emissions. Although the focus of our analysis is on identifying sites that experience coincidences between STT-S and enhanced O_3 concentrations, we believe it is important to also identify to what extent days with zero STT-S counts are contributing to the enhanced O_3 concentrations. We refer to this type of enhancement as "non-STT-S" events. Element "c" in each of the monthly coincidence tables that exhibited statistically significant coincidence values between STT-S and enhanced surface O_3 concentrations was used to determine the percentage of days when non-STT-S events were related to enhanced O_3 concentrations. For these tables, we identified the number of days in which the "c" element was greater than 3 (i.e., greater than 10% of the days). Using this information for each site, we determined over the 3-year period the number of months with a statistically significant coincidence value that also experienced a "c" element greater than 3. As an indicator for assessing the importance of non-STT-S events at a specific site, we then calculated the percentage of months with a statistically significant coincidence value, which experienced greater than 10% of the days when O_3 concentration enhancements were related to non-STT-S events. Sites that experienced fewer than 3 months with a statistically significant coincidence value were not summarized in this part of the analysis.

3. Results

3.1. Comparison of results applying previous and modified methodology

We compared the results using the methodology described in Lefohn et al. (2011) (i.e., considering all STT-S events) with the results using the methodology applied in this study (considering direct STT-S events only). Table 4 shows the comparison for six O_3 monitoring sites (Cheeka Peak, Trinidad Head, Glacier NP, Yellowstone NP, Voyageurs NP, and Chittenden County) used in both studies. While differences were observed for some of the monitoring sites, the comparison of methodologies shows that the differences do not change the conclusion reached in Lefohn et al. (2011) that STT processes appear to be related to enhanced O_3 concentrations at the six monitoring sites. No difference was found for Trinidad Head, Glacier NP, and Chittenden County. For Yellowstone NP, two of the 16 months (i.e., July 2007 and September 2007) were eliminated when the modified methodology was implemented. When the modified methodology was implemented for Voyageurs NP, the months of April 2007 and September 2007 were added, while for Cheeka Peak, the months of April 2008 and September 2008 were eliminated and June 2009 and September 2009 were added. We assume that the modified methodology is more reliable, in particular for stations close to potential anthropogenic emissions.

3.2. Seasonal and spatial patterns of enhanced O_3 concentrations associated with STT-S occurrences

Fig. 2 summarizes the number of months accumulated (over the 3-year period) by season in which the monitoring sites exhibited

Table 3
Examples of coincidence tables relating number of days of STT-S = 0 or STT-S > 0 with number of days when daily maximum hourly average O_3 concentration was either <50 ppb or ≥50 ppb for Yellowstone NP (WY), Rocky mountain NP (CO), and Georgia station (GA) for March–June 2007. Coincidence value is calculated by summing the on-diagonal elements and dividing by the number of measurement days in the month.

	March		April		May		June	
	STT-S = 0	STT-S > 0	STT-S = 0	STT-S > 0	STT-S = 0	STT-S > 0	STT-S = 0	STT-S > 0
Yellowstone NP								
Max O_3 < 50 ppb	2	6	0	0	1	0	0	6
Max O_3 ≥ 50 ppb	2	21[b]	3	27[b]	0	27[b]	3	21[b]
Coincidence value	0.74[a]		0.90[a]		1.00[a]		0.70[a]	
Rocky Mountain NP								
Max O_3 < 50 ppb	2	4	1	4	0	0	0	0
Max O_3 ≥ 50 ppb	1	23[b]	2	23[b]	8	23[b]	10	20[b]
Coincidence value	0.83[a]		0.80[a]		0.74[a]		0.67[a]	
Georgia Station								
Max O_3 < 50 ppb	0	3	3	3	0	2	3	0
Max O_3 ≥ 50 ppb	6	22[b]	1	23[b]	9	20[b]	8	19[b]
Coincidence value	0.71[a]		0.87[a]		0.65[a]		0.73[a]	

[a] Coincidence value statistically significant. See Section 2.3 for explanation.
[b] STT contribution to enhanced O_3 concentration considered to be important when coincidence value was statistically significant.

Table 4
Comparison of months in which sites exhibited a statistically significant coincidence value and there was a coincidence between the number of days with daily maximum hourly average O_3 concentrations ≥50 ppb and STT-S > 0 using the original methodology described in Lefohn et al. (2011) and current study.

Site	Months (original methodology)	Months (modified methodology)
Cheeka Peak, WA	May 2007	May 2007
	August 2007	August 2007
	April 2008	–
	September 2008	–
	–	June 2009
	–	September 2009
Trinidad Head, CA	June 2008	June 2008
Glacier NP, MT	April 2008	April 2008
	April 2009	April 2009
Yellowstone NP, WY	March 2007	March 2007
	April 2007	April 2007
	May 2007	May 2007
	June 2007	June 2007
	July 2007	–
	August 2007	August 2007
	September 2007	–
	March 2008[a]	March 2008[a]
	April 2008	April 2008
	May 2008	May 2008
	June 2008	June 2008
	July 2008	July 2008
	March 2009	March 2009
	April 2009	April 2009
	May 2009	May 2009
	June 2009	June 2009
Voyageurs NP, MN	–	April 2007
	–	September 2007
	April 2008	April 2008
	April 2009	April 2009
	May 2009	May 2009
Chittenden County, VT	April 2008	April 2008
	May 2008	May 2008
	April 2009	April 2009
	May 2009	May 2009

[a] Data capture less than 90% but statistically significant coincidences existed.

a statistically significant value for the coincidence between the number of days with daily maximum hourly average O_3 concentrations ≥50 ppb and direct STT-S > 0. Monitoring sites that are located at high-elevations (i.e., ≥1.3 km) are illustrated in bold. The average number of days per month when STT-S is coincident with O_3 enhancements by geographic region is shown in Fig. 3. The numbers within each bar are the number of site-months associated with the average. The information in Tables 1 and 2 provides the list of sites in each geographic region in Fig. 3. The figure indicates the frequency of coincidences within each geographic region. Supplemental Tables S-1–S-6 provide a detailed description of the number of days in which the daily maximum hourly average O_3 concentration was ≥50 ppb and coincident with a direct STT-S event for each of the statistically significant STT-S months. They also provide an indication of the variability of the coincidences at each site across years.

The Intermountain West experienced some of the greatest number of months and days when direct STT-S was coincident with hourly averaged enhanced O_3 concentrations (i.e., ≥50 ppb) (Fig. 2). For the high-elevation site at Yellowstone NP, numerous days occurred during the spring and summer months when O_3 concentration enhancements were statistically coincident with STT-S. During the late winter, spring, summer, and early fall, the Centennial, Gothic, Rocky Mountain NP, and Pinedale sites exhibited frequent days within a specific month in which O_3 concentration enhancements were statistically coincident with STT-S. The sites at Mesa Verde NP, Grand Canyon NP, Chiricahua NM, Great Basin NP, Canyonlands NP, and Jefferson County (CO) experienced statistically significant coincidences between enhanced O_3 concentrations and STT-S during the springtime, as well as other seasons. In 2009, both Canyonlands NP (November) and Rocky Mountain NP (November and December) experienced late fall and early winter months in which the enhanced O_3 concentrations were statistically coincident with STT-S. In contrast to the high-elevation sites, the low-elevation site Glacier NP (MT) in the Intermountain West, experienced only two months (April 2008 and April 2009) in which O_3 concentration enhancements were statistically coincident with direct STT-S events.

In the West, the two high-elevation sites, Lassen Volcanic NP and Yosemite NP, similar to the sites in the Intermountain West, exhibited numerous days during the spring, summer, and fall months when statistically significant coincidences occurred between STT-S and hourly averaged enhanced O_3 concentrations. At the high-elevation Crestline site, numerous days were observed during the spring and fall when STT-S and hourly averaged enhanced O_3 concentrations were coincident. The low-elevation site at El Dorado, near Sacramento (CA), appeared to be influenced frequently during the spring and fall months by STT-S events. In contrast, the low-elevation sites, Cheeka Peak, King County, Mount Rainier, and Trinidad Head, experienced infrequent numbers of days and months when O_3 concentration enhancements were coincident with STT-S.

In the Midwest, the low-elevation sites at Theodore Roosevelt NP (ND) and Voyageurs NP (MN), along the Northern Tier of the US, experienced frequent O_3 concentration enhancements for the spring months associated with STT-S; the sites at Ann Arbor (MI) and Cook County (MI) experienced statistically significant coincidences during the spring and summer months. The Harris County (TX) site, near Houston, experienced coincidences between O_3 concentration enhancements and STT-S during the spring, summer, and fall months. However, the number of enhancement days within each of the statistically significant STT-S months was less than at many of the other sites in the Midwest.

In the East, the two stations located in Georgia (Rockdale and Georgia Station) experienced most of their O_3 concentration enhancements coincident with STT-S during the statistically significant STT-S spring months and in September. The sites at Cuyahoga County (OH) and Bucks County (PA) experienced most of their O_3 concentration enhancements coincident with STT-S events during the spring and occasionally during the summer. At the Shenandoah NP (VA) site, O_3 concentration enhancements were coincident with STT-S during the spring, summer, and fall months. Infrequent O_3 concentration enhancements coincident with STT-S occurred at Blackwater NWR (MD), Abington (CT), and Fairfield (CT). The high-elevation sites at Mount Washington (NH) (spring, summer, and fall) and Whiteface Mountain (NY) (spring and fall) experienced frequent enhancements coincident with STT-S events; the low-elevation site at Chittenden County (VT) frequently experienced O_3 concentration enhancements coincident with STT-S events during April and May.

3.3. Characterizing the frequency of coincidences and the relationship of daily maximum concentrations with STT-S events

Fig. 3 summarizes the average number of days per month by geographic region for those months in which a statistical significant coincidence value occurred between the STT-S events and the O_3 concentration enhancements. For the statistically significant months identified for each site, the average number of days per month is calculated by averaging the "d" element for each of the monthly coincidence tables described in Section 2.3. The figure illustrates that the average number of days per month for the

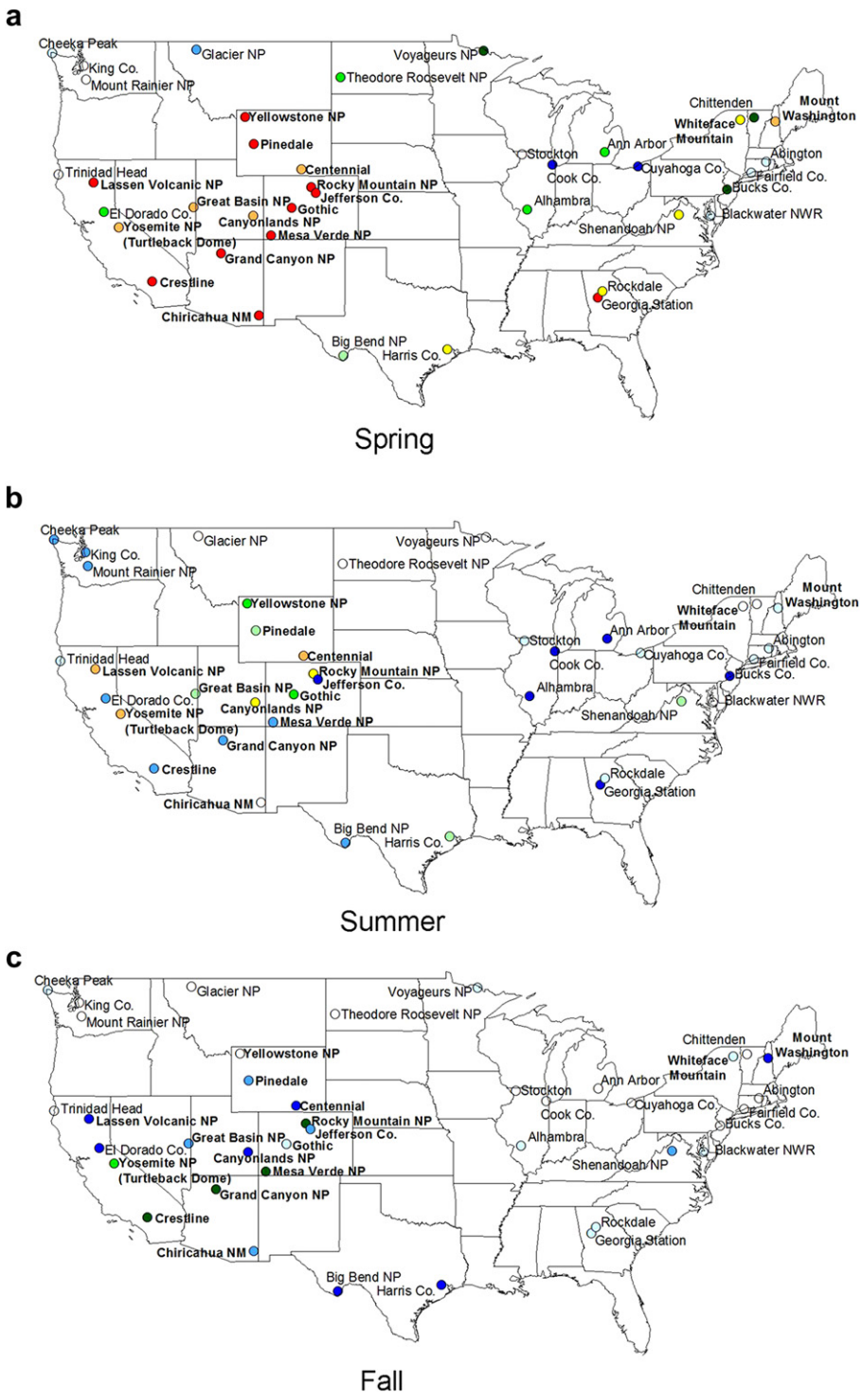

Fig. 2. The number of months during the (a) spring (MAM), (b) summer (JJA), (c) fall (SON), and (d) winter (DJF) seasons for the period 2007–2009 when a statistically significant coincidence value occurred with a coincidence between STT-S events greater than 0 and daily maximum hourly average O_3 concentrations $\geq$50 ppb. High-elevation sites are identified in bold lettering.

coincidences is greatest at the high-elevation West and Intermountain West sites. Table 5 summarizes by site over the 3-year period the (1) average number of days when coincidences occurred and (2) number of statistically significant coincident months. All of the high-elevation sites in the West and Intermountain West experienced a monthly average of 23 or 24 days of coincidence between STT-S events and enhanced O_3 concentrations. This region of the US experienced the highest number of months when statistically significant coincidences occurred (i.e., 13–22 months). The two high-elevation sites in the East (Mount Washington and Whiteface Mountain) exhibited monthly averages of 19–20 days when coincidences occurred, respectively. There were 12 and 8 statistically significant coincident months experienced, respectively. In the Midwest and East, the low-elevation sites generally experienced fewer statistically significant coincident months than the high-elevation sites across the US. The

Fig. 2. (*continued*).

exceptions to this pattern were the sites in Harris County, Georgia Station (GA), and Shenandoah NP (VA).

Besides the tendency for the high-elevation sites in the West, Intermountain West, and East to experience more frequent coincidences than the low-elevation sites across the US, the high-elevation sites experienced more days with concentrations ≥50 ppb and ≥60 ppb on days when STT-S > 0 for statistically significant months in which STT-S and enhanced O_3 concentrations were coincident. For example, over the spring period for those days in which STT-S > 0, the high-elevation Yellowstone NP site experienced approximately 86% and 72% of the total days with 1-h and 8-h daily maximum concentrations ≥50 ppb, respectively. In comparison, the low-elevation sites experienced lower percentages than the higher-elevation locations when STT-S > 0. As an example, Voyageurs NP experienced approximately 66% and 44% of the days with 1-h and 8-h daily maximum concentrations ≥50 ppb, respectively. We found that there was no distinction between the high- and low-elevation sites for the 1-h and 8-h daily maximum concentrations ≥70 ppb when STT-S > 0. Maximum concentrations, which all occurred in spring, showed little dependence on altitude.

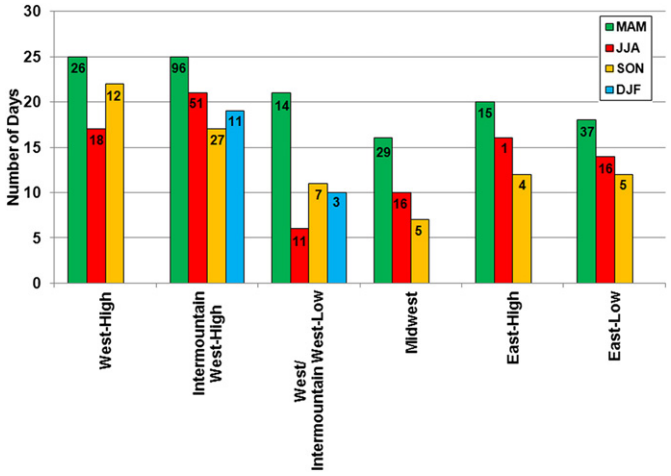

Fig. 3. Average number of days per month when STT-S is coincident with O_3 enhancements by geographic region for spring (March, April, and May), summer (June, July, and August), fall (September, October, and November), and winter (December, January, and February). The numbers within each bar are the number of site-months contributing to the average.

3.4. Ozone concentration enhancements coincident with days with zero STT-S counts

Fig. 4 illustrates the relative ranking for those sites affected by days with zero STT-S counts coinciding with enhanced O_3 days during the months in which there occurred a statistically significant coincidence value between STT-S events and O_3 enhancements. The relative ranking reflects the importance of the "c" element in the coincidence table described in Section 2.3. Fig. 4 summarizes by site the percentage of months that experienced a statistically significant coincidence value and exhibited more than 3 days when O_3 concentration enhancements were related to non-STT-S events. At Voyageurs NP (MN), Chittenden County (VT), King County (WA), Cuyahoga County (OH), Yellowstone NP (WY), and Lassen Volcanic NP (CA) this occurred during 10% or less of all months with a statistically significant coincidence value. Georgia Station (GA), Harris County (TX), Whiteface Mountain (NY), Shenandoah NP (VA), Crestline (CA), Canyonlands NP (UT), Jefferson County (CO), Bucks County (PA), Alhambra (IL), Mesa Verde NP (CO), El Dorado County (CA), and Yosemite NP (CA) appeared to be most affected by days with zero STT-S counts. These sites are located in areas of the US that are exposed to local or regional anthropogenic emissions.

4. Discussion

This section discusses the seasonal and spatial patterns, the frequency of coincidence patterns, and compares the results with those from previous observations. In discussing the importance of STT-S processes and their influence on US background O_3 concentrations, we identify some of the caveats associated with the applied methodology.

4.1. Seasonal and spatial patterns and the relative importance of STT versus long-range transport of Asian emissions

We found that the greatest contribution to surface O_3 concentration from STT-S processes occurred at the high-elevation sites in

Table 5
Summary of average number of days per month over the 3-year period (2007–2009) for those months in which a statistically significant coincidence value occurred. The number of months with statistically significant coincidence values is listed.

Site	Region	Avg. days per month	Num. of months
Lassen Volcanic NP, CA	West – High	24	20
Yosemite NP (Turtleback Dome), CA	West – High	24	21
Crestline, CA	West – High	23	15
Yellowstone NP, WY	Intermountain West – High	23	14
Pinedale, WY	Intermountain West – High	23	18
Centennial, WY	Intermountain West – High	23	20
Gothic, CO	Intermountain West – High	23	17
Rocky Mountain NP, CO	Intermountain West – High	23	22
Chiricahua NM, AZ	Intermountain West – High	23	13
Grand Canyon NP, AZ	Intermountain West – High	23	17
Great Basin NP, NV	Intermountain West – High	24	16
Canyonlands NP, UT	Intermountain West – High	23	19
Jefferson County, CO	Intermountain West – High	22	14
Mesa Verde NP, CO	Intermountain West – High	23	15
Cheeka Peak, WA	West/Intermountain West – Low	3	4
King County, WA	West/Intermountain West – Low	1	3
Mount Rainier NP, WA	West/Intermountain West – Low	7	2
Trinidad Head, CA	West/Intermountain West – Low	2	1
El Dorado County, CA	West/Intermountain West – Low	20	10
Glacier National Park, MT	West/Intermountain West – Low	18	2
Big Bend NP, TX	West/Intermountain West – Low	16	13
Theodore Roosevelt NP, ND	Midwest	18	5
Voyageurs NP, MN	Midwest	15	5
Ann Arbor, MI	Midwest	19	8
Cook County, IL	Midwest	15	6
Stockton, IL	Midwest	1	1
Alhambra, IL	Midwest	17	9
Harris County, TX	Midwest	7	16
Mount Washington, NH	East – High	19	12
Whiteface Mountain, NY	East – High	20	8
Georgia Station, GA	East – Low	17	13
Rockdale, GA	East – Low	17	9
Cuyahoga County, OH	East – Low	18	4
Bucks County, PA	East – Low	15	7
Shenandoah NP, VA	East – Low	18	15
Blackwater NWR, MD	East – Low	10	2
Abington, CT	East – Low	14	2
Fairfield County, CT	East – Low	13	2
Chittenden County, VT	East – Low	18	4

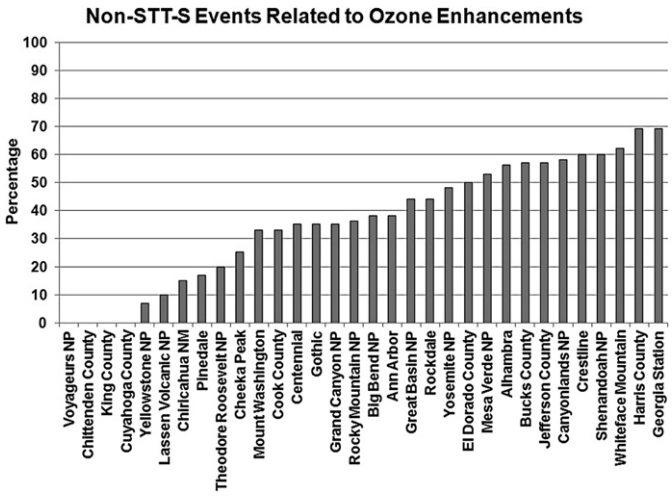

Fig. 4. The percentage of months over the 3-year period (2007–2009) that experienced a statistically significant coincidence value and exhibited greater than 10% of the days (i.e., 3 days) when O_3 concentration enhancements were related to non-STT-S events (lightning, wildfires, and anthropogenic sources). Sites that experienced fewer than three statistically significant months were not summarized.

the West and Intermountain West during the spring, summer, and fall. The Upper Midwest and a band stretching from Texas into the northeastern US also exhibited enhancements associated with STT-S. For the Upper Midwest, frequent occurrences were exhibited when O_3 concentration enhancements were coincident with STT-S events. Similar to Ludwig et al. (1977), we observed that some sites in the Midwest experienced statistically significant STT-S months while other sites in the region experienced few enhancements associated with STT-S. Some of the sites in the Midwest and South received substantially less O_3 concentration enhancement from STT-S than observed in the Intermountain West. Ludwig et al. (1977) noted that STT-S enhancements occurred frequently in the Northeast. Our results indicated that the high-elevation sites in the East, as well as the low-elevation site in Chittenden County (VT), exhibited numerous days when enhanced O_3 concentrations were coincident with STT-S.

At times, enhancement of hourly average O_3 concentrations associated with STT events are combined with enhancements associated with long-range transport of Asian emissions. Several studies (Bertschi et al., 2004; Bertschi and Jaffe, 2005; Oltmans et al., 2010; Pfister et al., 2010) have documented the presence of Eurasian biomass burning effluent and its impact on O_3 levels in western North America, both in the spring and summer. There is evidence that descending stratospheric intrusions and Asian pollution plumes influence O_3 concentrations in the West and Intermountain West (Oltmans et al., 2010; Cooper et al., 2011; Ambrose et al., 2011; Lefohn et al., 2011). Lin et al. (2012) reported that a mixture of long-range transport from Asian emissions and STT contributes to high-O_3 episodes over the high-elevation sites in the western US. Ambrose et al. (2011) provide evidence that the contribution of STT on surface O_3 concentrations in the western US may be greater than from Asian emissions. Lin et al. (2012) indicate that to the east of the Intermountain West, the contribution of long-range transport of Asian emissions may be less important.

4.2. The frequency of STT-S events enhancing O_3 concentrations in rural and urban areas of the US

Our results indicate that the O_3 concentrations associated with STT-S frequently enhance the measured O_3 concentrations during specific months of the year, with the result that natural background O_3 concentrations appear to be affected by STT-S events not just in the West and Intermountain West, but also in other regions across the US. These findings complement the results reported by Lefohn et al. (2011), who noted that STT events were of sufficient magnitude and frequency to enhance O_3 concentrations at the high-elevation site at Yellowstone NP, and low-elevation sites in the West and along the northern tier of the US. Although the frequency of the coincidences between STT-S and enhanced O_3 concentrations

may be larger in the West and Intermountain West than other parts of the US (Table 5), the magnitude and frequency of the STT-S events appear to be sufficient to affect the level of natural background O_3 concentrations across the entire US.

Sites influenced by anthropogenic emissions, such as Jefferson County, El Dorado County, Crestline, Harris County, Shenandoah NP, and Georgia Station, while frequently experiencing coincidences between STT-S events and enhanced O_3 concentrations, also experienced non-STT-S related O_3 concentration enhancements (Fig. 4). One site of interest is Crestline, which is located in southern California and is heavily influenced by anthropogenic sources. Langford et al. (2012) investigated the stratospheric influence on surface O_3 in the Los Angeles area during late spring and early summer of 2010 and showed that at times STT processes appear to enhance surface O_3 concentrations. Our results agree with those of Langford et al. (2012) that the Crestline site, while heavily influenced by anthropogenic emissions, is also influenced by STT-S.

4.3. Caveats of the methodology

This study, like Lefohn et al. (2011), is based fundamentally upon the calculation of large samples of trajectories using gridded wind fields from global reanalyses. Clearly, these calculations are affected by various error sources associated with the quality of the reanalysis wind fields (observational errors, data assimilation issues), their spatial and temporal resolution (which does not capture mesoscale features on scales smaller than about 100 km and shorter-lived than 6 h), and with errors in the trajectory computation. Unfortunately, these errors cannot be quantified and are likely to vary strongly from case to case. We address this uncertainty by choosing a statistical approach; the fairly large set of events and a thorough statistical test provide robustness to our results.

The O_3 concentration in stratospheric intrusions is highly variable due to concentration differences in the stratospheric origin and in chemical and mixing processes during their descent. The motivation for using our statistical methodology was the inability to characterize statistical associations between the enhanced hourly average concentrations $\geq$50 ppb and the number of stratospheric intrusions. We believe we have reduced the opportunity for STT-S events to lose their stratospheric O_3 signature due to turbulent mixing in the boundary layer prior to arriving at the site by restricting the trajectories' near-surface residence time to 18 h or less. The comparison of the methodology described in Lefohn et al. (2011), which did not apply the additional 18-h residence time criterion, with the methodology applied in this study found only minor differences, which did not impact the conclusion reached in Lefohn et al. (2011) that STT processes were statistically related to enhanced O_3. However in this study, which investigated the potential influence of STT at less remote sites, it was useful to include this additional criterion to distinguish between direct and indirect STT-S events.

In our analysis, we observed other months, beyond those months with a statistically significant coincidence between O_3 enhancements and STT-S reported in the tables and figures, when STT-S appeared to contribute to enhanced O_3 concentrations. By applying the stringent coincidence value described in Section 2.3, we established a statistical approach that provided us with a degree of certainty regarding the coincidences between STT-S and enhanced O_3 concentrations. It is therefore possible that our methodology may result in an underestimate of the role that STE plays in affecting enhanced surface O_3 concentrations. Applying alternative statistical methodologies in the future (as done by Bourqui et al., 2012) may provide refined estimates of the contribution of STT-S to enhanced O_3 concentrations.

5. Conclusion

This study expands the conclusions of Lefohn et al. (2011) that STT plays an important role in affecting natural background O_3 concentrations at both high- and low-elevation monitoring sites in the US. Thirty-nine O_3 monitoring sites across the US at rural and urban locations were assessed for STT processes affecting surface O_3 concentrations for the period 2007–2009 (Table 5). A forward trajectory-based approach was used to address the relationship between stratospheric intrusions and enhancements in hourly average O_3 concentrations. All of the monitoring sites exhibited months in which a statistically significant coincidence occurred between the number of days with a daily maximum hourly average O_3 concentration $\geq$50 ppb and direct stratospheric intrusion that reached the surface (i.e., STT-S $>$ 0).

Our results, similar to those reported by other investigators describing empirical and modeling results, indicate that STT events contribute to enhanced surface O_3 hourly averaged concentrations ($\geq$50 ppb) at sites across the US in varying degrees. The coincidences occur most frequently at the high-elevation sites in the Intermountain West, as well as at the high-elevation sites in the West and East, with a preference for springtime and in some cases summer, fall, and late winter. Besides the high-elevation monitoring sites, low-elevation monitoring sites across the US also experience enhanced O_3 concentrations that are coincident with STT-S.

For those sites that exhibited months in which a statistically significant coincidence exists between STT-S events and enhanced O_3 concentrations, Voyageurs NP (MN), Cuyahoga County (OH), Chittenden County (VT), Yellowstone NP (WY), and Lassen Volcanic NP (CA) appear to be least affected by days that experienced days with zero STT-S counts coinciding with enhanced O_3 days. In contrast, sites such as Jefferson County (CO), El Dorado County (CA), Mesa Verde NP (CO), Harris County (TX), Canyonlands NP (UT), Crestline (CA), Shenandoah NP (VA), and Georgia Station (GA) appear to be most influenced by days with zero STT-S counts coinciding with enhanced O_3 days.

Our results and those published by others support the observation that STT-S events appear to frequently enhance the measured O_3 concentrations, with the result that natural background O_3 concentrations are affected by STT-S events in the West, Intermountain West, and other regions across the US during specific seasons of the year, with variation occurring across years.

Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.atmosenv.2012.09.004.

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