EXPLANATIONS FOR THE SPRING PEAK IN GROUND-LEVEL OZONE IN THE SOUTHWESTERN UNITED STATES

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Abstract: Many remote locations in the Northern Hemisphere have a spring peak, rather than a summer peak, in ground-level ozone concentrations, and the principal cause is presumed to be stratosphere-troposphere exchange (i.e., stratospheric intrusions). Grand Canyon National Park (GCNP) in northern Arizona also has a spring peak, and the purpose of this study is to explore the impact of stratospheric intrusions and another process synoptic-scale pollutant transport on ground-level ozone levels at GCNP from 1996 to 2000. The primary methods involve the stratification of days to identify stratospheric-intrusion days and the compositing of days to assess the impact of pollutant transport on ground-level ozone concentrations. Results indicate that stratospheric intrusions contributed little to the ozone budget at GCNP. In fact, atmospheric pollution originating in southern California was the likely cause of the May peak in ozone. The transported pollution also appeared to be responsible for high ozone days during all spring months. Tracer-based research (i.e., beryllium-7 and methylchloroform) at multiple locales in the southwestern United States is needed to fully confirm the weak impact of stratospheric intrusions and the strong contribution of ozone and its precursors originating in southern California. [Key words: ozone, stratospheric intrusions, pollutant transport, southwestern United States.]

INTRODUCTION

Ground-level ozone at a given locale is a function of locally produced ozone and ozone that has been transported to the locale. Ozone is an atmospheric pollutant, thus it is an important entity for two reasons: it has been proven rigorously to impact forests negatively (Woodman, 1987; Cowling, 1989) and it may exacerbate respiratory ailments of humans (e.g., Friedman et al., 2001). Tropospheric ozone is produced typically by the oxidation of volatile organic compounds (VOCs) in the presence of nitrogen oxides (NO_x) and sunlight (Chameides et al., 1992). The transport of ozone and its precursor chemicals (i.e., VOCs and NO_x) can occur over a range of spatial scales. Moreover, the transport can occur at all levels of the troposphere, and it can involve exchanges between the stratosphere and the troposphere.

A spring maximum, rather than a summer maximum, characterizes the annual ozone cycle in remote regions in the Northern Hemisphere (Davies and Schuepbach, 1994). This cycle reflects the pre-industrial ozone cycle, which was presumably controlled by ozone exchange from the stratosphere to the troposphere (Stohl et al., 2000). In the absence of human activities and thus increased emissions of VOCs and NO_x, little *in situ* ozone production should occur (Lefohn et al., 1990; Lefohn et al., 2001).

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Locales in the interior portion of the western United States also have the pre-industrial ozone cycle (Diem, 2003); however, the exact cause is unknown. Diem (2003) hypothesized that the spring peak (i.e., peak in May) in the southwestern United States, especially at rural locales, results from stratosphere-troposphere ozone exchange, synoptic-scale pollutant transport, or both. Using the south rim of Grand Canyon National Park (GCNP) as the study location, this paper provides an exploration of those speculations.

It is vitally important to identify the processes responsible for periods of elevated ozone levels, because knowledge of the relative impacts of those processes may influence air-quality policy decisions. Lefohn et al. (2001) noted that the occurrence of episodic, naturally occurring events might increase ozone levels to a point where reductions of anthropogenic pollutant emissions may not prevent exceedances of the National Ambient Air Quality Standard (NAAQS) for ozone. The same general linkage can be applied to events associated with the long-distance, horizontal transport of pollutants. Although these transport events are certainly not "natural," they are not impacted directly by emissions-control measures introduced in the locale where the ozone exceedance may occur.

STUDY REGION

The GCNP site, which is located 2,152 m a.s.l. on the south rim of the Grand Canyon (35.95°N, -112.15°W), was chosen because it contains the only rural, high-elevation ozone monitor in the southwestern United States that is relatively distant from large urban areas (Fig. 1). Several other high-elevation monitors exist in central Arizona, but those monitors receive pollutants regularly from the Phoenix area via a mountain-valley circulation (Ellis et al., 2000). Because stratospheric air may not penetrate to heights lower than ~1600 m a.s.l. (Reiter, 1991), high-elevation monitors are needed to observe stratosphere-derived ozone.

RATIONALE FOR POLLUTANT TRANSPORT TO THE SOUTHWEST

Since GCNP is a rural locale distant from large NO_x sources, the two largest contributors of ozone at GCNP may be stratosphere-troposphere exchange and synoptic-scale pollutant transport. This section explores from a theoretical stance the respective potential of those processes for causing the spring peak in ground-level ozone at GCNP.

Stratosphere-Troposphere Ozone Exchange

Because stratosphere-troposphere ozone exchange is a significant process for the budget of ozone in the free troposphere (Bonasoni et al., 2000), it is possible that the exchange may be prevalent at a high-elevation site such as GCNP. Troughs and cut-off low-pressure cells (i.e., upper-level lows) in the upper troposphere are the most important causes of these stratospheric intrusions (Stohl et al., 2000). The degree of exchange from the stratosphere to the troposphere is proportional to the

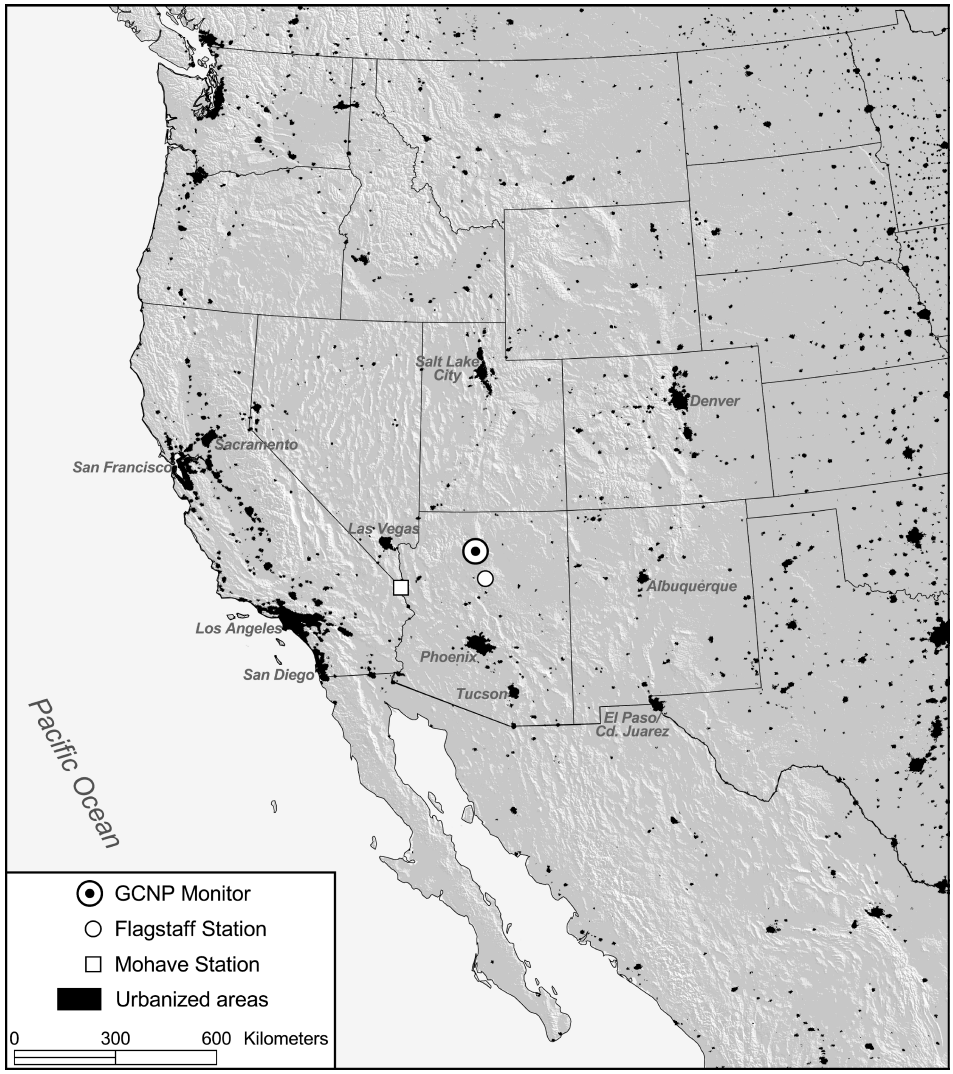

Fig. 1. Western United States showing urban areas, topography, and the locations of the GCNP ozone monitor and several meteorological stations. Urban areas were determined using satellite-measured nighttime light-intensity data available for the period 1996–1997 from the National Geophysical Data Center.

strength of the upper-level low-pressure system (Johnson and Viezee, 1981), and typical intrusion events can cause elevated ground-level ozone concentrations to persist for several hours to several days (Davies and Schuepbach, 1994). Danielson's (1980) conceptual model linked trough-related intrusions to tropopause folding, which occurs on the western side of a trough. The folding leads to the subsidence of ozone-rich air to a surface anticyclone. Downward transport also

can occur through convective mixing in cut-off lows (Davies and Schuepbach, 1994). Lastly, stratospheric air can undergo substantial horizontal transport before reaching the surface (Bonasoni et al., 2000), which results in additional mixing with tropospheric air.

Stratospheric intrusions have seasonal and spatial variations, with a spring maximum of stratospheric-tracer concentrations in the troposphere in middle latitudes (Appenzeller et al., 1996). The spring maximum can be attributed primarily to the accumulation of ozone in the stratosphere over winter at high latitudes coupled with the horizontal movement of that ozone-rich air to lower latitudes through increased meridional flow (Davies and Schuepbach, 1994). Consequently, a relatively large reservoir of stratospheric ozone exists just above the tropopause in the middle latitudes in spring; in fact, there is a two-fold decline from winter/spring to summer in the ozone content of the lower stratosphere (Singh, 1980). Synoptic-scale systems, which are prevalent in spring, can initiate the transport of the stratospheric ozone to the low levels of the troposphere: for example, Gettelman and Sobel (2000) noted that mid-latitude storm-track regions have the highest values of cross-tropopause ozone flux.

There is considerable troughing—which includes cut-off lows—over the southwestern United States in winter and spring, thus it is plausible that stratospheric intrusions should add to the ozone budget at GCNP and other high-elevation sites in the region. Burnett (1994) found that southwestern troughs during the 1946–1987 period were most frequent in March and April, and troughing occurred on more than 25% of the days from March through May. For the 1979–1988 period, a mid-tropospheric troughing pattern over the Southwest was most frequent in May and least frequent during July and August (Davis and Walker, 1992; Davis and Gay, 1993a). Also supporting the notion that stratospheric intrusions peak in spring, observations made near Denver, Colorado indicated that stratosphere-troposphere ozone transfer peaks in late spring in that region (Langford, 1999).

Synoptic-Scale Pollutant Transport

There also is evidence that ozone in the southwestern United States may have its origins in distant urban areas. In fact, trans-Pacific transport of pollutants has been documented along the western coast of North America (Jaffe et al., 2003); therefore, Asia may be a significant source of the atmospheric pollution in the Southwest. Nevertheless, nearly all researchers (Ashbaugh, 1983; Malm et al., 1990; White et al., 1990; Pryor and Hoffer, 1992; Davis and Gay, 1993a, 1993b; Johnson et al., 1994; White et al., 1994; de P. Vasconccelos et al., 1996; Pryor et al., 1995; Henmi and Bresch, 1998) who have examined particulate transport to GCNP and nearby locales have found southern California to be the most likely source region (refer to Fig. 1). In fact, a majority of the days during any given year will have air entering the Grand Canyon area from the southwest (White et al., 1994; Henmi and Bresch, 1998).

The formal techniques that have been employed to examine pollutant transport include back-trajectories, tracer studies, and synoptic typing. White et al. (1990) implicated the Los Angeles basin as the source of most above-background

methylchloroform (i.e., "tracer of opportunity") observed in the summer at sites west of GCNP, for the weekly methylchloroform cycles at those sites lagged similar cycles observed in the Los Angeles basin by one to two days. Similarly, Pryor and Hoffer (1992) observed a synoptically driven transport episode of methylchloroform from Los Angeles to the western portion of the Grand Canyon throughout a summer night. Ashbaugh (1983) determined that the prevailing pathway for air trajectories terminating at GCNP was over the deserts of southern California. Back-trajectories also indicated that southern California was the dominant source region of fine sulfur for the Grand Canyon (Malm et al., 1990). It was concluded that visibility at GCNP was associated with the direction of the large-scale flow: hazy air tended to come from the southwest (White et al., 1994). Results from back-trajectories for the 1984–1989 period linked high particulate concentrations at the Grand Canyon with air arriving from the southwest (de P. Vasconcelos et al., 1996). Finally, Davis and Gay (1993b) determined that the highest standardized particulate concentrations at GCNP for the 1984–1988 period coincided with 800-mb circulation patterns having southwesterly flow.

Pollutant transport from southern California to GCNP should be maximized in spring and minimized in summer. Synoptic-scale circulation systems with southwesterly flow are most prevalent in April, May, and June (Davis and Walker, 1992). Davis and Gay (1993a) found that for the 1979–1988 period, a southwesterly flow synoptic pattern associated with high particulate concentrations at GCNP was most frequent during the transition months of April and November and nearly absent from June through September. Likewise, Pryor et al. (1995) found the influence of synoptic-scale conditions on methylchloroform concentrations at locations just west of the Grand Canyon to be weaker during the summer season than during the winter season. Lastly, Johnson et al. (1994) concluded that high concentrations of particulate nitrate in spring at a high-elevation site located just west of Las Vegas, Nevada was due to nitrate transport from the Los Angeles basin.

PURPOSE AND OBJECTIVES

Several major deficiencies in the peer-reviewed literature provide the impetus for the research presented in this paper. Those research "gaps" are as follows: no research has been conducted on stratospheric intrusions in the southwestern United States; and, despite the presence of multiple pollutant-transport studies for the Grand Canyon area, the transport of ozone and its precursors (i.e., VOCs and NO_x) to GCNP has never been examined. As discussed in the previous section, both stratospheric intrusions and synoptic-scale transport may influence ground-level ozone concentrations at GCNP. Only several studies (Wooldridge et al., 1997; Lefohn et al., 2001) have noted the occurrence of stratospheric intrusions in the interior portion of the western United States and their impact on ground-level ozone concentrations. Although Diem (2003) did discuss the conceptual basis for the transport of ozone from metropolitan areas (e.g., Las Vegas, Los Angeles, and Phoenix) to GCNP, an empirical study was never conducted.

The purpose of this study is to determine the likely causes of the spring peak in ground-level ozone in the southwestern United States. Therefore, the key objectives

are as follows: (1) identify stratospheric-intrusion days; (2) estimate the ozone contribution from the identified stratospheric intrusions; and (3) examine the potential of nonintrusion days for synoptic-scale transport of pollutants within the troposphere to GCNP.

DATA

The core data used in this study consisted of ground-level ozone concentrations, near-surface meteorological values, and upper-level meteorological values corresponding to the 1996–2000 period (Fig. 1). Hourly ozone concentrations for the GCNP monitor (U.S. Environmental Protection Agency [EPA] number 040058001442011) were obtained from the EPA. Daily near-surface measurements of temperature, dew-point temperature, sea-level pressure, and wind speed were acquired from the National Climatic Data Center (NCDC) for a nearby weather station (WMO identification number 72378). Daily solar-radiation totals were obtained from the Arizona Meteorological Network (AZMET) for the Mohave station (34.97°N, 114.6°W). Daily radiosonde measurements of geopotential height, temperature, dew-point temperature, wind speed, and wind direction were obtained for Flagstaff (35.23°N, -111.82°W; 2,179 m a.s.l.) from the Forecast Systems Laboratory of the National Oceanic and Atmospheric Administration (NOAA; WMO Flagstaff Airport identification number 72376). Daily geopotential-height surfaces were extracted from the National Center for Environmental Protection/National Center for Atmospheric Research (NCEP/NCAR) Reanalysis dataset (Kalnay et al., 1996), which was provided by NOAA's Climate Diagnostics Center. Estimates of backward trajectories from the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model were provided by NOAA's Air Resources Laboratory.

METHODS

Identification and Examination of Stratospheric-Intrusion Days

Stratospheric-intrusion days were identified using a three-stage procedure. The following criteria were tested: (1) presence of a 500-mb circulation pattern conducive to stratosphere-troposphere exchange; (2) presence of a relatively high hourly ozone concentration; and (3) presence of air-parcel descent over a period of several hours to several days. Days that fulfilled the first criterion, first and second criteria, and all three criteria were classified as potential intrusion days, probable intrusion days, and highly probable intrusion days, respectively.

Daily 500-mb geopotential-height surfaces were examined in order to extract days with the highest potential for stratospheric intrusions. Days when GCNP was west of a trough, under a trough, or under an upper-level low were considered potential intrusion days. Therefore, the days were placed into the following categories: west of trough, under trough, under upper-level low, and other. Days in the "other" category were not considered potential intrusion days.

Days with relatively high ozone concentrations were flagged through the joint analysis of hourly ozone concentrations and daily near-surface atmospheric conditions. Irrespective of the influences of stratospheric intrusions and synoptic-scale pollutant transport, it was assumed that days having similar near-surface atmospheric conditions should have similar ozone concentrations. Therefore, air-mass typing (e.g., Kalkstein et al., 1990) was used to place days at GCNP into various air-mass classes. The air-mass typing was performed for all days for the following groups of months: March–April, May–June, July–August, September–October, and November–February. The procedure involved a P-mode principal components analysis (PCA) using a correlation matrix followed by a clustering of days using the score of each day on the extracted, unrotated components. Eigenvalues and scree plots were assessed to determine the number of components to extract. The variables used in the PCA were mean temperature, daily temperature range, mean relative humidity, mean sea-level pressure, and mean wind speed. The clustering was a two-stage process, with hierarchical clustering used initially to determine the appropriate number of clusters and nonhierarchical clustering (i.e., k-means) used subsequently to place the days into the final air-mass types. Hourly ozone concentrations were converted into Z-scores for each hour/air-mass type combination. In this study, for a given potential intrusion day, if one of its Z-scores was greater than one (i.e., greater than one standard deviation above the mean) then that day was retained as a probable intrusion day.

The third and final criterion, air-parcel descent, was tested by examining multi-day backward trajectories from the HYSPLIT model. The HYSPLIT model was configured in the following manner: NCEP/NCAR Reanalysis data and the meteorological model's vertical velocity fields were used; and -112.15ºW and 35.95ºN were the starting longitude and starting latitude, respectively. Probable intrusion days having downward trajectories from at least the middle troposphere were retained as highly probable intrusion days. Specific details of each trajectory were not considered, because the accuracies of the trajectories were unknown. Therefore, it was assumed that the trajectories were only accurate enough for determining the general vertical movement of air.

Estimates of the impact of the highly probable intrusion days on the ground-level ozone budget were made through comparisons with other days. The SUM0 index, which is the total of all hourly ozone concentrations, was calculated for each day. Since it was impossible to properly estimate what the SUM0 levels may have been had the intrusion not occurred, the impact of each highly probable intrusion event was its daily SUM0 value minus the smallest daily SUM0 value for the month of occurrence of the event. For example, if a highly probable intrusion day occurred in May, then the smallest daily SUM0 value in May from 1996 to 2000 was subtracted from the SUM0 value for the intrusion day. The overestimation procedure was intended to counteract underestimates resulting from the employment of daily data instead of hourly data.

For comparison purposes, the maximum impact of stratospheric intrusions was estimated by examining SUM0 levels for the potential intrusion days. By using daily data to estimate the contribution of stratospheric intrusions to the overall ozone budget it is possible that the quantity of ozone linked to stratospheric intrusions was

underestimated. For example, if the intrusion occurred over a period of 3 hrs. and the concentration during all 3 hrs. was 80 ppb and the other 21 hrs. had ozone concentrations of 45 ppb, then the daily SUM0 value was 1185 ppb. If the day to which the intrusion day was compared had an ozone concentration of 50 ppb for all 24 hrs., then the daily SUM0 value for that day was 1200 ppb. At the daily level, the intrusion was associated with a deficit of 15 ppb, while during the time when the intrusion actually occurred the surplus was 90 ppb. The impact of each potential intrusion event was its daily SUM0 value, for it was assumed that absolutely no ground-level ozone would exist on those days if stratospheric intrusions had not occurred. Although the resulting estimates of stratospheric contributions to the ground-level ozone budget are unrealistic, the values do represent the upper limit of stratospheric-intrusion ozone contributions.

Examination of Synoptic-Scale Pollutant Transport

The potential for synoptic-scale transport on nonintrusion days was investigated using several different compositing approaches. First, the characteristics of nonintrusion days during each ozone-season month (i.e., March through August) were compared. Second, an environment-to-circulation approach (e.g., Yarnal, 1993) was employed to assess differences in atmospheric conditions and circulation patterns between high ozone days (HODs) and the remainder of the nonintrusion days for each month.

Characteristics of the nonintrusion days were assessed using graphical and statistical approaches. Differences between months in near-surface atmospheric conditions, activity of local sources of pollutants (i.e., number of park visitors), and 700-mb circulation patterns were assessed graphically. The Mann-Whitney *U* test was used to determine statistically significant ($\alpha = .05$) differences in near-surface and upper-level atmospheric conditions between the months. This nonparametric test can be used with small samples and it does not require a normal distribution, thus it was much more appropriate for this study than were parametric tests (e.g., Student's *t*-test).

Regarding the environment-to-circulation approach, the SUM60 index, rather than the SUM0 index, was used to extract the month-specific nonintrusion HODs, which were then compared with the remainder of the nonintrusion days. SUM60 is the sum of all hourly ozone concentrations greater than or equal to 60 ppb, and high SUM60 values are indicative of days with anomalously high hourly ozone concentrations. The SUM0 index was not used, because SUM0 values can be increased substantially by decreased ozone deposition resulting from the presence of snow cover (Wooldridge et al., 1997). Nonintrusion days that had SUM60 values in the top 20% of all days in a given month were classified as HODs (e.g., Diem and Comrie, 2001); therefore, each month had at least 30 HODs. Statistically significant ($\alpha = .05$) differences in near-surface and upper-level atmospheric conditions between the two types of days were determined using the Mann-Whitney *U* test.

Table 1. Results of the Principal Components Analysis and the Clustering Analysis for the Groups of Months

Months	Number of extracted components[a]	Number of clusters (air-mass types)[b]
March–April	2 (80)	3 (136, 78, 81)
May–June	2 (77)	2 (92, 206)
July–August	2 (71)	2 (55, 233)
September–October	3 (87)	4 (33, 141, 94, 34)
November–February	2 (74)	3 (232, 131, 218)

[a]Percent of variance explained in parentheses.

[b]Number of days in each cluster in parentheses.

RESULTS AND DISCUSSION

Stratospheric Intrusions

There were a relatively small number of stratospheric-intrusion days from 1996 to 2000, and those days were not confined to the spring months. Of the initial 1827 days, only 415 days (~23% of the days) over the 5-yr. period were potential intrusion days (Fig. 2A). Combined results from the upper-level circulation analyses and the air-mass-based ozone analyses (results related to the air-mass typing are provided in Table 1) revealed that only 181 days were probable intrusion days (Fig. 2B). Only 62 of those days occurred in April, May, and June. The final criterion of substantial air-parcel descent produced the final set of 60 highly probable intrusion days (Fig. 2C). Only 19 of those days occurred in April and May; no highly probable events occurred in June.

Based on the troughing-based model of Danielson (1980), tropopause folding is assumed to be the main mechanism by which ozone is transferred from the stratosphere to the troposphere above GCNP. GCNP was located under the western limb of an upper-level trough (i.e., left-of-trough situation) on over 70% of the highly probable intrusion days, with most of the remaining days characterized by the presence of an upper-level low. Nearly all the days did not have near-surface atmospheric conditions (i.e., relatively warm, high solar radiation, dry, and calm conditions) conducive to enhanced *in situ* ozone production (Table 2). In addition, the synoptic-scale transport of pollutants from large urbanized areas to GCNP was unlikely for nearly all the intrusion days, for the air was arriving at GCNP mostly via descent from the northern sector. Wind directions at 700-mb and 500-mb also indicate that large urbanized areas in the western United States were rarely upwind of GCNP (Table 2).

The general characteristics of the 60 highly probable intrusion days can be gleaned through the visualization of 500-mb circulation patterns on two days: April 14, 1999, and April 1, 2000. The highest hourly ozone concentration (87 ppb) measured at GCNP from 1996 to 2000 occurred on April 14, 1999 (Fig. 3A). This day, like many other highly probable intrusion days, contained a deep trough over the interior portion of the Southwest that provided air-parcel descent from the

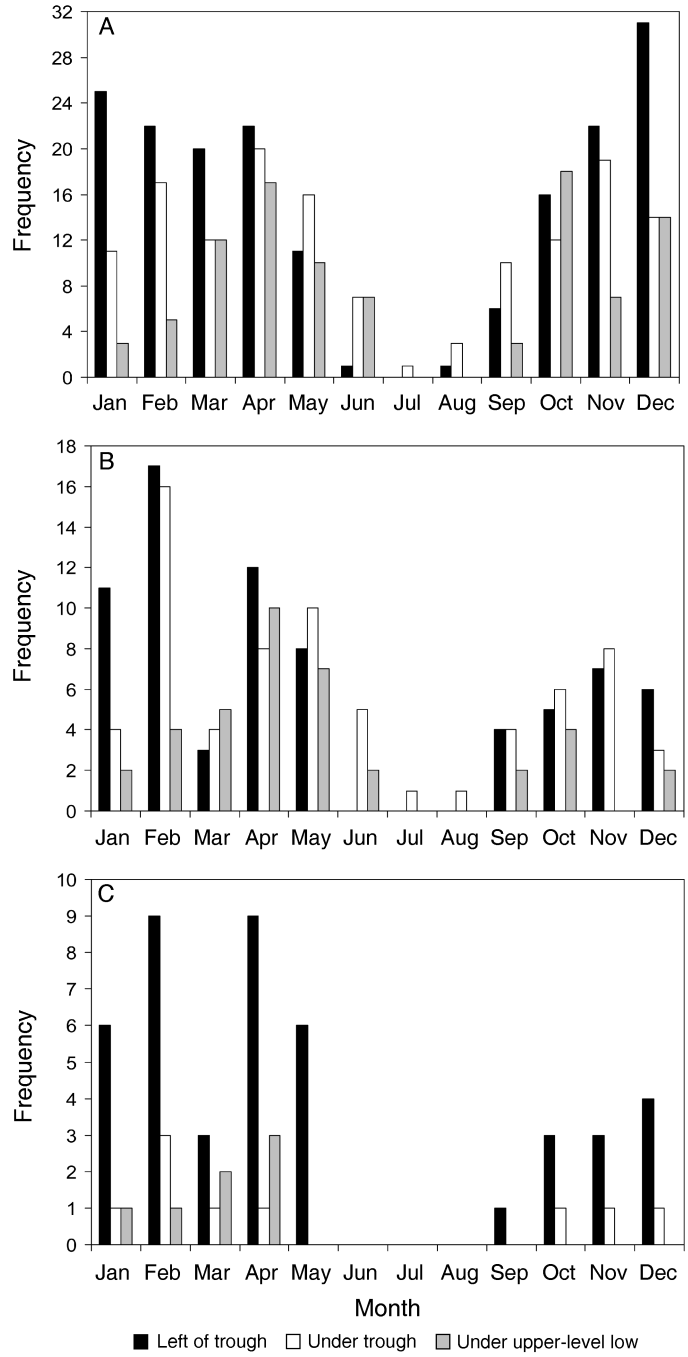

Fig. 2. Monthly variation in (A) potential intrusion days, (B) probable intrusion days, and (C) highly probable intrusion days.

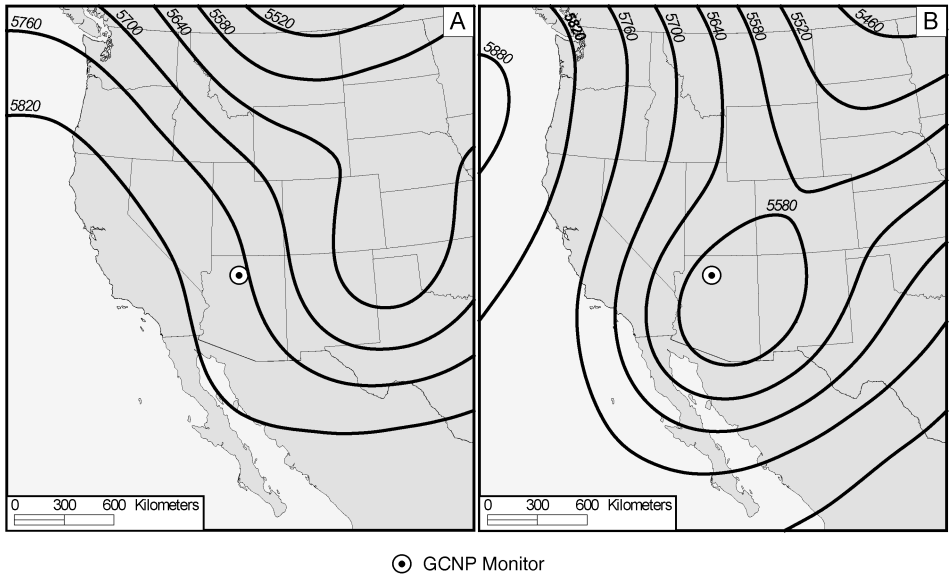

Fig. 3. Geopotential-height surface at 500-mb surface for (A) April 14, 1999, and (B) April 1, 2000. Contour interval is 60 m.

northwest to GCNP; however, unlike most highly probable intrusion days, local near-surface atmospheric conditions on April 14, 1999 may have been conducive to increased ozone concentrations. An upper-level low was present on April 1, 2000 (Fig. 3B), and this day did not have atmospheric conditions favorable for increased *in situ* ozone production.

Results indicate that stratospheric intrusions contributed little to the ground-level ozone budget at GCNP from 1996 to 2000, and stratospheric intrusions probably were not the cause of the May peak in ozone levels at GCNP during the 1996–2000 period (Fig. 4). It is unlikely that stratospheric intrusions were responsible for more than 2% of both the annual SUM0 and the May SUM0 at GCNP (Fig. 5). Even if all 415 potential intrusion days were considered and their daily impact were maximized, stratospheric intrusions still would have contributed no more than 30% of the annual SUM0. The 37 potential intrusion days in May would have been responsible for only 25% of the May SUM0. Consequently, either the synoptic-scale transport of pollutants or local factors or both caused the peak in ozone levels to occur in May.

Synoptic-Scale Pollutant Transport

Synoptic-scale pollutant transport from southern California was the most likely cause of the May peak in ozone levels at GCNP. Localized precursor emissions and ozone production probably were not major contributors to the peak, for emissions and production were maximized from June through August when near-surface temperatures were higher, solar radiation was more intense, and pollutant emissions

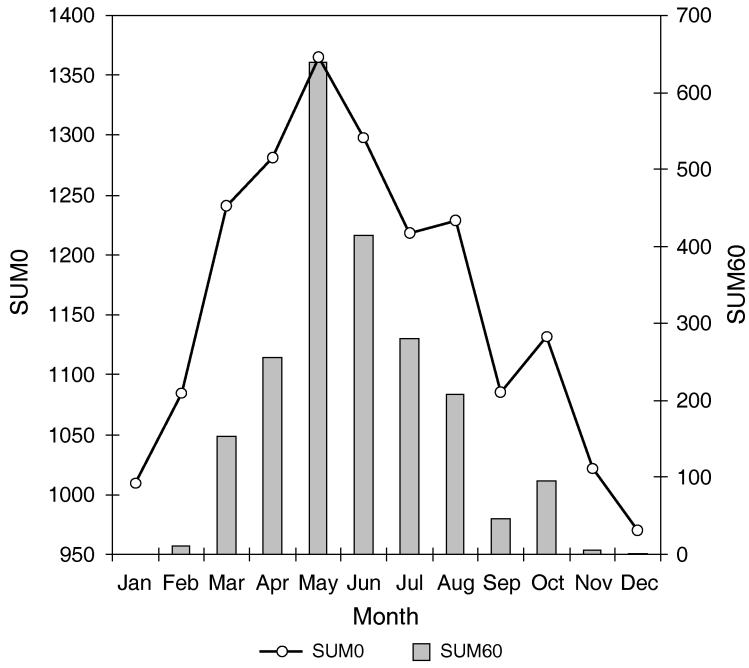

Fig. 4. Monthly variation in daily SUM0 levels and daily SUM60 levels at GCNP.

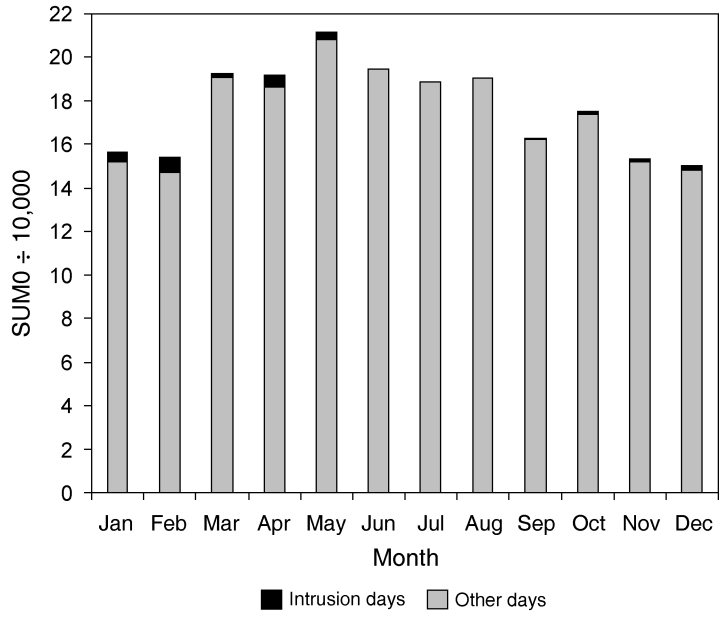

Fig. 5. Monthly variation in estimated contribution from stratospheric intrusions.

Table 2. Near-Surface Atmospheric Conditions at GCNP, Mid-Tropospheric Wind Directions at Flagstaff, and HYSPLIT-Produced Backward Air-Parcel Origins for the 60 Highly Probable Intrusion Days at GCNP[ab]

Day/period	TEMP	RANGE	RH	SLP	WS	WD700	WD500	HYOR
01/18/1996	-5.4	23.4	42	1,018	2.0	337	309	NW
01/30/1997	2.2	13.0	59	1,029	3.8	115	36	N
01/22/1998	-2.9	16.0	63	1,020	1.8	310	320	NW
01/02/1999	-1.9	22.2	45	1,023	1.6	350	320	N
01/03/1999	-3.4	21.0	48	1,025	1.6	10	320	N
01/09/1999	-3.3	24.0	48	1,029	1.2	15	335	N
01/22/1999	-0.4	18.3	37	1,020	2.4	355	335	NW
01/29/1999	-5.8	25.0	54	1,026	1.9	35	5	NE
JAN (mean)	-0.7	17.4	62	1,020	2.7	278	285	
02/23/1996	-2.2	25.6	53	1,018	2.7	10	317	NW
02/27/1996	-9.6	18.9	82	1,016	3.0	272	277	W
02/19/1997	1.2	21.0	37	1,021	2.3	*	8	N
02/26/1997	-4.2	19.0	62	1,011	2.1	316	328	N
02/12/1998	-3.9	21.6	63	1,023	1.4	*	*	N
02/28/1998	-9.1	25.0	48	1,025	1.4	355	325	NW
02/02/1999	-2.3	22.2	52	1,027	1.3	340	310	NW
02/11/1999	-8.3	15.0	45	1,030	4.5	355	320	N
02/16/1999	1.1	18.0	46	1,021	1.6	280	300	NW
02/22/1999	2.6	27.2	36	1,021	3.4	*	*	NW
02/27/1999	2.1	23.3	40	1,021	1.5	295	320	NW
02/01/2000	-0.9	18.0	56	1,026	2.9	15	345	NW
02/29/2000	0.2	18.4	62	1,020	2.1	320	300	NW
FEB (mean)	0.9	16.4	60	1,018	3.1	250	271	
03/15/1996	2.1	21.2	72	1,020	1.3	3	19	NE
03/16/1996	4.2	16.1	56	1,016	1.9	349	341	N
03/15/1998	4.9	15.0	78	1,014	2.9	40	50	NE
03/16/1998	4.5	19.4	60	1,011	1.6	345	315	N
03/13/1999	1.2	28.9	42	1,020	1.8	50	355	N
03/23/2000	1.4	25.5	63	1,020	2.4	340	340	E
MAR (mean)	3.5	20.5	50	1,016	3.1	260	276	
04/29/1996	7.7	24.4	17	1,025	4.3	13	337	N
04/17/1998	0.7	18.0	61	1,020	1.9	35	355	N
04/19/1998	4.6	23.9	40	1,021	2.0	330	305	N
04/28/1998	6.6	16.6	71	1,019	2.7	320	320	NE
04/29/1998	6.7	20.0	62	1,019	2.4	125	105	NE
04/30/1998	8.8	22.8	48	1,019	1.6	145	190	NE

(*table continues*)

Table 2. *Continued*

Day/period	TEMP	RANGE	RH	SLP	WS	WD700	WD500	HYOR
04/14/1999	8.3	22.8	32	1,018	2.3	355	340	NW
04/15/1999	4.7	25.0	31	1,021	3.8	25	320	N
04/16/1999	3.2	26.7	30	1,024	2.7	10	345	N
04/30/1999	0.4	13.3	83	1,013	2.7	230	235	W
04/01/2000	3.4	12.7	44	1,015	6.0	40	50	E
04/02/2000	2.3	23.3	39	1,018	1.8	30	25	N
04/30/2000	7.7	27.2	26	1,019	3.2	25	345	N
APR (mean)	6.1	20.3	45	1,015	3.3	246	270	
05/02/1999	5.9	20.0	61	1,013	3.5	280	295	N
05/04/1999	5.3	12.2	63	1,006	4.7	280	310	NW
05/05/1999	5.2	18.8	43	1,015	2.5	330	320	NW
05/06/1999	7.6	24.4	39	1,019	1.8	355	315	NW
05/17/1999	9.9	28.4	28	1,019	2.4	225	285	NW
05/01/2000	9.3	31.6	26	1,020	2.1	15	250	N
MAY (mean)	12.4	23.1	33	1,013	3.7	234	253	
09/27/1996	10.5	13.0	27	1,018	4.1	357	313	N
SEP (mean)	14.7	18.5	55	1,016	2.6	217	244	
10/12/1997	1.0	9.0	72	1,016	2.5	327	322	N
10/02/1998	9.0	24.4	52	1,015	2.7	295	300	NW
10/17/1998	1.8	20.0	61	1,021	3.2	65	360	N
10/17/1999	4.8	18.9	23	1,025	6.3	*	280	NE
OCT (mean)	7.9	20.4	49	1,018	2.6	223	264	
11/06/1996	1.6	13.0	57	1,016	1.2	*	*	N
11/21/1997	-2.5	19.4	63	1,021	1.4	318	305	NW
11/10/1998	-4.1	18.3	63	1,023	0.8	*	*	NW
11/25/1999	-4.1	28.9	31	1,031	0.9	350	340	NW
NOV (mean)	2.3	19.3	54	1,020	2.3	257	276\	
12/28/1997	-4.1	16.0	40	1,029	2.1	19	342	N
12/11/1998	-6.1	26.6	41	1,035	0.7	25	10	N
12/24/1998	*	*	*	*	*	40	15	NW
12/28/1998	-0.6	23.9	49	1,027	1.2	335	305	NW
12/02/1999	-1.3	23.0	42	1,018	2.6	15	335	NW
DEC (mean)	-1.3	17.7	56	1,023	2.8	295	294	

[a]Monthly mean conditions are provided for comparison purposes.

[b]TEMP = mean daily temperature (ºC); RANGE = mean daily temperature range (ºC); RH = mean daily relative humidity (%); SLP = mean daily sea-level pressure (mb); WS = mean daily wind speed (m s^{-1}); WD700 = 700-mb wind direction; WD500 = 500-mb wind direction; HYOR = origin of air parcels as determined by HYSPLIT model.

*Data were unavailable.

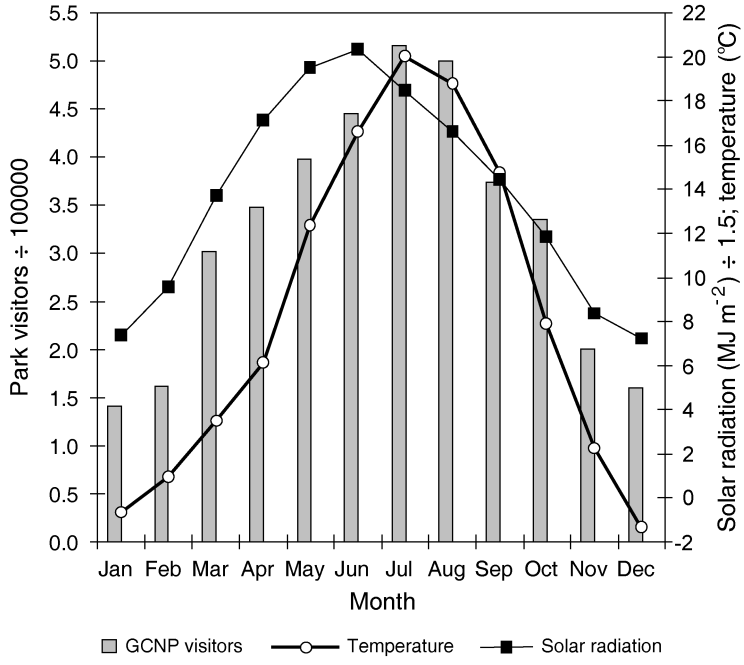

Fig. 6. Monthly variation in visitors to GCNP, temperature, and solar radiation. Park-visitor totals only include counts at the south and east gates of the park.

from local sources (e.g., motor vehicles of park visitors) was highest (Fig. 6). Monthly park-visitation data for 1996–1999 were provided by Grand Canyon National Park. Among all the ozone-season months, August had the most consistent near-surface atmospheric conditions which were generally conducive to enhanced *in situ* ozone production—among all the ozone-season months (Fig. 7). Based on near-surface atmospheric conditions alone for May and the neighboring months of April and June, the month with the highest ozone levels should have been June owing to more solar radiation and significantly higher temperatures (Table 3). In addition, May and June should have had similar magnitudes of ventilation (Holzworth, 1962). But typical 700-mb wind directions on nonintrusion days in May were significantly different from those in April and June, with GCNP being roughly downwind of southern California in May and to a lesser extent in June (Table 3). Pollutant transport along a straight trajectory at 700 mb from southern California to GCNP would be maximized when the wind direction was between 220° and 265°. Specifically, southern California was upwind of GCNP on over 40% of the nonintrusion days in May, while the percentage for both April and June was ~30% (Fig. 8). Not only did May have the highest daily SUM60 values of all the months, but those SUM60 values had the least amount of interdiurnal variability (Fig. 7). Predominant southwesterly air flow in May seems to have resulted in steady pollutant transport from southern California and thus consistently high ozone levels at GCNP.

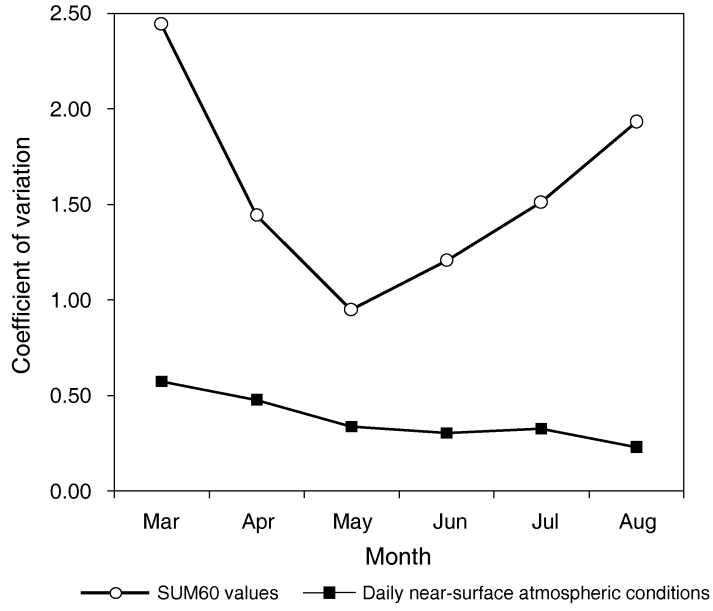

Fig. 7. Monthly variation in coefficients of variation on nonintrusion days for daily SUM60 values and daily near-surface atmospheric conditions, which is the mean of coefficients of variation for mean daily temperature, mean daily temperature range, mean daily relative humidity, and mean daily wind speed.

It needs to be noted that examinations of back-trajectories for randomly selected days in May also indicated that southern California is nearly always the most likely source region for atmospheric pollution at GCNP. Rarely did the trajectories reach urbanized areas of Asia. Therefore, unlike remote locales in the Pacific Northwest (e.g., Jaffe et al., 2003), GCNP was not impacted substantially by Asian-derived atmospheric pollution.

Atmospheric pollution from southern California probably assisted in causing nonintrusion HODs at GCNP from March through June. Although nonintrusion HODs in April and especially March may have been caused partially by enhanced *in situ* ozone production (Table 4), pollutant transport from southern California to GCNP also contributed: those days typically had southwesterly air flow over northern Arizona in the middle troposphere (Table 5; Figs. 8 and 9). Conversely, westerly flow prevailed on the remainder of the nonintrusion days as well as on a typical nonintrusion day (Table 5; Figs. 8 and 9). The situation was slightly different in May and June, for *in situ* ozone production was less important than in the earlier months. In fact, atmospheric conditions on nonintrusion HODs and the remainder of the nonintrusion days in May were not statistically different (Tables 4 and 5). A typical nonintrusion day in May—and to a lesser extent a typical nonintrusion day in June—had conditions conducive to pollutant transport from southern California to GCNP (Fig. 10).

Table 3. Mean Near-Surface and Upper-Level Atmospheric Conditions on Nonintrusion Days in April, May, and June[ab]

Month	TEMP	RANGE	RH	SLP	WS	700H	700T	700DPT	700WS	700WD	500H	500T	500DPT	500WS	500WD
April	**6.1**	**20.3**	**44.5**	**1,014.6**	3.3	**3,064**	**-0.5**	**-12.9**	10.3	**246**	**5,669**	**-17.3**	**-33.2**	16.6	**269**
May	12.4	23.1	33.1	1,013.2	3.7	3,101	5.4	-10.3	10.3	233	5,761	-12.2	-27.5	15.3	253
June	**16.6**	**24.6**	**29.0**	1,013.2	3.5	**3,134**	**9.2**	**-7.2**	9.8	**222**	**5,828**	**-9.2**	-28.0	**11.8**	**233**

[a]Values in bold type for each month indicate that May days are significantly ($\alpha = .05$) different from days in that particular month. [b]TEMP = mean daily temperature (ºC); RANGE = mean daily temperature range (ºC); RH = mean daily relative humidity (%); SLP = mean daily sea-level pressure (mb); WS = mean daily wind speed (m s^{-1}); 700H = geopotential height (m) at 700 mb; 700T = temperature (ºC) at 700 mb; 700DPT = dew-point temperature (ºC) at 700 mb; 700WS = wind speed (m s^{-1}) at 700 mb; 700WD = wind direction (º) at 700 mb; 500H = geopotential height (m) at 500 mb; 500T = temperature (ºC) at 500 mb; 500DPT = dew-point temperature (ºC) at 500 mb; 500WS = wind speed (m s^{-1}) at 500 mb; 500WD = wind direction (º) at 500 mb.

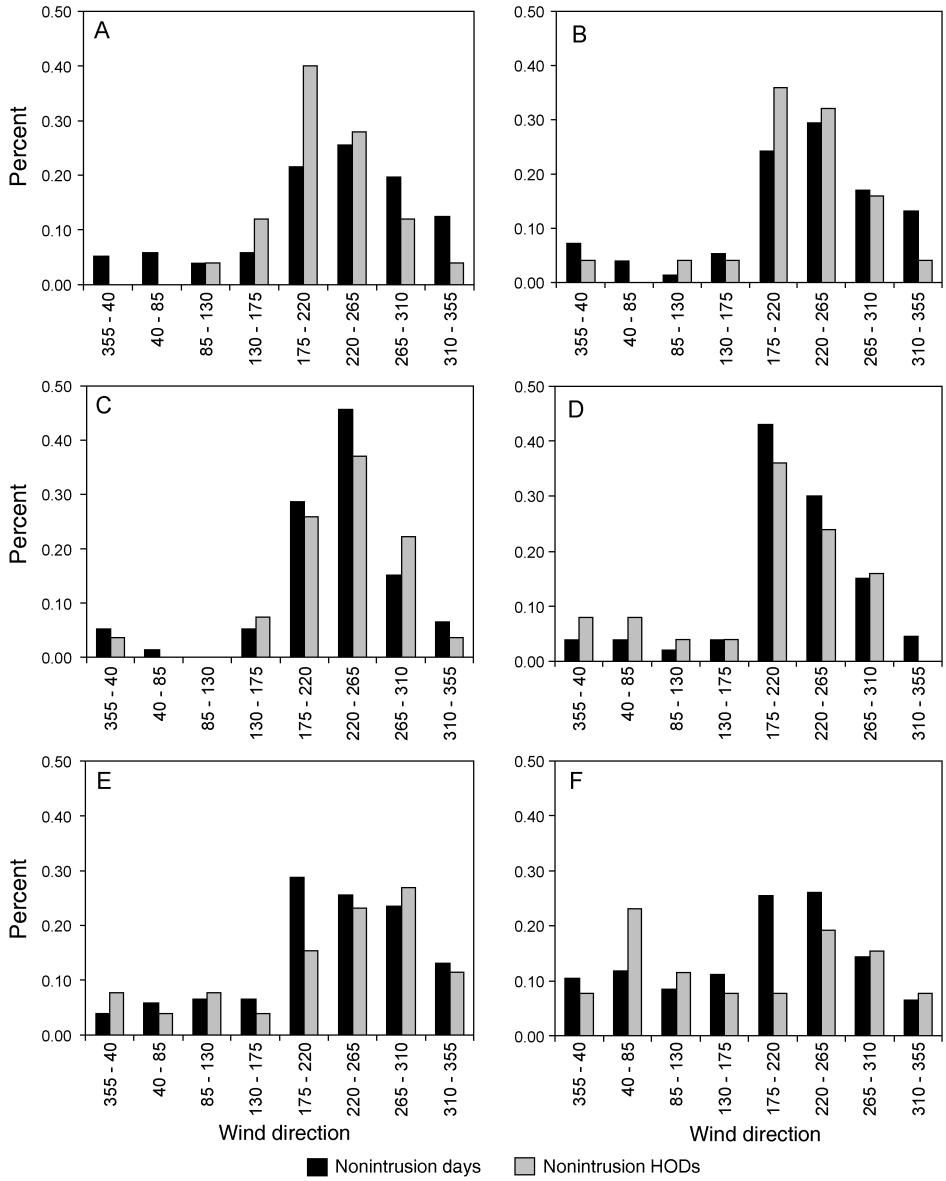

Fig. 8. Histograms of 700-mb wind directions on nonintrusion days and nonintrusion HODs for (A) March, (B) April, (C) May, (D) June, (E) July, and (F) August.

Although pollutant transport from southern California to GCNP on nonintrusion HODs may have been present in July and August, it was not the dominating factor. The expansion of a subtropical high-pressure zone resulted in weak and variable mid-tropospheric winds (Table 5; Figs. 8 and 11). Therefore, instead of pollutant

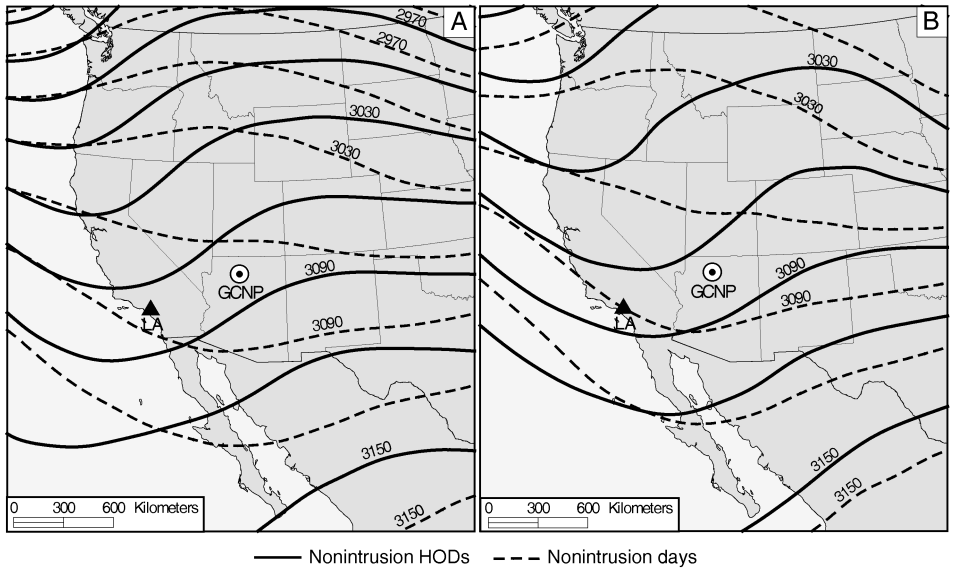

Fig. 9. Typical 700-mb surfaces for nonintrusion HODs and all nonintrusion days in (A) March and (B) April. Contour interval is 30 m.

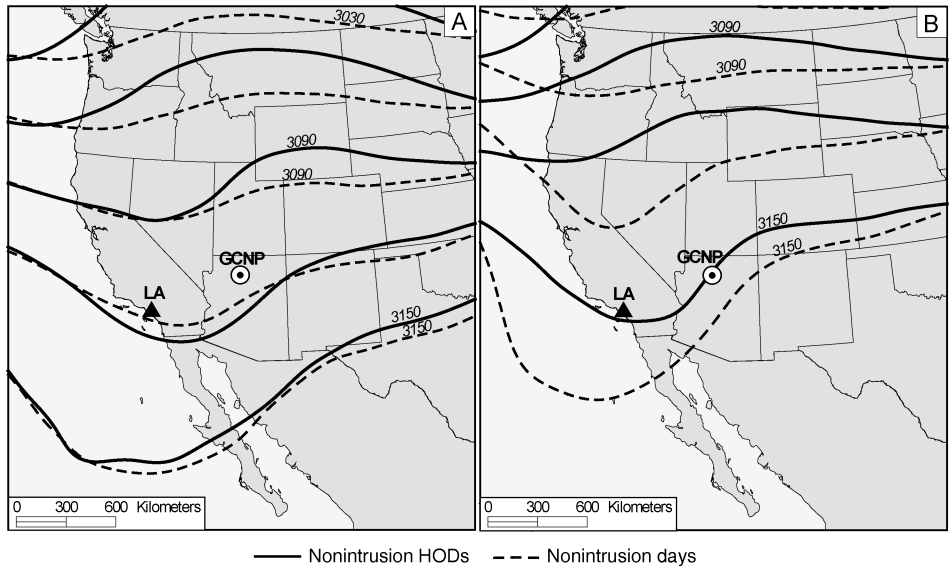

Fig. 10. Typical 700-mb surfaces for nonintrusion HODs and all nonintrusion days in (A) May and (B) June. Contour interval is 30 m.

transport from southern California, the likely cause of nonintrusion HODs at GCNP during the summer months was increased *in situ* ozone production. Nonintrusion HODs had higher temperatures, more solar radiation, and a drier atmosphere than

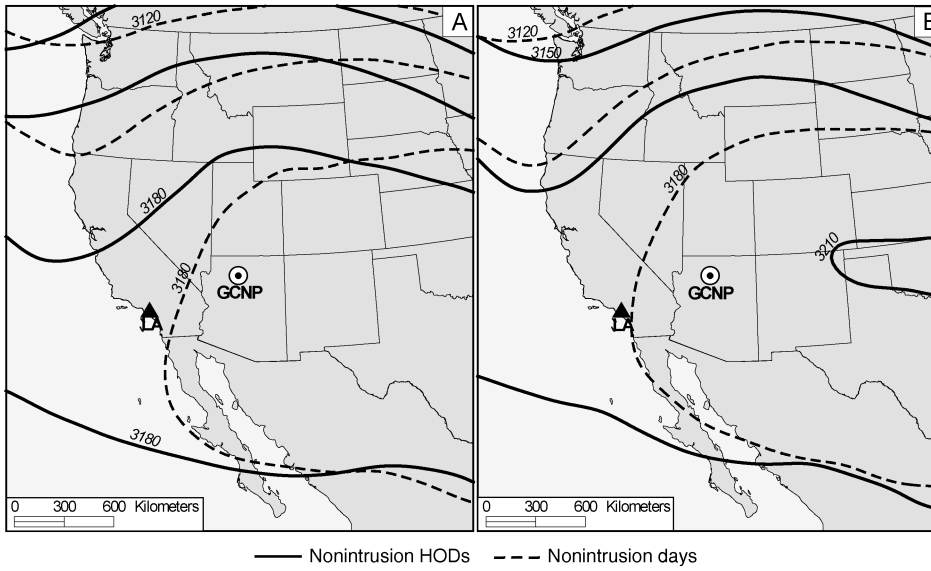

Fig. 11. Typical 700-mb surfaces for nonintrusion HODs and all nonintrusion days in (A) July and (B) August. Contour interval is 30 m.

did the remainder of the nonintrusion days (Tables 4 and 5). Ozone-precursor emissions and *in situ* ozone production at GCNP theoretically reached a pinnacle on nonintrusion HODs in July and August.

The ozone budget at GCNP during the spring months appears to be supplemented by pollution from southern California. The hypothesized pollutant-transport process is as follows: convection and mountain venting in southern California inject pollutants into the free troposphere; as a result, pollutants have the potential to be transported over relatively large distances before being down-mixed to the surface by the afternoon deepening of a relatively thick atmospheric boundary layer at GCNP (refer to review in McKendry and Lundgren, 2000). The lifetime of ozone in the free troposphere typically surpasses 10 days (Liu et al., 1980), and the transport time from Los Angeles or San Diego to GCNP is probably less than one day. The distance from the approximate centroid of the heavily urbanized lands of southern California to GCNP was -550 km. The mean wind speed in May at 700-mb over northern Arizona was -10 m s^{-1}. The travel time was distance divided by velocity. By May, the maximum depth of the atmospheric boundary layer should approach 3,000 m over GCNP (Holzworth, 1962), thus the boundary layer expands vertically to entrain elevated pollutant layers in the free troposphere.

CONCLUSIONS

Analyses of ground-level ozone concentrations, near-surface atmospheric conditions, and upper-level atmospheric conditions from 1996 to 2000 at GCNP

Table 4. Mean Near-Surface Atmospheric Conditions on Nonintrusion HODs and Remainder of Nonintrusion Days in March, April, May, June, July, and August[a]

Month	TEMP	RANGE	RH	SLP	WS
MAR					
HOD	**5.2**	**23.7**	**35.8**	1,015.2	3.4
Remainder	3.1	19.8	52.1	1,016.3	3.1
APR					
HOD	**7.4**	20.7	40.3	1,013.5	**3.9**
Remainder	6.0	20.1	45.5	1,014.3	3.2
MAY					
HOD	12.4	23.9	30.5	1,013.0	3.3
Remainder	12.6	23.0	33.2	1,013.1	3.8
JUN					
HOD	16.8	25.7	**25.2**	**1,014.2**	3.1
Remainder	16.5	24.4	29.9	1,013.0	3.6
JUL					
HOD	**20.7**	**24.5**	**31.4**	1,015.6	2.6
Remainder	19.9	19.0	46.0	1,015.9	2.6
AUG					
HOD	**20.1**	**21.7**	**42.8**	1,017.4	2.2
Remainder	18.5	18.1	54.5	1,017.1	2.2

[a]Values in bold type indicate that HODs are significantly ($\alpha = .05$) different from non-HODs

[b]TEMP = mean daily temperature (ºC); RANGE = mean daily temperature range (ºC); RH = mean daily relative humidity (%); SLP = mean daily sea-level pressure (mb); WS = mean daily wind speed (m s^{-1}).

revealed that the May peak in ozone concentrations at the site is probably caused by atmospheric pollution originating from southern California. This conclusion supports the findings by other researchers (Ashbaugh, 1983; Malm et al., 1990; White et al., 1990; Pryor and Hoffer, 1992; Davis and Gay, 1993a, 1993b; Johnson et al., 1994; White et al., 1994; de P. Vasconcelos et al., 1996; Pryor et al., 1995; Henmi and Bresch, 1998) for particulate matter in the general vicinity of GCNP. GCNP was typically downwind of southern California in May, and it is doubtful that the occurrence of the highest ozone levels at GCNP in May and the presence of southwesterly mid-tropospheric winds were coincidental. Southern California pollution also was the most logical cause of the highest daily ozone levels in the other spring months.

The stratosphere was a minor contributor to the ground-level ozone budget at GCNP during all months. This stems from an overall lack of days (i.e., <25%) with mid-tropospheric conditions conducive to stratospheric intrusions. Moreover, only ~3% of the days were classified as highly probable intrusion days, and the

Table 5. Mean Upper-Level Atmospheric Conditions on Nonintrusion HODs and Remainder of Nonintrusion Days in March, April, May, June, July, and August[ab]

Month	700H	700T	700DPT	700WS	700WD	500H	500T	500DPT	500WS	500WD
March										
HOD	3,067	**0.4**	**-21.5**	**11.6**	**221**	5,686	-16.7	**-38.3**	17.6	**249**
Remainder	3,060	-1.6	-15.1	9.2	276	5,660	-18.0	-32.6	16.0	283
April										
HOD	3,065	0.3	-12.9	11.0	**221**	5,670	-17.0	-33.9	18.6	**247**
Remainder	3,061	-0.6	-12.6	10.4	250	5,667	-17.3	-32.8	16.5	269
May										
HOD	3,100	5.4	-11.3	9.6	231	5,761	-12.3	-28.4	14.4	239
Remainder	3,103	5.4	-9.7	10.5	232	5,765	-12.1	-27.0	15.3	253
June										
HOD	**3,147**	10.3	**-9.3**	8.9	224	5,845	-9.2	-28.8	**9.7**	240
Remainder	3,131	9.0	-6.7	10.0	222	5,824	-9.2	-27.7	12.3	232
July										
HOD	3,186	**13.8**	**-5.8**	6.1	258	**5,911**	-6.9	**-26.7**	7.9	**255**
Remainder	3,179	11.9	0.3	6.0	230	5,895	-7.4	-17.6	6.3	194
August										
HOD	**3,200**	**13.4**	**-0.5**	5.1	**257**	**5,921**	-7.3	-22.5	8.8	186
Remainder	3,185	11.5	2.7	5.4	219	5,897	-7.7	-18.2	7.2	202

[a]Values in bold type indicate that HODs are significantly ($\alpha = .05$) different from non-HODs.

[b]700H = geopotential height (m) at 700 mb; 700T = temperature (ºC) at 700 mb; 700DPT = dew-point temperature (ºC) at 700 mb; 700WS = wind speed (m s^{-1}) at 700 mb; 700WD = wind direction (º) at 700 mb; 500H = geopotential height (m) at 500 mb; 500T = temperature (ºC) at 500 mb; 500DPT = dew-point temperature (ºC) at 500 mb; 500WS = wind speed (m s^{-1}) at 500 mb; 500WD = wind direction (º) at 500 mb.

intrusions occurring on those days appeared to be responsible for less than 2% of the annual SUM0 value. Nevertheless, a suspected intrusion on April 14, 1999, contributed to the highest hourly ozone concentration measured at GCNP from 1996–2000. The overall conclusion that stratospheric intrusions provide little enhancement of the ground-level ozone budget at GCNP differs from that of Wooldridge et al. (2000) who estimated that stratospheric intrusions contributed ~10% to ground-level ozone concentrations in May at a high-elevation site in southeastern Wyoming. The major difference in contribution (2% vs. 10%) is related to the location of GCNP: synoptic-scale pollutant transport from urban areas in the western United States has a strong potential for supplementing the ground-level ozone budget at GCNP, especially in May.

Tracer-based research in more remote areas of the southwestern United States is needed to verify fully the conclusions presented in this paper. Firstly, continuous ozone monitors need to be established in high-elevation locations more remote than GCNP to improve the detection of stratospheric intrusions. Secondly, surface

measurements of beryllium-7 need to be made to reduce the uncertainty involved in identifying stratospheric intrusions (Davies and Schuepbach, 1994). Thirdly, methylchloroform should be measured intensively over space and time to determine the influence of urban areas in southern California and other parts of the western United States. At the present time for GCNP, it can be stated with only a moderate amount of certainty that there is a weak contribution of ozone from stratospheric intrusions and that southern California is responsible for the spring peak in ozone levels.

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