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Background ozone levels of air entering the west coast of the US and assessment of longer-term changes

Samuel J. Oltmans [a,*], Allen S. Lefohn [b], Joyce M. Harris [a,c], Douglas S. Shadwick [d]

[a] NOAA Earth System Research Laboratory, Global Monitoring Division, 325 Broadway, Boulder, CO 80305, USA
[b] A.S.L. & Associates, 302 North Last Chance Gulch, Helena. MT 59601, USA
[c] STC, Boulder, CO 80305, USA
[d] 320 Eastwood Road, Chapel Hill, NC 27514, USA

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ABSTRACT

An analysis of surface ozone measurements at a west coast site in northern California (Trinidad Head) demonstrates that this location is well situated to sample air entering the west coast of the US from the Pacific Ocean. During the seasonal maximum in the spring, this location regularly observes hourly average ozone mixing ratios $\geqslant$50 ppbv in air that is uninfluenced by the North American continent. Mean daytime values in the spring exceed 40 ppbv. A location in southern California (Channel Islands National Park) demonstrates many of the characteristics during the spring as Trinidad Head in terms of air flow patterns and ozone amounts suggesting that background levels of ozone entering southern California from the Pacific Ocean are similar to those in northern California. Two inland locations (Yreka and Lassen Volcanic National Park) in northern California with surface ozone data records of 20 years or more are more difficult to interpret because of possible influences of local or regional changes. They show differing results for the long-term trend during the spring. The 10-year ozone vertical profile measurements obtained with weekly ozonesondes at Trinidad Head show no significant longer-term change in tropospheric ozone.

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1. Introduction

Ozone in the troposphere plays a significant role (1) as an absorber of infrared radiation (i.e., greenhouse gas), (2) in the cleansing capacity of the atmosphere as a precursor of hydroxyl radical formation, and (3) as a regulated air pollutant capable of deleterious health and ecosystem effects (US EPA, 2006). Knowledge of the ozone budget in the troposphere over North America (NA) is required to properly understand the various mechanisms that contribute to the measured distribution and to develop and test models capable of simulating and predicting this important component in atmospheric chemical and physical processes (Forster et al., 2007). In developing a strategy for future mitigation of the impacts of climate- and anthropogenic-caused changes in tropospheric ozone levels, it is necessary to have a quantitative assessment of the current status of ozone levels in the troposphere.

Several investigators (e.g., Jaffe et al., 2003; Parrish et al., 2004; Jaffe and Ray, 2007) have proposed that the effect of Asian emissions on surface ozone concentrations may be enhancing ozone levels in the US inferring that it may be more difficult to attain environmental goals. Models have also considered the role of ozone produced outside of the North American boundary layer on background ozone levels over the continental US (Fiore et al., 2002, 2003). Because of anthropogenic and natural contributions, it is important to understand the levels of background ozone entering and ozone exiting the North

* Corresponding author. Tel.: +1 303 497 6676; fax: +1 303 497 5590.
E-mail address: Samuel.J.Oltmans@noaa.gov (S.J. Oltmans).

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American continent and the modification of tropospheric ozone over a continent, where a number of chemical and dynamical mechanisms occur. At almost all continental locations where surface ozone measurements are made, multiple sources are likely to contribute to the observed ozone distribution. Depending on the flow pattern of air parcels reaching a near-coastal western US location, the observed ozone amounts may represent air that is almost exclusively from long-range transport over the Pacific Ocean, air that has recirculated over the ocean from the North American continent, air that has been influenced by direct over-continent flow, or air contaminated by nearby ozone precursor emissions that reflect locally enhanced or depleted ozone values. In this work, surface ozone observations from two western US locations are evaluated to determine to what extent the sites represent air primarily representative of ozone levels with minimal NA continental or nearby local influence and hence provide information on tropospheric ozone behavior of air entering the US from the Pacific Ocean. By identifying

Fig. 1. Locations of monitoring sites included in this analysis. Top panel—Yreka (41.7°N, 122.6°W), Trinidad (41.1°N, 124.1°W), Lassen Volcanic NP (40.5°N, 121.5°W), Redding (40.6°N, 122.4°W). Bottom panel–Channel Islands NP (34.5°N, 119.4°W).

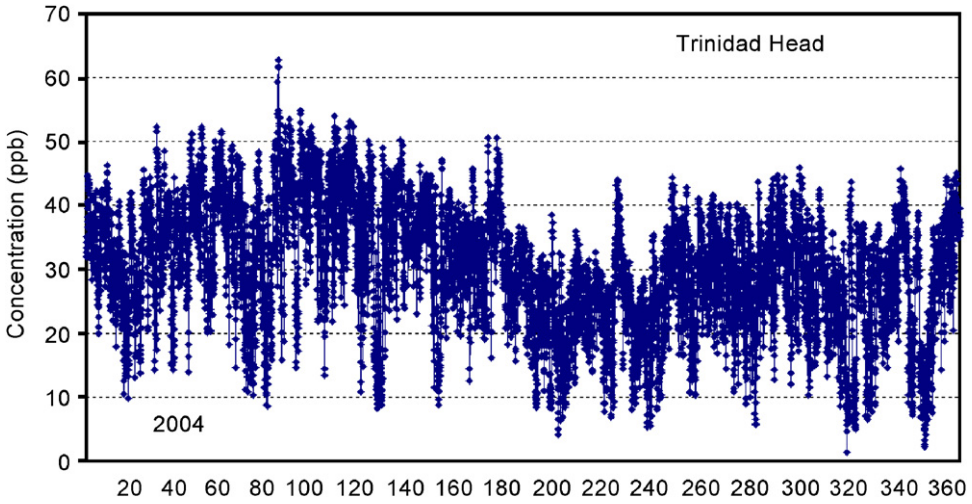

Fig. 2. Hourly average ozone-mixing ratios at Trinidad Head for the year 2004.

Table 1
Summary of hourly average percentiles (ppm), number of hourly occurrences ⩾0.05 ppm (N50), 24-h SUM06 (ppm-h), and 24-h W126 (ppm-h) cumulative exposure values for 2002–2007 at Trinidad Head

Year	Month	*N*	Min	P10	P30	P50	P70	P90	P95	P99	Max	N50	SUM06	W126
2002	April	288	0.015	0.028	0.035	0.041	0.044	0.050	0.052	0.054	0.054	36	0.000	0.642
2002	May	744	0.009	0.022	0.032	0.039	0.043	0.047	0.048	0.051	0.053	18	0.000	1.142
2002	June	718	0.007	0.017	0.023	0.028	0.032	0.037	0.038	0.040	0.041	0	0.000	0.235
2002	July	744	0.008	0.014	0.020	0.024	0.027	0.031	0.032	0.034	0.045	0	0.000	0.115
2002	August	710	0.007	0.013	0.018	0.023	0.027	0.032	0.034	0.038	0.043	0	0.000	0.118
2002	September	720	0.006	0.014	0.021	0.026	0.030	0.035	0.038	0.041	0.044	0	0.000	0.187
2002	October	744	0.005	0.018	0.024	0.029	0.033	0.038	0.040	0.046	0.048	0	0.000	0.321
2002	November	711	0.005	0.011	0.022	0.031	0.035	0.040	0.044	0.049	0.052	5	0.000	0.422
2002	December	737	0.013	0.025	0.031	0.035	0.039	0.043	0.044	0.048	0.050	2	0.000	0.667
2003	January	739	0.008	0.014	0.021	0.028	0.034	0.038	0.041	0.044	0.046	0	0.000	0.312
2003	February	660	0.014	0.023	0.029	0.034	0.038	0.044	0.045	0.048	0.050	1	0.000	0.587
2003	March	713	0.012	0.028	0.034	0.038	0.042	0.047	0.049	0.051	0.052	23	0.000	1.076
2003	April	720	0.023	0.036	0.042	0.046	0.048	0.052	0.054	0.058	0.064	187	0.310	2.722
2003	May	743	0.011	0.029	0.036	0.041	0.045	0.050	0.052	0.054	0.057	96	0.000	1.773
2003	June	720	0.016	0.025	0.032	0.037	0.040	0.044	0.046	0.053	0.054	13	0.000	0.816
2003	July	744	0.003	0.015	0.020	0.023	0.026	0.031	0.032	0.036	0.041	0	0.000	0.109
2003	August	740	0.007	0.020	0.024	0.027	0.030	0.034	0.035	0.038	0.041	0	0.000	0.185
2003	September	720	0.006	0.016	0.025	0.030	0.035	0.040	0.042	0.054	0.060	13	0.120	0.507
2003	October	743	0.004	0.016	0.025	0.028	0.033	0.038	0.040	0.044	0.047	0	0.000	0.300
2003	November	696	0.006	0.023	0.028	0.032	0.035	0.039	0.041	0.046	0.047	0	0.000	0.396
2003	December	725	0.004	0.024	0.029	0.033	0.037	0.041	0.042	0.045	0.050	1	0.000	0.492
2004	January	742	0.010	0.022	0.028	0.032	0.036	0.041	0.043	0.047	0.052	2	0.000	0.504
2004	February	691	0.014	0.027	0.033	0.038	0.041	0.046	0.048	0.051	0.052	15	0.000	0.934
2004	March	744	0.009	0.019	0.028	0.036	0.042	0.048	0.050	0.054	0.063	46	0.186	1.192
2004	April	719	0.011	0.026	0.035	0.041	0.044	0.048	0.050	0.052	0.054	41	0.000	1.382
2004	May	734	0.008	0.016	0.031	0.036	0.040	0.044	0.045	0.048	0.050	4	0.000	0.731
2004	June	719	0.013	0.023	0.029	0.033	0.036	0.040	0.044	0.047	0.051	2	0.000	0.476
2004	July	744	0.004	0.014	0.019	0.023	0.027	0.031	0.033	0.036	0.039	0	0.000	0.109
2004	August	743	0.005	0.014	0.020	0.024	0.027	0.033	0.036	0.041	0.044	0	0.000	0.149
2004	September	697	0.010	0.020	0.026	0.029	0.033	0.038	0.040	0.043	0.044	0	0.000	0.286
2004	October	744	0.006	0.020	0.025	0.029	0.034	0.038	0.041	0.044	0.046	0	0.000	0.339
2004	November	719	0.001	0.011	0.020	0.025	0.030	0.035	0.037	0.041	0.044	0	0.000	0.177
2004	December	744	0.002	0.012	0.023	0.030	0.034	0.039	0.041	0.044	0.046	0	0.000	0.347
2005	January	744	0.007	0.017	0.025	0.031	0.036	0.040	0.043	0.045	0.048	0	0.000	0.422
2005	February	670	0.011	0.022	0.028	0.032	0.037	0.041	0.044	0.048	0.051	4	0.000	0.480
2005	March	700	0.006	0.021	0.032	0.040	0.046	0.050	0.052	0.054	0.056	92	0.000	1.553
2005	April	720	0.011	0.028	0.036	0.041	0.045	0.051	0.053	0.055	0.057	87	0.000	1.640
2005	May	721	0.006	0.012	0.028	0.034	0.037	0.043	0.046	0.050	0.053	10	0.000	0.627

Table 1 (*continued*)

Year	Month	*N*	Min	P10	P30	P50	P70	P90	P95	P99	Max	N50	SUM06	W126
2005	June	720	0.008	0.020	0.025	0.029	0.033	0.040	0.042	0.045	0.047	0	0.000	0.333
2005	July	744	0.005	0.015	0.020	0.023	0.027	0.032	0.034	0.037	0.040	0	0.000	0.121
2005	August	744	0.003	0.012	0.018	0.022	0.027	0.031	0.033	0.036	0.038	0	0.000	0.105
2005	September	720	0.005	0.013	0.020	0.026	0.030	0.035	0.036	0.040	0.045	0	0.000	0.178
2005	October	744	0.003	0.014	0.020	0.025	0.031	0.037	0.039	0.043	0.048	0	0.000	0.244
2005	November	719	0.003	0.013	0.021	0.028	0.034	0.039	0.042	0.045	0.047	0	0.000	0.321
2005	December	737	0.006	0.017	0.023	0.029	0.035	0.041	0.042	0.049	0.051	6	0.000	0.430
2006	January	692	0.009	0.027	0.032	0.034	0.037	0.043	0.045	0.048	0.049	0	0.000	0.622
2006	February	671	0.010	0.024	0.031	0.037	0.041	0.046	0.047	0.050	0.056	8	0.000	0.817
2006	March	741	0.019	0.034	0.040	0.043	0.046	0.049	0.051	0.054	0.057	69	0.000	1.924
2006	April	720	0.013	0.030	0.037	0.042	0.045	0.050	0.052	0.058	0.061	88	0.061	1.804
2006	May	743	0.008	0.025	0.034	0.039	0.043	0.049	0.052	0.061	0.066	60	0.571	1.596
2006	June	720	0.007	0.020	0.027	0.031	0.034	0.038	0.040	0.044	0.048	0	0.000	0.347
2006	July	744	0.010	0.016	0.021	0.024	0.027	0.031	0.032	0.035	0.036	0	0.000	0.115
2006	August	715	0.007	0.017	0.023	0.027	0.031	0.033	0.035	0.039	0.040	0	0.000	0.170
2006	September	718	0.005	0.015	0.023	0.028	0.033	0.040	0.044	0.049	0.051	5	0.000	0.397
2006	October	744	0.006	0.018	0.027	0.030	0.035	0.040	0.042	0.049	0.052	4	0.000	0.443
2006	November	720	0.007	0.020	0.028	0.034	0.039	0.044	0.045	0.047	0.049	0	0.000	0.625
2006	December	685	0.010	0.020	0.026	0.031	0.036	0.042	0.043	0.046	0.050	2	0.000	0.437
2007	January	696	0.005	0.016	0.024	0.028	0.034	0.038	0.040	0.043	0.045	0	0.000	0.287
2007	February	671	0.008	0.024	0.032	0.037	0.041	0.045	0.047	0.049	0.050	3	0.000	0.806
2007	March	743	0.008	0.023	0.031	0.036	0.041	0.048	0.051	0.053	0.054	54	0.000	1.129
2007	April	719	0.007	0.028	0.036	0.041	0.045	0.050	0.052	0.058	0.060	82	0.119	1.690
2007	May	744	0.014	0.027	0.034	0.039	0.044	0.049	0.051	0.054	0.057	71	0.000	1.470
2007	June	720	0.013	0.021	0.026	0.029	0.033	0.036	0.038	0.040	0.041	0	0.000	0.258
2007	July	700	0.004	0.014	0.020	0.024	0.026	0.030	0.031	0.034	0.034	0	0.000	0.093
2007	August	743	0.006	0.016	0.022	0.025	0.028	0.032	0.034	0.041	0.045	0	0.000	0.149
2007	September	720	0.011	0.019	0.026	0.030	0.034	0.039	0.040	0.044	0.046	0	0.000	0.327
2007	October	742	0.006	0.018	0.024	0.029	0.033	0.040	0.042	0.047	0.049	0	0.000	0.360
2007	November	720	0.004	0.016	0.024	0.028	0.032	0.039	0.043	0.048	0.050	1	0.000	0.333
2007	December	569	0.004	0.020	0.028	0.033	0.037	0.042	0.043	0.044	0.045	0	0.000	0.380

the conditions under which air entering the west coast of the US is free from the influence of the North American continent, changes in tropospheric ozone on a hemispheric or global scale can be separated from those more directly attributable to North American ozone sources.

Surface ozone data are characterized by diurnal and seasonal patterns. Three-dimensional trajectories are employed to identify ozone measurements that have clear flow patterns from off the Pacific Ocean and the ozone amounts associated with these observations are categorized. The spring season is identified as having the strongest flow from off the ocean along with the highest average ozone amounts of the four seasons for the sites included in this study.

Although background ozone is generally thought of in terms of surface measurements, air flowing into the North American continent associated with long-range transport over the Pacific occurs above the surface. Hence air reaching the west coast and the interior of NA from over the Pacific has likely been mixed into the boundary layer from above (Cooper et al., 2004). The 10-year record of weekly ozone profile observations (with several intensive periods of daily measurements) at Trinidad Head, California provide a picture of the background ozone content of air in the lower troposphere above the boundary layer entering the continental US. This record is of sufficient length to give some indication of changes in the ozone content of air entering NA at altitudes above the boundary layer, where some of the complications related to near-surface processes are not present.

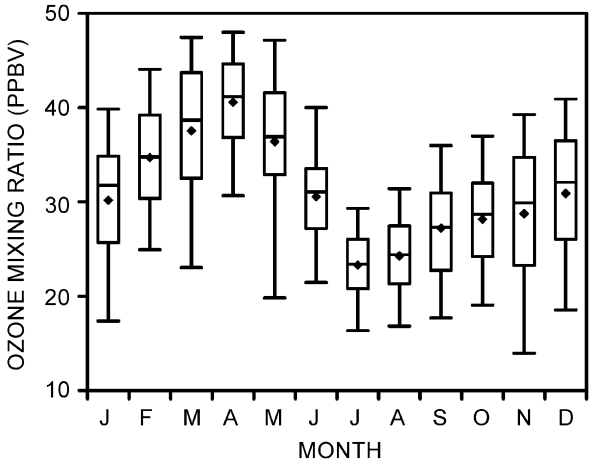

Fig. 3. Seasonal variation in surface ozone mixing ratio at Trinidad Head, California. The diamond is the monthly mean, the horizontal bar the median, the box the inner 50th percentile (25–75th percentile), and the whiskers the inner 90th percentile (5–95th percentile). Data are for the period 2002–2007 and are based on daily averages for all hours of the day.

Because several investigators (Yienger et al., 2000; Jaffe et al., 2003; Parrish et al., 2004; Jaffe and Ray, 2007) have described possible longer-term changes in the chemical characteristics of air entering the west coast of the US, we have characterized the trends at three California monitoring sites (i.e., Trinidad Head, Yreka, and Lassen Volcanic National Park). Although the two longer-term surface ozone measurement sites in the western US (Yreka and Lassen) considered in this work are not ideally located to measure background ozone levels, their length of record and continuing observations make it worthwhile to investigate their usefulness in providing information on possible changes in background ozone levels over time.

2. Description of observations and approach

A key site for measuring background ozone levels of air entering the west coast of the US is the NOAA Baseline Station at Trinidad Head, California (see Fig. 1 for location of sites discussed). The observing location is on an elevated piece of land that extends out from the main coastline near the small town of Trinidad. During the day, this site almost exclusively intercepts air coming in off the Pacific Ocean during the spring months as will be shown later in a discussion of the air trajectory and local wind measurement results. During other times of the year, the predominant flow is also from off the Pacific Ocean. At night, however, the land breeze often dominates and near-surface observations reflect air that has traveled over the heavily vegetated coastline. This site began surface ozone measurements in April 2002 as part of the Intercontinental Transport and Chemical Transformation 2002 (ITCT 2K2) experiment. During the initial study that ran from mid April to early May 2002, intensive measurements were made of the chemical composition of the atmosphere at this site including ground-based observations, aircraft measurements, and near-daily balloon ozone profile soundings. Surface ozone observations, as well as continuous aerosol and meteorological measurements, continue at this site. A site at Channel Islands National Park off the southern coast of California has a 10-year record of surface ozone observations. By comparing the diurnal and seasonal ozone variations and flow patterns at Channel Islands with those at Trinidad Head, ozone levels are characterized for conditions when background air is observed at Channel Islands. Surface ozone data for Channel Islands, as well as from Yreka, California, Lassen Volcanic National Park, California, and the site in Redding, California were extracted from the US EPA's Air Quality System (AQS) database. Surface ozone measurements from Channel Islands National Park were discontinued at the end of 2004.

Ozone profile measurements at Trinidad, California (the small village located ~3 km to the east of Trinidad Head on the mainland) were obtained with standard electrochemical (ECC) ozonesondes. The performance of ozonesondes has been discussed extensively in the literature (Johnson et al., 2002; Thompson et al., 2007; Smit et al., 2007). At Trinidad consistent procedures have been used throughout the 10-year measurement record that began in August 1997 (Johnson et al., 2002) with a precision of tropospheric profiles of ±5%. Soundings are routinely obtained on a weekly schedule; however, several intensive campaigns of approximately 1 month's duration with daily measurements also provide a picture of the strong day-to-day variability that occurs at this location.

Air parcel back trajectory calculations are based on the model of Harris and Kahl (1994) and use the NCEP/NCAR reanalysis data set (Kalnay et al., 1996). Trajectories are calculated for arrival times of 00, 06, 12, and 18 GMT. In order to avoid the influence of effects very close to the surface, the calculations were made at an altitude of 300 m, which is well within the boundary layer for the sites considered here.

Two approaches were used to investigate the longer-term changes in surface ozone at Yreka and Lassen NP and vertical profile changes at Trinidad Head. For the surface sites the analysis of the changes focused on the daytime (1200–1800 LST) ozone values since these are likely to be the most representative of the mixed boundary layer behavior. At night very local effects in a shallow boundary layer may dominate. The overall ozone trend is computed in a two-step process (Harris et al., 2001). The first step uses an autoregressive model that incorporates explanatory variables and a cubic polynomial fit rather than a straight line for better representation of the long-term

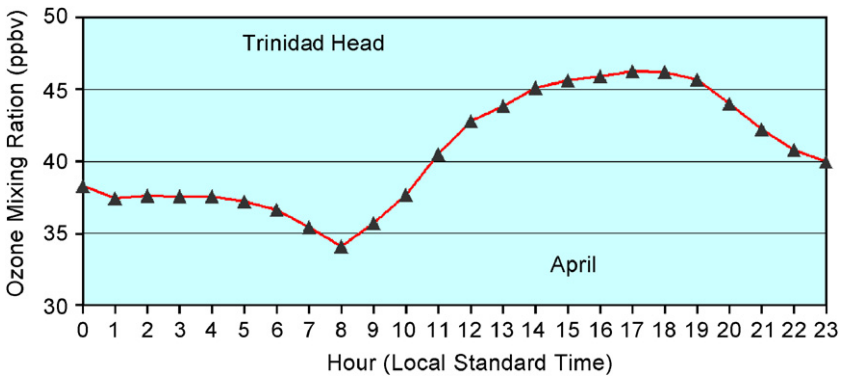

Fig. 4. Composite (2002–2005) diurnal pattern of surface ozone mixing ratio at Trinidad Head, California for the month of April. The 13–21 LST average concentration is 46 ppbv.

variations. The model accounts for serial correlation in the ozone data and minimizes the residual variance of the model fit by regressing to known sources of ozone variability. The second step determines the ozone tendency (trend line) and growth-rate curves. A bootstrap method produces 100 statistical realizations of the population from which the original data were drawn by combining the tendency curve with randomly selected residuals (difference between the model fit and the observed monthly values) from the curve according to the month (Harris et al., 2001). The growth-rate curve is obtained by numerical differentiation of the tendency curve with the 95% confidence limits determined using the standard deviation of the growth-rate curves from the 100 bootstrap samples. The average ozone growth rate is a measure of the overall change.

In order to investigate the manner in which the overall changes are a function of the season, the monthly time trend in the distribution of hourly average concentrations for each monitoring site was also investigated. By characterizing the time trend in frequency (expressed as a percent) within each 10 ppb increment (bin) of the distribution for months with significant changes, the shifts in the distribution may help identify processes that dominate during a particular time of the year and are more likely to influence particular portions of the distribution. Overall, 75% of possible hourly values for a given site, month, and year had to be present for the site, month, and year data to be included in the analysis. The 10 ppb bin data were then arranged as a time series by month. The Thiel estimate (Hollander and Wolfe, 1999) was used to characterize the trend estimate. To test for

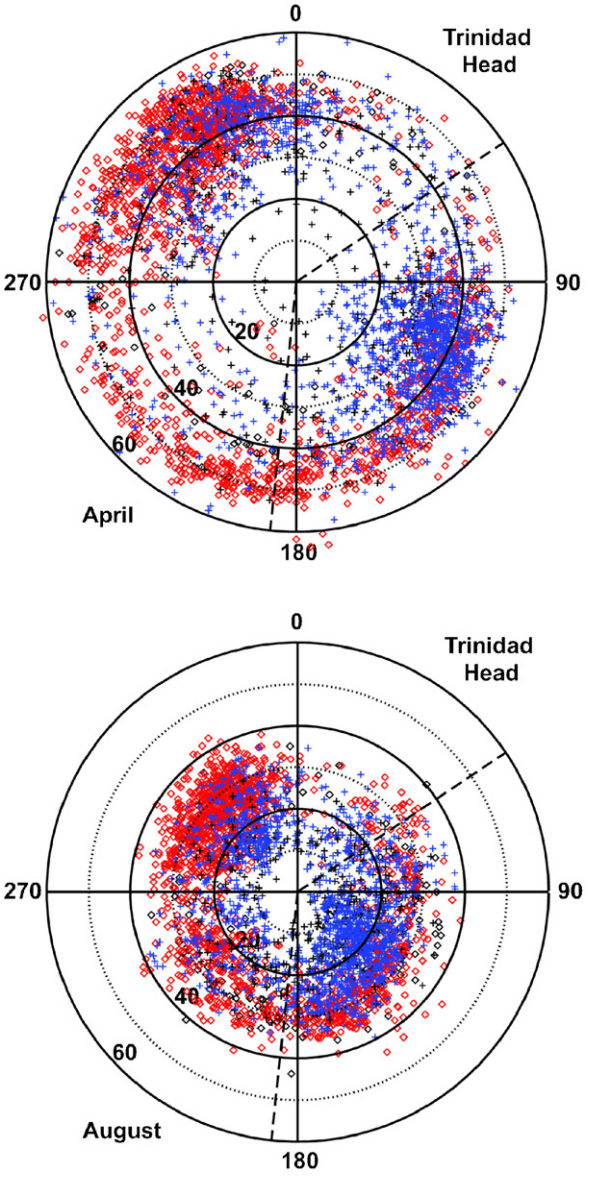

Fig. 5. Wind rose for ozone mixing ratios at Trinidad Head for April and August for the period 2002–2007. The concentric circles mark the ozone mixing from 0 ppbv at the center to 60 ppbv for the outer circle. The red diamonds are daytime (10–21 LST) values and the blue plusses are nighttime (22–09 LST) values. When wind speeds are $<0.5\,m\,s^{-1}$ the symbol is black. The wind direction is labeled by the direction from which the wind is blowing. Between the dashed lines (56–186°), the wind is blowing from over the land to the observing site.

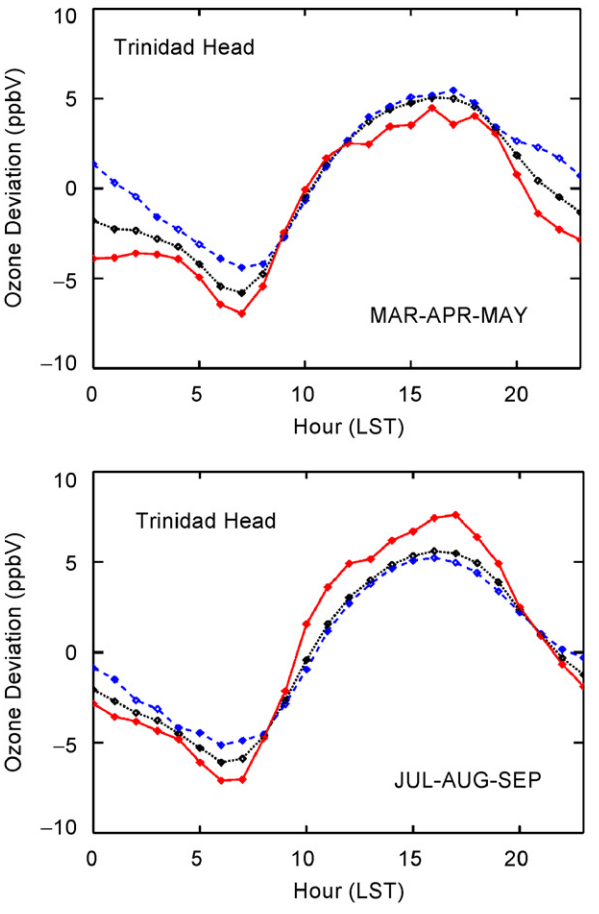

Fig. 6. Average (2002–2007) diurnal pattern of surface ozone at Trinidad Head plotted as a deviation from the seasonal mean for two seasons (March–April–May and July–August–September). The black (dotted) line is for data from all wind directions. The blue (dashed) line is observations from off the ocean and the red (solid) line from off the land (56–186°). Designation of the curves for B/W is given in parenthesis.

statistical significance, Kendall's tau test (Lefohn and Shadwick, 1991) was used to determine significance at the 10% level for the specified increments.

3. Characterizing Trinidad Head surface ozone

3.1. Diurnal and seasonal variation

Surface ozone amounts at Trinidad Head show significant variability on scales from diurnal to seasonal with significant synoptic (weather system) scale variability (Fig. 2) apparent in the record. As will be shown later, this synoptic variability can have a significant impact on ozone amounts over several day periods that reflects transport of air from widely varying sectors even for flow from off the Pacific Ocean. Although measured hourly average ozone levels have never exceeded 70 ppbv occasional hourly amounts exceed 60 ppbv and hourly values of $\geqslant$50 ppbv (N50) are relatively common during the spring (Table 1), which is the season of both the highest individual hourly averages and the average monthly mixing ratios (Fig. 3). A detailed characterization of the surface ozone for each month of the data record is provided in Table 1. The values are given in units of ppm (0.001 ppb) for comparison with various air quality metrics usually expressed in these

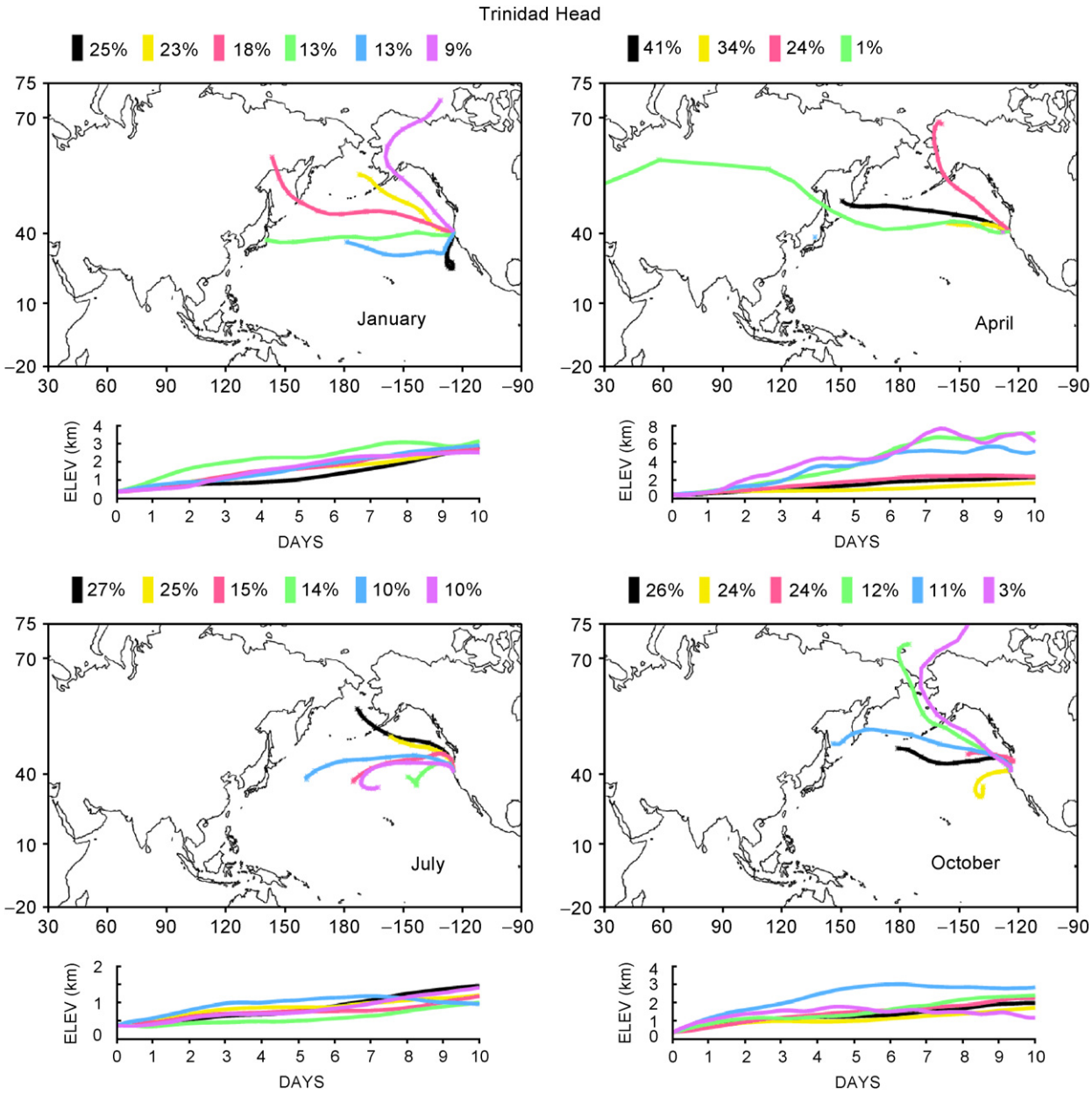

Fig. 7. Clustered back trajectories (1996–2005) for air parcels reaching Trinidad Head, California at 300 m for January, April, July, and October.

units. In addition to the breakdown for various percentile concentration levels two indices for exposures (SUM06 and W126) were calculated. The SUM06 sums all hourly values $\geqslant$0.06 ppm (US EPA, 2006). The cumulative W126 exposure index uses a sigmoidally weighted function as described by Lefohn and Runeckles (1987) and Lefohn et al. (1988). The W126 index focuses on the biologically important higher hourly average concentrations, while retaining the mid- and lower-level values (0.04–0.09 ppm). The high springtime ozone values at Trinidad Head are not driven by active local or regional ozone photochemical ozone production (Goldstein et al., 2004) but rather by ozone transported to the site from the Pacific Ocean as will be shown in the next section. The prominent diurnal variation with highest amounts from late morning through the evening and lowest amounts during the night and early morning (Fig. 4) is strongly influenced by the local meteorology and the ventilation of the observing site (Goldstein et al., 2004). Daytime measurements primarily reflect airflow that reaches the observing site at Trinidad Head from directly off the ocean, while nighttime measurements are often from off the land to the northeast through the south of the observing site. This is illustrated by the wind roses for May and August in Fig. 5, where all of the hourly ozone values for the 6-year data record (2002–2007) are plotted as a function of the wind direction measured at the same time and height as the ozone measurement. For the half-day period 10–21 LST, representing the daytime period, mixing ratios are plotted as red diamonds and nighttime values (22–09 LST) are shown as blue plus symbols. For wind speeds $<$0.5 m s^{-1}, the corresponding symbol (day—diamond, night—plus) is

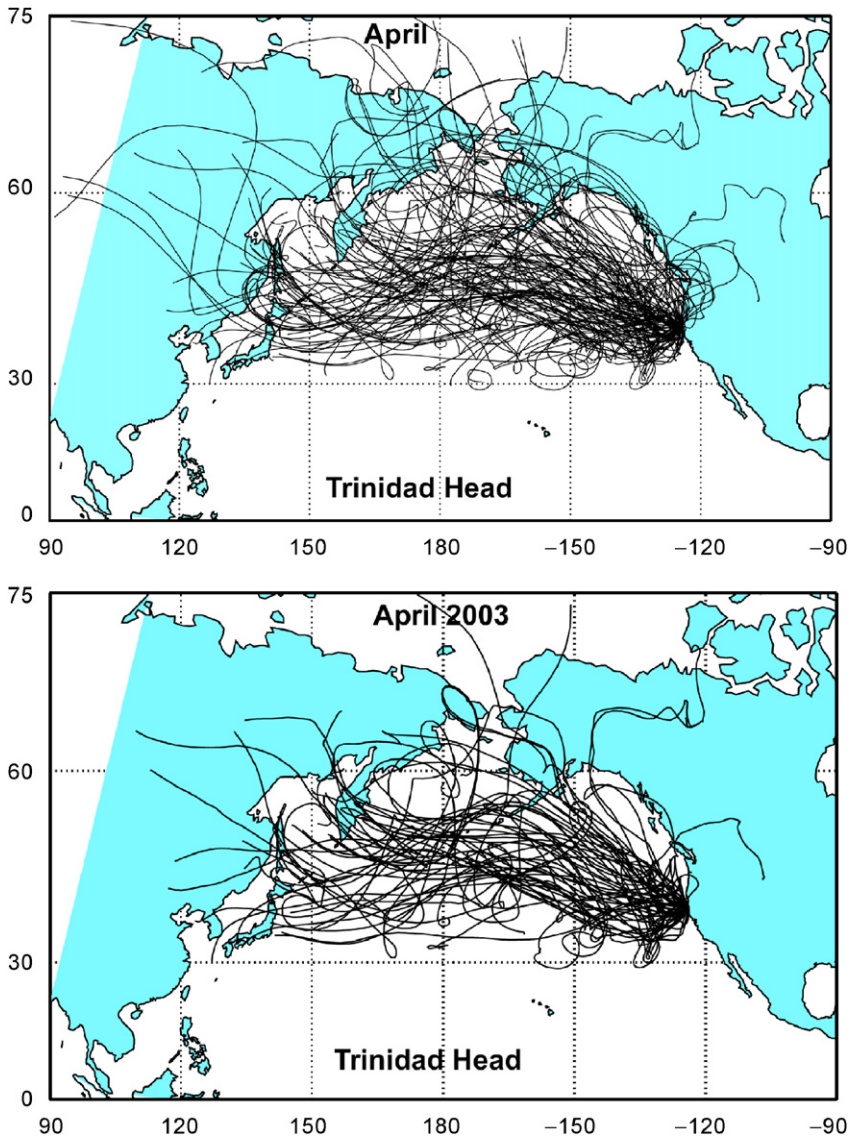

Fig. 8. Back trajectories to Trinidad Head for all days in April 2002–2005 on which there was an hourly average ozone concentration $\geqslant$50 ppbv (upper panel) and for April 2003 (lower panel).

plotted in black. Ozone values when air is coming from off the ocean generally exceed those from air flow off the land. Nighttime ozone amounts even when the local wind is from off the ocean are lower than daytime amounts (Fig. 6). During the spring (March–April–May), ozone values for air coming off the ocean on average exceed those from off the land during all hours. Although relatively infrequent, during the summer (July–August–-September), daytime values from the land can exceed the ocean values. During this period, the overall daytime average is very close to the off ocean value. The observation that nighttime ozone amounts are lower than daytime even when the wind direction is from off the ocean indicates that it is not only deposition at night, when air is coming from over the land that decreases the nighttime values, but that the mixing of air from above the boundary layer also plays a significant role in producing the diurnal variation.

3.2. Air flow and selection of background measurements

The large-scale flow pattern shows that a preponderance of the air parcels reaching the observing site at Trinidad Head are from off the Pacific Ocean as can be seen in the 10-year climatology of back trajectories to this site (Fig. 7), which are calculated four times per day 00, 06, 12 and 18 GMT (16, 22, 04, and 10 LST). On an average this back trajectory pattern is not significantly different for any single one of these times. During all months, a large portion of the trajectories on an average stay over the ocean for the full 10 days of the computed trajectory. During winter and spring, a significant fraction of the trajectories extend back toward Alaska or Asia.

During March, April, and May, there are numerous occurrences of hourly average ozone concentrations $\geqslant$50 ppbv (Table 1). These cases are largely associated with trajectories that reach Trinidad Head without intercepting the North American continent (Fig. 8) and are thus, representative of air reaching NA without the influence of local or regional sources within NA.

While the dominant airflow pattern to Trinidad Head is from the west or northwest during the spring, there are intervals when flow from the southwest may reach this site. Under these conditions daytime ozone levels are markedly less. For example during May 2007 (Fig. 9) there was a 3-day period when daytime averages (12–18 LST) were only about 30 ppb compared to the average daytime value for this season of about 40 ppbv. Although these periods are not frequent (none occurred during the 3-week ITCT-2K2 campaign in April and May 2002) there is usually at least one such event each spring. Back trajectories for 8 and 12 May 2007 (Fig. 10) clearly show the much different origin of air parcels reaching Trinidad Head during the period of lower daytime ozone levels and when ozone levels are at more typical levels for this time of year. Although the air parcels on 8 May come from the north in the 2 days immediately prior to reaching Trinidad Head, they clearly had their origin in the subtropical Pacific where lower altitude tropospheric ozone levels are systematically less than are seen at more northerly Pacific

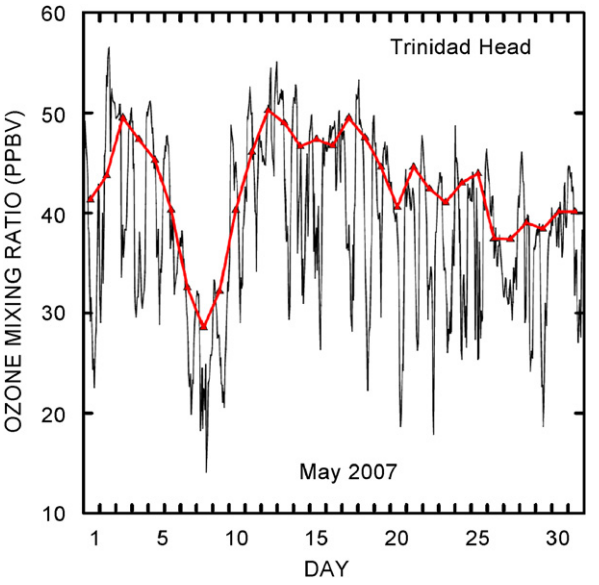

Fig. 9. Hourly surface ozone mixing ratios (thin solid line) and daytime average (11–19 LST) mixing ratios (red-thick solid line) for May 2007 at Trinidad Head.

latitudes (Oltmans et al., 2004). Even monthly median values can vary by ~5 ppbv from year to year during the spring months (Table 1—Column P50). Examples such as the one in May 2007 suggest that caution is appropriate in interpreting measurements obtained over relatively short periods or for a single year; such periods may not represent the overall distribution.

4. Channel islands as a background data set

4.1. Diurnal and seasonal variation and comparison with Trinidad Head

Based on the analysis of the Trinidad Head surface ozone record presented in the previous section, the northern California site provides a solid reference point for characterizing other locations as to their representativeness as background measurement sites for air reaching NA from the Pacific. During the winter through early summer months (December–July), the seasonal pattern of ozone mixing ratio measured at Channel Islands National Park replicates that at Trinidad Head (Fig. 11) but departs markedly from this pattern from late summer through the autumn months. From August through November, surface ozone amounts at Channel Islands rise to a secondary maximum in October that is nearly equal to the spring maximum that occurs in April. Unlike Trinidad Head, there are occurrences of hourly average ozone mixing ratios in both the spring (infrequently) and in other months (most frequently in autumn) that exceed 70 ppbv. As will be discussed in the following section, the cases where ozone exceeds 70 ppbv are associated with flow from the mainland and likely represent polluted air masses that reach the observing site at Channel Islands

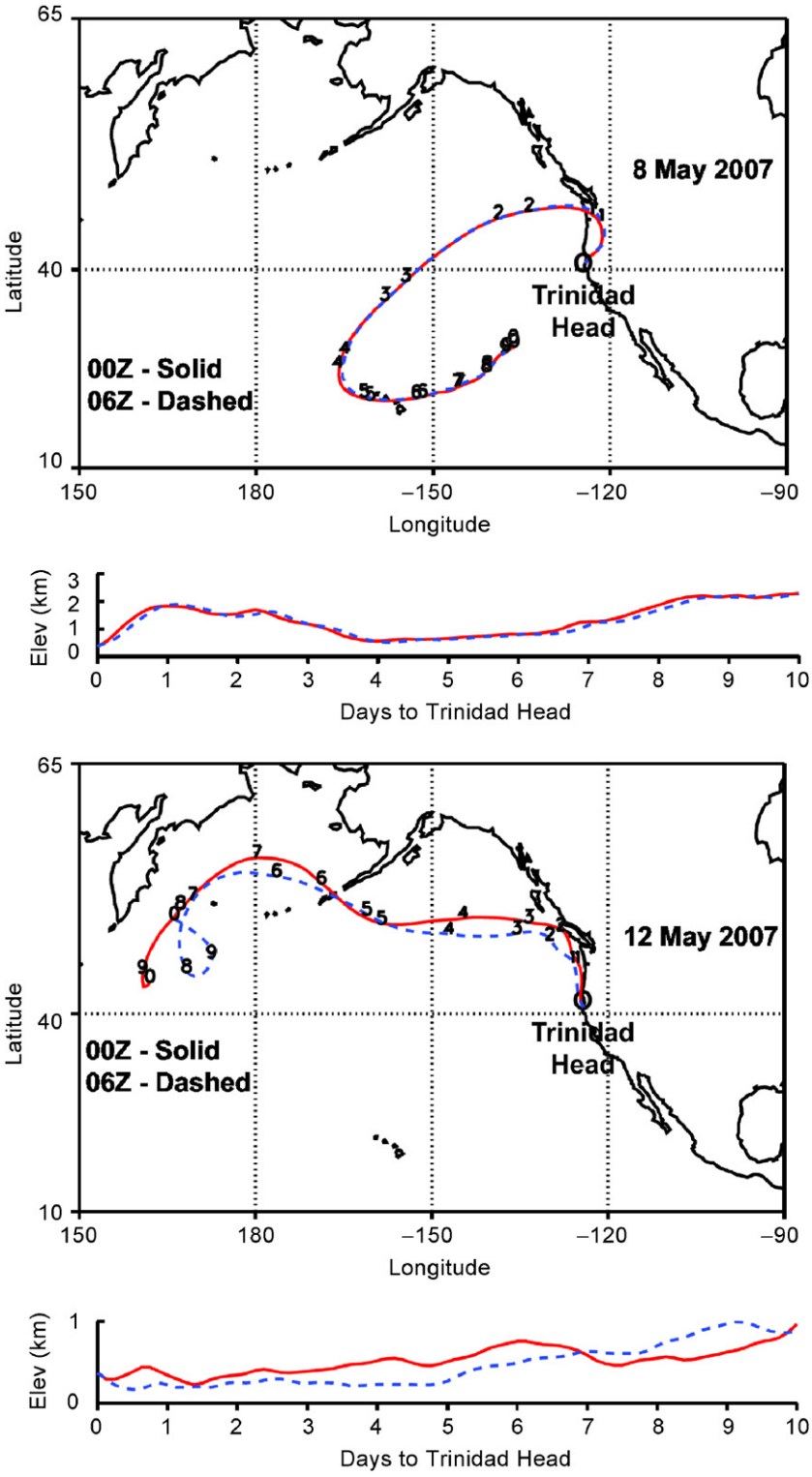

Fig. 10. Back trajectories from Trinidad Head on 8 May 2007 during the period of lower daytime (11–19 LST) ozone levels and on 12 May 2007 on a day when daytime ozone levels were at more typical values.

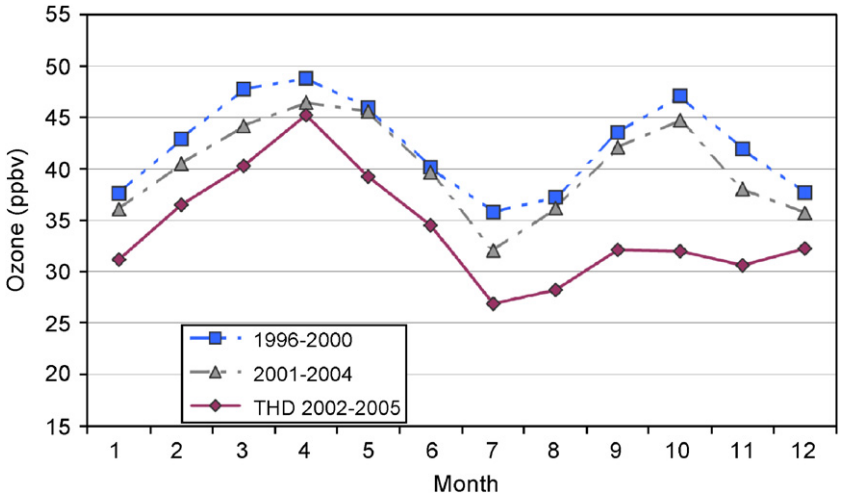

Fig. 11. Comparison of the seasonal cycle for daytime hours (12–18 LST) at Channel Islands NP, California during two periods (1996–2000 and 2001–2004) with the seasonal cycle at Trinidad Head (2002–2005).

NP from the vicinity of Los Angeles. It will also be shown that in the spring the preponderance of cases, when ozone mixing ratios fall in the range of 50–60 ppbv, the air flow, based on back trajectory calculations, has come from over the Pacific Ocean.

4.2. Air flow pattern

The clustered back trajectories to Channel Islands show flow during all seasons that arrives from off the Pacific Ocean (Fig. 12). There is also a tendency for the trajectories to have a significant component from the north with air traveling parallel to the California coast before arriving at Channel Islands. During the autumn months (illustrated by October in Fig. 12), air regularly intercepts the southern California coast before reaching the Channel Islands location. Examination of individual trajectories for ozone values $\geqslant$50 ppbv for a spring (April 2003) and late summer/autumn month (September 2004) shown in Fig. 13 provide a comparison with the behavior at Trinidad Head. During April both locations show air coming dominantly from off the ocean for mixing ratios $\geqslant$50 ppbv. In September, on the other hand, high ozone levels ($\geqslant$50 ppbv) are common at Channel Islands as seen in the number of cases in a single year (September 2004) with the preponderance of cases coming from over land to the east of the observing site (Fig. 13). By contrast, at Trinidad Head there were only 3 days (a total of 18 h) in 5 years for which ozone reached 50 ppbv during September (Table 1). For these cases, trajectories did show air coming from off the land, but as indicated they were infrequent. For the December to June period, the Channel Islands monthly averages are 3–4 ppbv higher than those at Trinidad Head. This reflects higher values at Channel Islands through the entire distribution of ozone amounts, not just in the high values. For the great majority of cases in the winter and spring at Channel Islands, air does not intercept the North American continent when ozone levels are $<$60 ppbv; thus, ozone levels at Channel Islands can be considered representative of background values.

5. Ozone profiles at Trinidad Head

Ozone vertical profiles measurements at Trinidad, California provide additional information on the ozone content of air entering the west coast of the US. They also give a picture of ozone amounts that can mix from above the boundary layer to the surface at Trinidad Head and to the east as air is transported in the westerly flow (trajectories not shown).

5.1. Seasonal variation and the ozone content of the lower free troposphere

The seasonal variation at the surface, as measured by the ozonesondes (Fig. 14), agrees closely with the continuous surface measurements at Trinidad Head (Fig. 3). In the lower troposphere (1–5 km) above the boundary layer, the maximum also occurs in the spring but remains elevated through the summer with a minimum during the winter. Near the surface, there is a distinct summer minimum that may reflect the greater amount of time that air spends in the marine boundary layer (Fig. 5), where under lower NO_X conditions, there is significant photochemical loss (Ayers et al., 1992). The average profiles in the lower troposphere for the months of April–September are shown in Fig. 15, which are the months when highest ozone in the lower troposphere occurs. From the surface to 2 km, the mixing ratio nearly doubles with the steepest gradient between the surface and 1 km and during the summer months when surface values are lowest. At 2 km during all months, the average is $\geqslant$50 ppbv. This suggests that there is a significant reservoir of ozone just above the boundary layer at Trinidad Head that can be mixed down to the surface.

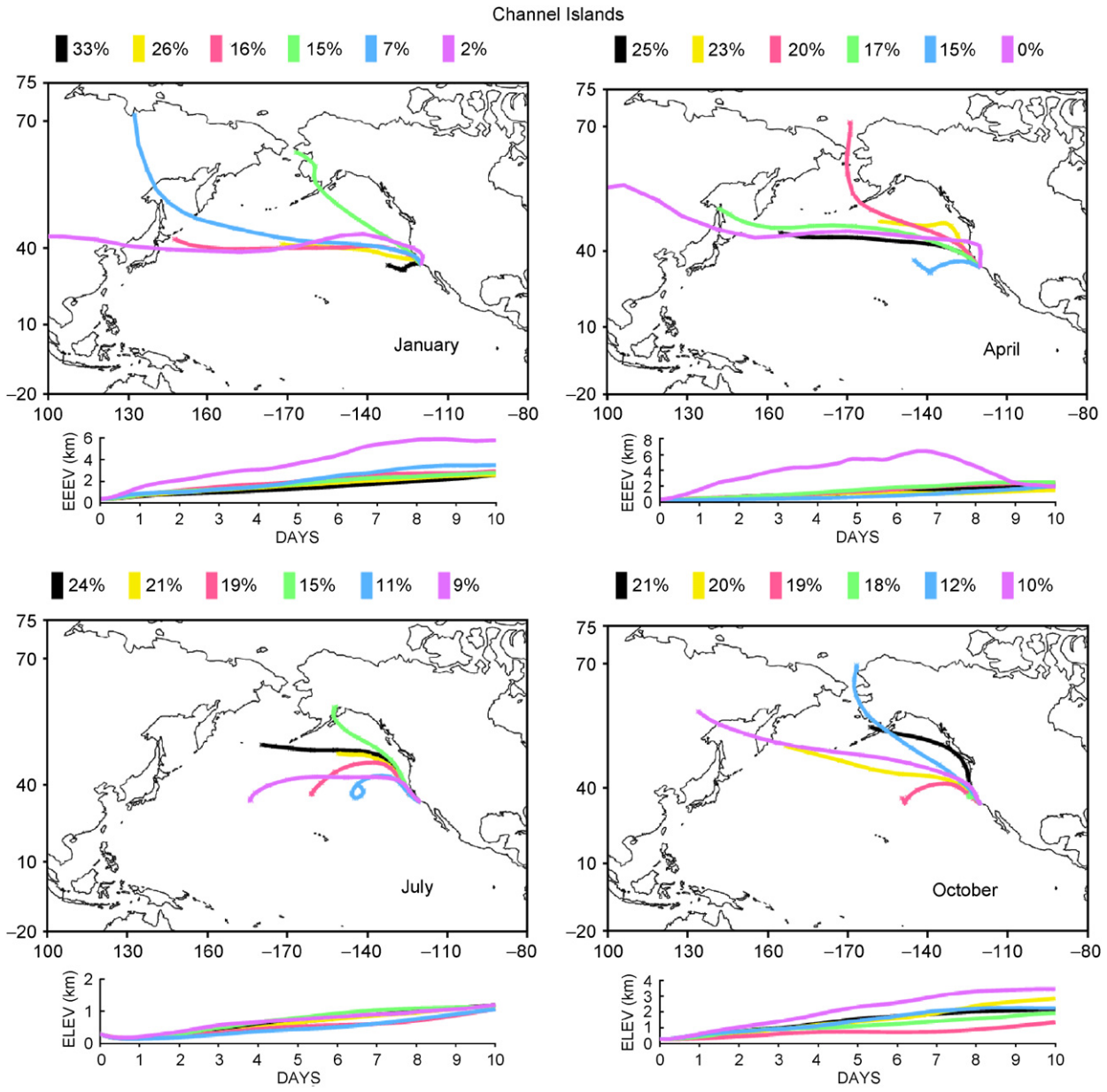

Fig. 12. Clustered back trajectories (1996–2005) for air parcels reaching Channel Islands NP, California at 300 m for January, April, July, and October.

The characteristics of air over the Trinidad Head surface site are illustrated for two periods (April–May 2002 and August 2006), when near daily ozone profiles were obtained (Fig. 16). In the upper troposphere (>5 km), ozone enhancements tend to extend downward from the tropopause region (the tropopause in Fig. 16 is denoted by the black line outlined in white). Below 5 km, the enhancements (i.e., ozone mixing ratios greater than approximately 65 ppbv) are sometimes associated with high ozone amounts in the upper troposphere but also may not be directly linked to ozone above. In both the spring and summer it is clear that there is a high degree of variability in the ozone amount over a several-day period of time and with altitude in layers of varying depth.

5.2. *Longer-term variation*

Although the vertical profile observational period at Trinidad is relatively short, the 10-year record provides an initial opportunity to evaluate possible changes to the ozone content of tropospheric air reaching the west coast of the US. In all layers that are representative of the troposphere (up to approximately 10 km), there are no significant changes (illustrated for the ~1.5–3 km layer in

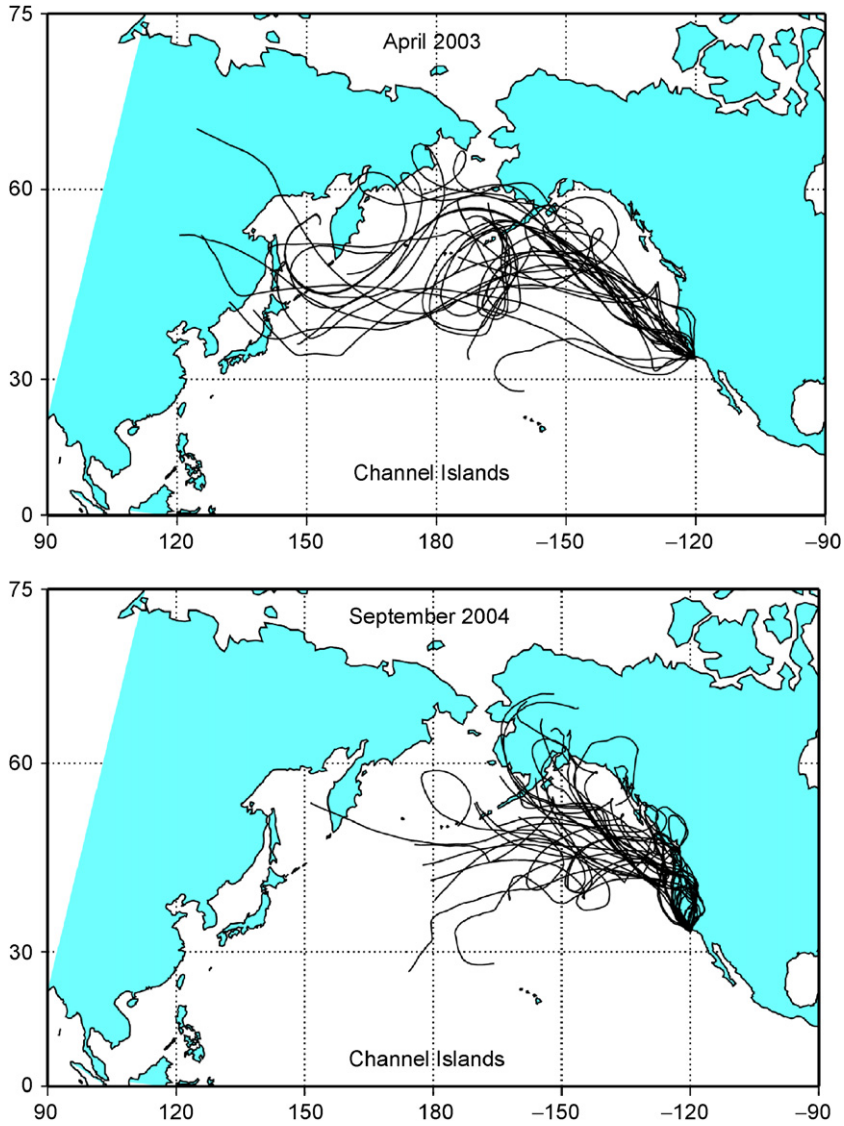

Fig. 13. Back trajectories to Channel Islands for days in April 2003 on which there was an hourly average ozone concentration $\geqslant$50 ppbv but $<$60 ppbv (upper panel) and for September 2004 for all days with an hourly average ozone concentration $\geqslant$50 ppbv (lower panel).

Fig. 17a) over the 10-year record of ozonesonde observations (Fig. 17b).

6. Longer-term surface ozone changes

At the Trinidad Head monitoring site, Fig. 18 shows the 6-year record of surface ozone observations. While there is year-to-year variation in the monthly means, the trend analysis of the distribution of the hourly values for each month shows no significant change for the limited period of surface observations.

At Yreka, California (see location in Fig. 1), there is a 25-year record of surface ozone observations. Although the location is not as optimal as the Trinidad Head site for observing changes in ozone concentrations that may be associated with emissions or climate change, the site does offer a basis of comparison. The Yreka site is in close proximity to a small town (population $<$5000). The spring values based on daytime ozone amounts when the sampled air tends to be well mixed are comparable to Trinidad Head (Fig. 19). However, during the summer and winter the continental location of this site exhibits seasonal amounts that differ from those at Trinidad Head. The monitoring site at Yreka for the period 1981–2006 (2001 was excluded due to measurement problems) for the 1200–1800 LST hours experienced an overall increase of $0.53 \pm .0.13\%\,yr^{-1}$ increase (Fig. 20). This increase is a result of changes during the summer and autumn seasons. The only statistically significant changes in the distribution of the hourly values for individual months were for the months of December, April, and May, with hourly average concentrations in fact decreasing from higher to lower concentration categories (Fig. 21). This at least

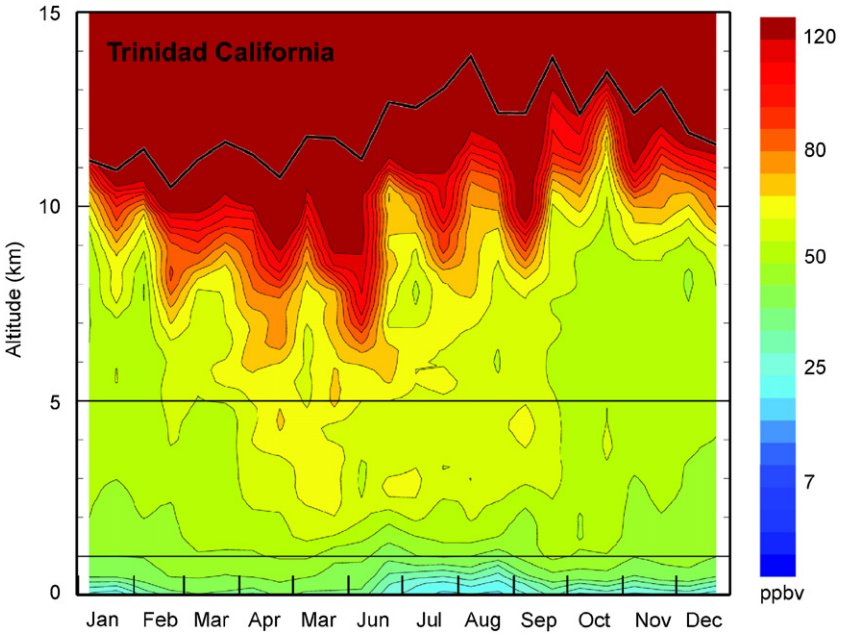

Fig. 14. Cross-section of ozone mixing ratio (ppbv) in the troposphere over Trinidad, California based on 2-week averages of 10 years of ozone soundings (1997–2007). The solid line above 10 km denotes the average thermal tropopause height. The solid lines at 1 and 5 km show the altitude range in the lower troposphere where air coming onshore over northern California is likely to move inland over the North American continent.

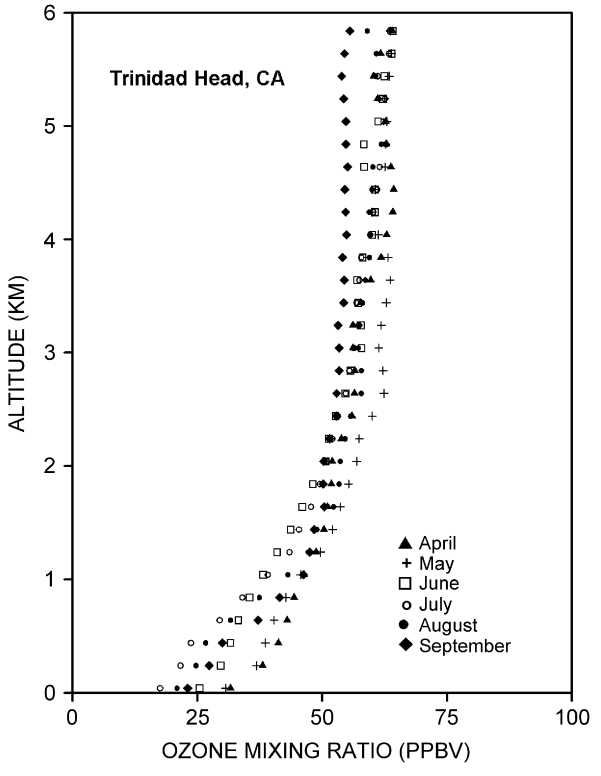

Fig. 15. Average monthly profiles (1997–2007) of ozone mixing ratio (ppbv) at Trinidad, California in the lower troposphere for the 6 months of April to September.

suggests that for the months of strongest transport across the Pacific (winter and spring) for this site there likely has not been significant impact of changing background ozone amounts reaching the west coast of the US.

The monitoring site at Lassen Volcanic National Park experienced a positive statistically significant overall trend of $0.82 \pm 0.14\%\,yr^{-1}$ in daytime ozone concentrations for the period 1987–2006 (Fig. 20). For hourly average concentrations all months for the period 1987–2006 based on the analysis of the 24 h values for each day showed significant increases with shifts to higher concentration bins. If the analysis is limited to daytime hours (12–18 LST) only, December, January, March, April, June, and July show shifts from the lower concentration bins to the higher levels (Fig. 22). In almost every case concentrations moved from bins <50 to $\geqslant 50$ ppb both when 24 h values were considered (hence for all months) or for the more limited months where there were significant increases in the daytime hours. Changes across multiple seasons suggest that more than one set of processes is responsible for the change.

7. Discussion and conclusions

Several investigators have proposed that the effect of Asian emissions on surface ozone concentrations might be responsible for enhancing ozone levels in the US (e.g., Jaffe et al., 2003; Parrish et al., 2004; Jaffe and Ray, 2007). For example, using a 15-year record of ozone from Lassen Volcanic NP and data from two aircraft campaigns, plus

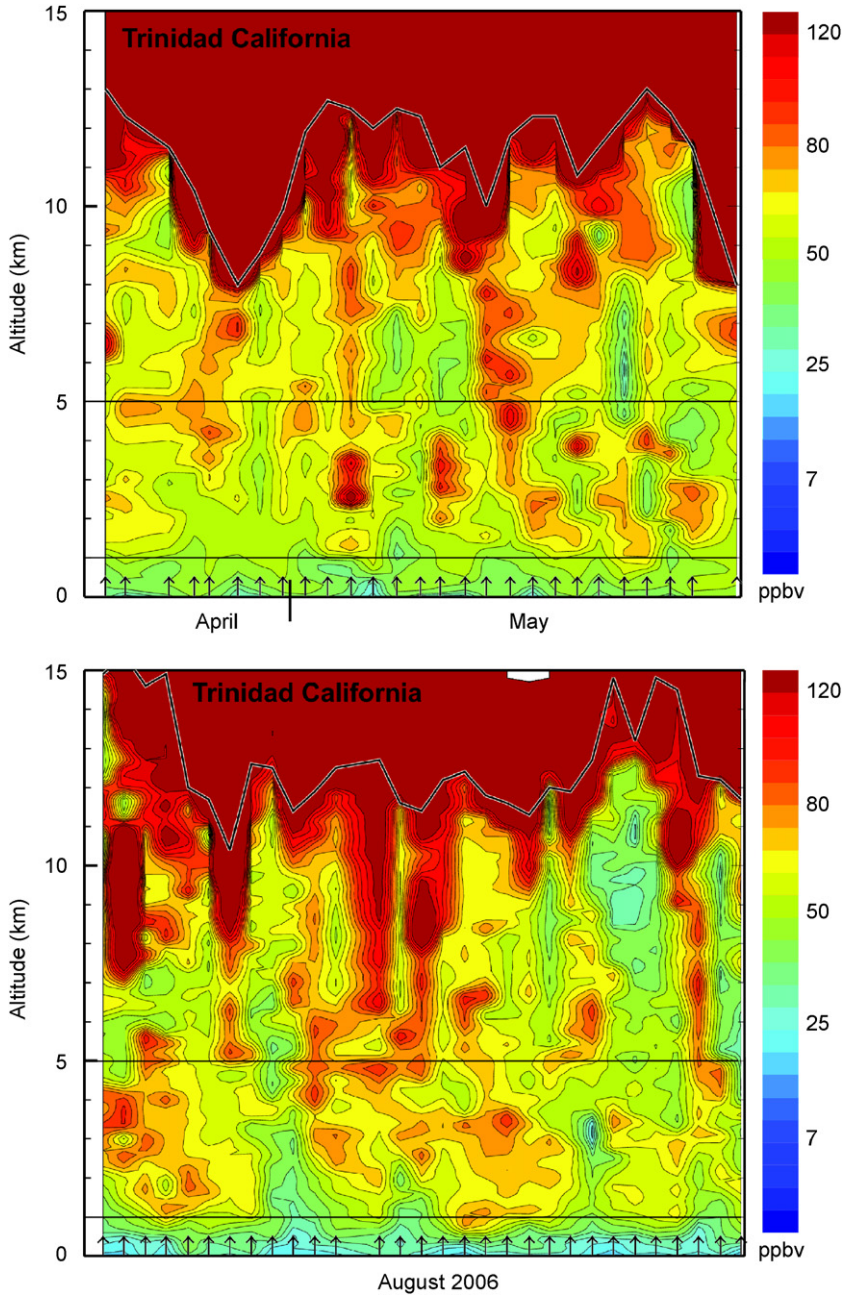

Fig. 16. Cross-section of daily ozone mixing ratio profiles for two periods (April–May 2002 and August 2006) with daily ozonesonde measurements. The thin black line with white outline connects the thermal tropopause determined from the individual sounding.

observations spanning 18 years from five US west coast marine boundary layer sites, Jaffe et al. (2003) estimated that the amount of ozone in air arriving from the Eastern Pacific in spring has increased by approximately 10 ppb from the mid-1980s to the present.

The monitor in Lassen Volcanic National Park in northern California is located in the northwest corner of the park, away from any major emission sources or urban centers. However, the monitor is located approximately 240 km inland from the Pacific coast. The nearest city is Redding, California, 70 km to the west. Jaffe et al. (2003) noted that because of prevailing westerly winds, most trajectories arrive at Lassen out of the west. Jaffe et al. (2003) reported that trends were statistically significant in all seasons. Although acknowledging that trends during the summertime were associated with photochemistry, the authors were not certain whether the trends in the other seasons reflected changes in background ozone or changes in regional photochemistry.

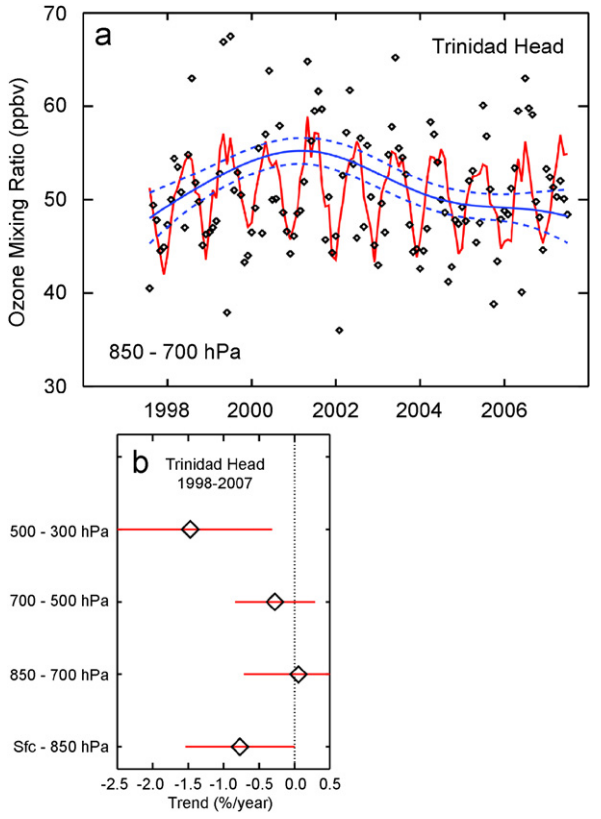

Fig. 17. (a) Monthly average ozone mixing ratio (diamonds) over Trinidad Head, California in the 850–700 hPa layer (~1.5–3 km). The red (thin solid) line is the model fit to the monthly means, the blue (thick, smooth solid) line is a fit to the residuals (difference between the model fit and the observed monthly values with the overall mean added back to the curve fit to allow plotting on the same scale), and the blue dashed lines are the 95% confidence interval of the fit (Harris et al., 2001). See the discussion of the method and the terminology in the text (Section 2). (b) Summary of the trend in $\%\,yr^{-1}$ in four tropospheric layers at Trinidad Head. Designation of the curves for B/W is given in parenthesis.

Hourly average concentrations during April and May at the Lassen National Park monitoring site (Table 2) are greater than those occurring at Trinidad Head (Table 1) suggesting that air has been modified as it has moved inland. Table 3 summarizes the maximum hourly average concentrations experienced at a monitoring site at Redding CA, west (Fig. 1) of the Lassen NP monitoring site, during the 1999–2007 period. It appears that local photochemical production may be very important in the Redding, CA area during the springtime. Elevated hourly average concentration levels of ozone occur both to the west (i.e., Redding, CA and Anderson, CA) and to the south (Chico, CA; Tuscan, CA; Red Bluff, CA) of the park site. As illustrated from the hourly average data measured in Redding, levels in the 0.09–0.10 ppm range occur during the April–May period at these locations and well above 0.10 ppm during the summertime. Lassen Volcanic National Park site may thus be influenced by the transport of ozone resulting from anthropogenic emissions within the regional area during the spring as well as summer months.

Although the exposure of the Yreka sampling site is likely not ideal since it is inland from the coast and not elevated above the surrounding terrain, the 25-year record of measurements makes this the one of the longest records available for investigating long-term changes. The location is not downwind of major emission sources when the site is under the influence of the broad scale flow, which is generally from the west during the spring, based on the 10-year climatology of back trajectories (not shown).

The detailed analysis of ozone measurements at the Trinidad Head CA site, that appears to be optimally located for sampling background air reaching the west coast of the US, shows that during the spring hourly surface ozone levels can equal or exceed 50 ppbv, which occur almost exclusively with air flow from directly off the Pacific. Due to the enhanced hourly average concentrations, mean daytime ozone mixing ratios during the

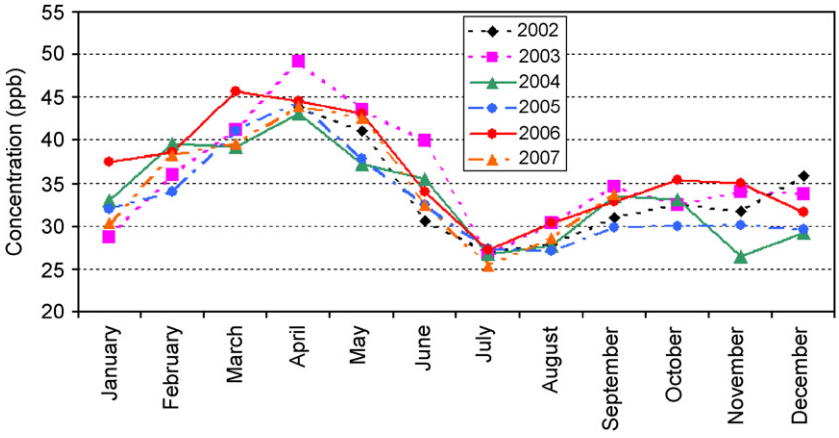

Fig. 18. Monthly average daytime (12–18 LST) ozone mixing ratios for 2002–2007 at Trinidad Head, California.

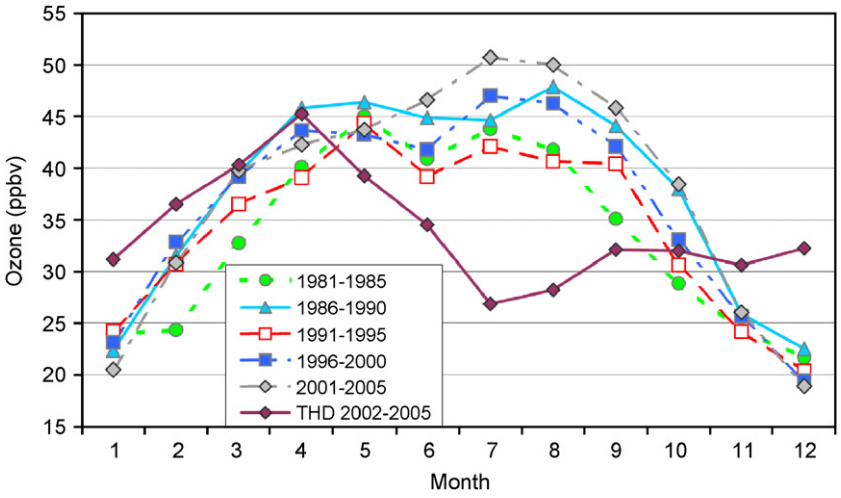

Fig. 19. Monthly ozone mixing ratios based on daytime hourly values (12–18 LST) for 5-year periods at Yreka (1981–1985, 1986–1990, 1991–1995, 1996–2000, 2001–2005) and Trinidad Head (2002–2005), California.

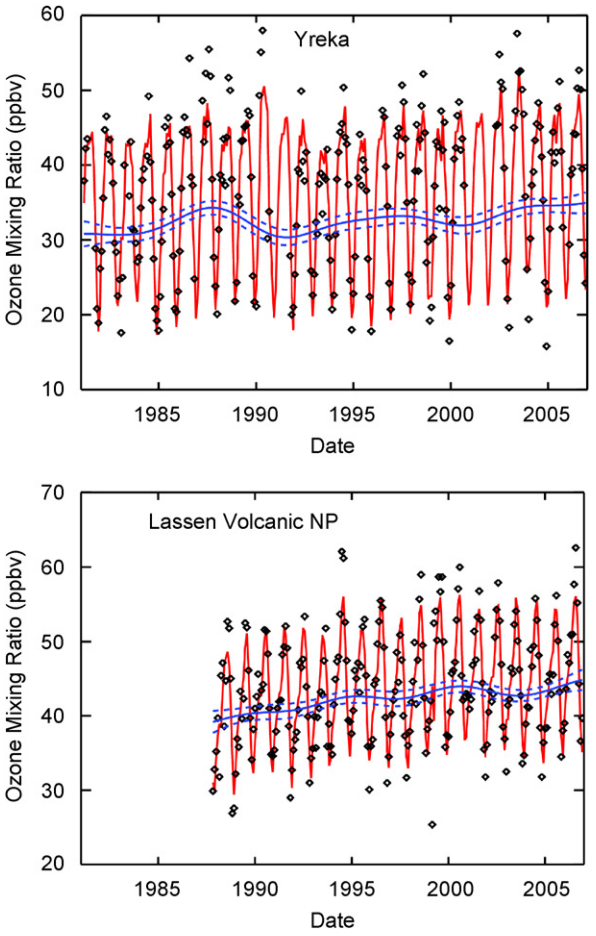

Fig. 20. Monthly mean daytime (12–18 LST) surface ozone mixing ratios (diamonds), model fit (red–thin solid) and long-term variation (blue–thick, smooth solid) as in Fig. 17a at Yreka, California (upper panel) and Lassen Volcanic NP (lower panel). Designation of the curves for B/W is given in parenthesis.

spring (March–April–May) exceed 40 ppbv implying that in this season ozone reaching the US west coast makes a significant contribution to background ozone levels over the US. Although a more complicated site than Trinidad Head for assessing background surface ozone levels, Channel Islands NP, located off the coast of southern California, also appears to be able to provide information on background ozone entering the US. During the winter and spring, surface ozone characteristics at Channel Islands strongly resemble those at Trinidad Head. This is consistent with the dominant air flow reaching this site, which is also over the Pacific Ocean. With a significant northerly component to air parcels arriving at this site in the spring, air from similar latitudes to those sampled at Trinidad Head is seen at Channel Islands. Median ozone values at Channel Islands during the spring exceed those at Trinidad Head by ~3 ppbv; however, the period of measurements overlap for only 3 years. In a few cases, high hourly average ozone levels (>60 ppb) in the spring at Channel Islands are associated with air coming off the land over southern California and do not represent background levels. Analysis of the Channel Islands surface ozone record strongly indicates that, as at Trinidad Head, hourly ozone mixing ratios in the spring of ⩾50 ppbv are associated with air that is not influenced by the North American continent, and the average daytime ozone levels in the spring under background conditions exceed 40 ppbv.

The 10-year record of ozone vertical profile measurements at Trinidad Head provides a picture of the ozone content of air entering the western boundary of the US. In the troposphere above the boundary layer, ozone levels do not show the strong seasonal decrease during the summer seen in the surface measurements. Average ozone mixing ratios near 50–60 ppbv are typical in the 2–6-km layer throughout the spring and summer. There has also been no significant change in tropospheric ozone above Trinidad Head over the 10 years of ozonesonde measurements and specifically during the spring season.

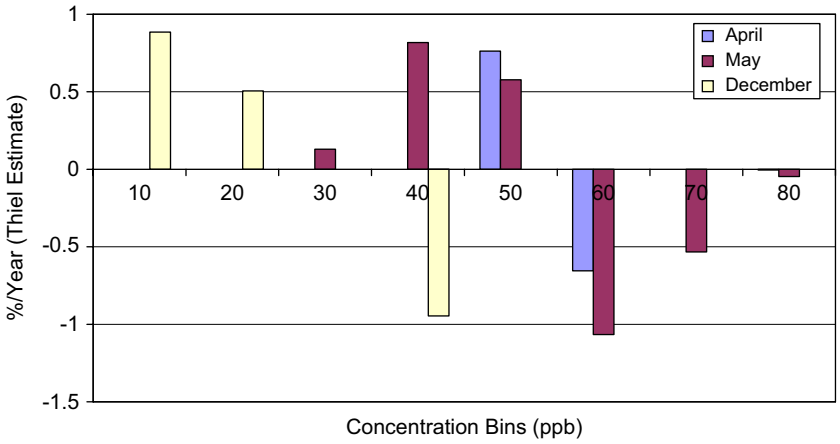

Fig. 21. Distribution of changes by month for Yreka, California (1982–2006) for the months with significant changes based on daytime (12–18 LST) values.

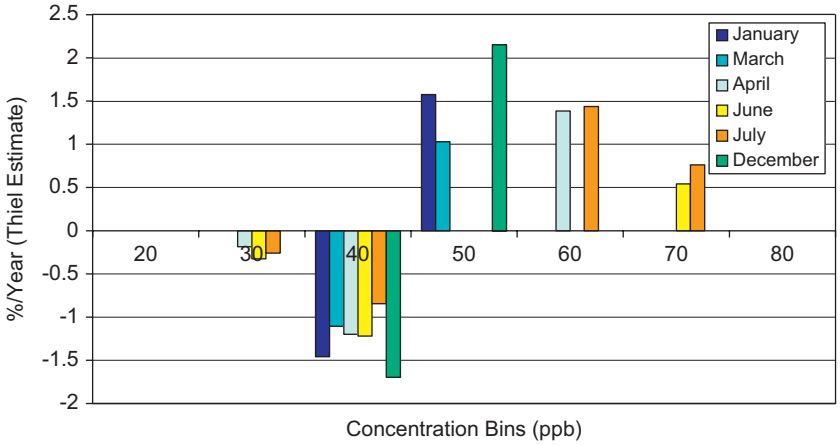

Fig. 22. Distribution of changes by month for Lassen Volcanic NP, California (1987–2006) for the months with significant changes based on daytime (12–18 LST) values.

Table 2
Maximum hourly average concentrations (ppmv) during spring measured at Lassen Volcanic National Park (AQS Site 060893003—40.5°N, 121.5°W) for the period 1988–2007

Year	March	April	May
1988	0.056	0.067	0.089
1989	0.050	0.072	0.067
1990	0.065	0.071	0.069
1991	0.064	0.062	0.060
1992	0.056	0.056	0.071
1993	0.051	0.058	0.084
1994	0.062	0.085	0.082
1995	0.062	0.066	0.078
1996	0.062	0.066	0.073
1997	0.058	0.072	0.072
1998	0.066	0.078	0.071
1999	0.050	0.085	0.082
2000	0.063	0.066	0.071
2001	0.056	0.068	0.078
2002	0.063	0.058	0.070
2003	0.061	0.063	0.069
2004	0.068	0.063	0.065
2005	0.059	0.077	0.077
2006	0.063	0.070	0.073
2007	0.065	0.069	0.077

Source: US EPA AQS Database.

Table 3
Maximum hourly average concentrations (ppmv) during spring measured at Redding, CA (AQS Site 060890004—40.6°N, 122.4°W) for the period 1999–2007

Year	March	April	May
1999	0.050	0.101	0.096
2000	0.072	0.073	0.094
2001	0.046	0.054	0.077
2002	0.044	0.060	0.079
2003	0.038	0.043	0.NA
2004	0.058	0.075	0.074
2005	0.064	0.060	0.089
2006	0.049	0.056	0.076
2007	0.059	0.069	0.077

Source: US EPA AQS Database.

Acknowledgments

Michael Ives of the Marine Sciences Laboratory of Humboldt State University has overseen both surface and ozone profile measurements at Trinidad Head. Funding for the ozonesonde measurements at Trinidad Head was provided through the NOAA Health of the Atmosphere program.

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