

Increasing background ozone during spring on the west coast of North America

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[1] Using a 15-year record of O₃ from Lassen Volcanic National Park, a rural elevated site in northern California, data from two aircraft campaigns conducted in 1984 and 2002 over the eastern North Pacific, and observations spanning 18 years from five U.S. west coast, marine boundary layer sites, we show that O₃ in air arriving from the Eastern Pacific in spring has increased by approximately 10 ppbv, i.e. 30% from the mid 1980s to the present. This positive trend in O₃ correlates with the increasing trend in global nitrogen oxide emissions, which is especially pronounced in Asia. As spring is the season of strongest transport of Asian emissions to the Pacific, we conclude that the emission trend is the most likely cause of the O₃ trend. *INDEX TERMS:* 0345 Atmospheric Composition and Structure: Pollution—urban and regional (0305); 0365 Atmospheric Composition and Structure: Troposphere—composition and chemistry; 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry. **Citation:** Jaffe, D., H. Price, D. Parrish, A. Goldstein, and J. Harris, Increasing background ozone during spring on the west coast of North America, *Geophys. Res. Lett.*, 30(12), 1613, doi:10.1029/2003GL017024, 2003.

1. Introduction

[2] It is now clear that large increases in anthropogenic emissions of the photochemical precursors to O₃ (particularly NO_x = NO + NO₂) have caused substantial increases in the global background O₃ mixing ratios over the past century [Marenco *et al.*, 1994]. Elevated levels are a serious concern due to the effects of O₃ on the respiratory system and vegetation damage at concentrations not far above ambient. In East Asia, NO_x emissions have grown by approximately 4%–6% per year over the past two decades [Akimoto and Narita, 1994; Streets *et al.*, 2001]. The increase in NO_x emission is largest in China and appears to have continued steadily through at least 1997 [Streets *et al.*, 2001].

[3] Several researchers have reported trends in tropospheric O₃ over Japan using ozonesonde data. Three sites,

Kagoshima, Tsukuba and Sapporo, have data since 1969 that show significant positive trends of 1.5–2.5%/year in the lower troposphere up to about 1990. In the past decade the reported trend, while still generally positive, has slowed [Oltmans *et al.*, 1998; Logan *et al.*, 1999]. Evaluation of data at Okinawa from 1989–1997 indicates an O₃ increase of 2.5%/year in Asian continental air during the winter-spring period [Lee *et al.*, 1998].

[4] While it has been known for some time that Asian desert dust is transported far out into the Pacific by the prevailing westerly winds [Duce *et al.*, 1980], it has also been shown that primary emissions and O₃ are transported at least as far as North America by the same mechanism [Parrish *et al.*, 1992; Jaffe *et al.*, 1999, 2003].

[5] In this work, we seek to quantify the recent trend in springtime background O₃ mixing ratios at the west coast of North America. Our focus is on the spring, since this is the season with the strongest outflow of Asian emissions to the Pacific and because it is the season with the most extensive observational database. For this work, we define “background air” to mean air from the Pacific with O₃ levels not significantly influenced by North American emissions within the previous three days.

2. Methods

[6] Most observations of O₃ in North America are made in urban and rural environments with substantial local perturbations to observed O₃ levels. However to examine trends in background O₃, measurements free from significant North American influence over decade time scales are required. Here we identify and examine the few such data sets that are available from surface and aircraft measurements along the U.S. West Coast.

[7] Table 1 summarizes these observations. The U.S. National Park Service has monitored ambient O₃ at three relatively remote locations (Lassen Volcanic National Park, Redwood National Park and Point Reyes National Seashore) on or near the West Coast of California that are not greatly influenced by local emissions. Each site also has coincident meteorological measurements for selection of marine air masses. Three intensive research campaigns have

Table 1. Springtime Data Sets Utilized in the Present Analysis

Site/Study	Dates	Lat./Long.	Elevation (m)	Wind selection ^a	% Data	Data Source
CITE-1C	April–May, 1984	30°–40°N 105°–135°W	0–8000	See text	See text	NASA GTE
Point Arena, CA	April 24–May 5, 1985	38°57′N 123°44′W	20	WD:225–360 WS: >3m/s	61	<i>Parrish et al.</i> [1992]
Lassen Volcanic N.P., CA	1988 to present	40°32′N 121°35′	1756	WD:190–360 WS: >3m/s	26	NPS-ARD ^b
Redwood N.P.	1988 to 1995	41°34′N 124°05′W	235	WD:225–360 WS: >3m/s	13	NPS-ARD ^b
Point Reyes N.S.	1988 to 1992	38°07′N 122°55′W	76	WD:225–360 WS: >3m/s	49	NPS-ARD ^b
Cheeka Peak, WA	Springtime, 1987 to present	48°18′N 124°36′W	480	WD:150–300 WS: >3m/s	51	<i>Jaffe et al.</i> [2001] and this work
Trinidad Head, CA	April–May, 2002	41°03′N 124°09′W	100	WD:225–360 WS: >3m/s	43	This work
ITCT 2K2	April–May, 2002	30°–40° 105°–135°W	0–8000	See text	See text	This work

^aWD refers to wind direction and WS refers to wind speed.

^bNational Park Service-Air Resources Division.

provided springtime, surface measurements along the West Coast at Point Arena, CA, Cheeka Peak, WA and Trinidad Head, CA. Two aircraft research campaigns have been conducted along the West Coast, also in spring - The NASA GTE CITE-1C study in 1984 [*Chameides et al.*, 1989] and the ITCT 2K2 study in 2002. Our approach here is to objectively select the measurements from these eight data sets that best represent background O₃ levels, and to quantify the long-term temporal trend.

[8] All O₃ measurements from the surface sites were made using standard UV analyzers. The National Park Service employed a consistent calibration procedure based on U.S. EPA methods. The intensive research campaigns have followed careful data validation procedures. The aircraft campaigns made high frequency measurements with chemiluminescence instruments that were calibrated against standard UV instruments. All of the data sets are believed to be accurate to $\pm 3\%$. The precision of the data points used in obtaining the averages discussed here is ± 1 ppbv. For all ground sites, we used hourly average ozone mixing ratios for our analysis.

[9] We have identified only one site, Lassen Volcanic National Park (hereafter referred to as “Lassen”) in Northern California, with a long-term (1988–present), continuous record of O₃ measurements. The monitoring station is located in the northwest corner of the park, away from any major emission sources or urban centers. However, it does lie approximately 240 km inland from the Pacific coast. The nearest city is Redding, California (1995 population of 79,000), 70 km to the west. This inland location requires close investigation of the possible effects of North American influences on observed O₃ levels. The five other surface sites all lie within a few km of the coast. Although none of them provides a continuous record of more than 8 years, the combination of measurements from these sites constitutes a discontinuous, 18-year record of springtime, background O₃ in the coastal marine boundary layer (MBL). Finally, the two aircraft studies, conducted 18 years apart, provide regional characterizations of O₃ in

the springtime lower troposphere in the eastern North Pacific.

[10] All of the measurement sites are affected to some degree by local influences including surface deposition (most importantly to vegetation), removal of O₃ by reaction with local emissions of NO, and photochemical production of O₃ from precursors emitted over North America. For each of the surface sites, we used objective criteria to select time periods characteristic of background O₃ using local wind speed and direction, as shown in Table 1. For the Pt. Arena

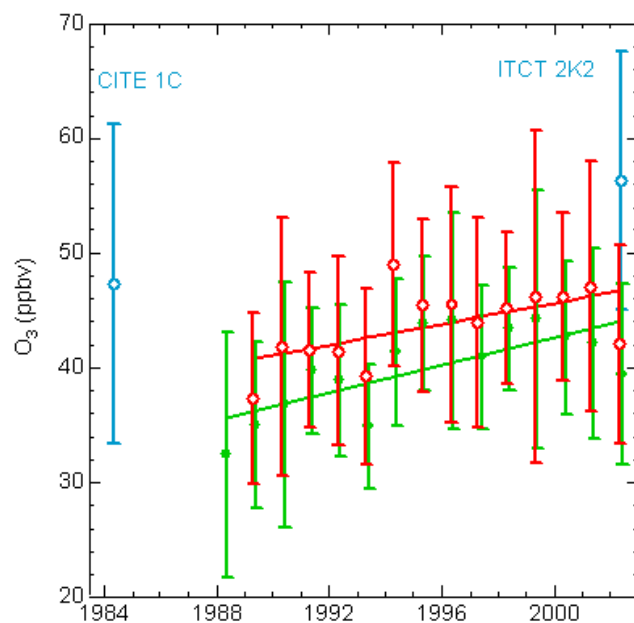


Figure 1. Spring mean mixing ratio ± 1 standard deviation for background O₃ at Lassen Volcanic NP using the marine trajectory dataset (green) and the wind selected dataset (red). Also shown is data from the CITE 1C and ITCT aircraft campaigns.

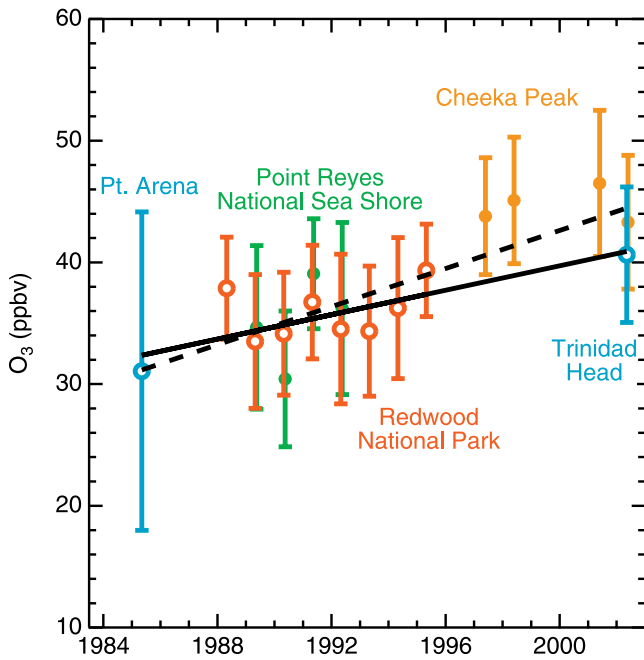


Figure 2. Spring mean mixing ratio ± 1 standard deviation for background O_3 at 5 MBL sites with linear regression lines. The data have been selected by local wind direction and speed to minimize local influences (see Table 1 for details). The linear fit to the data from the 4 sea level sites (solid line), yields a slope and year 2000 O_3 mixing ratio (with 95% confidence intervals) of 0.50 ± 0.36 ppbv/year, 39.9 ± 3.3 ppbv, and an r^2 of 0.44. If the higher altitude and latitude Cheeka Peak site is included (dashed line), the calculated slope and year 2000 mixing ratio become 0.78 ± 0.28 ppbv/year, 42.9 ± 2.4 ppbv, and the r^2 is 0.68.

and Cheeka Peak sites, we have shown previously that this wind selection effectively removes recent influence from the North American continent [Parrish et al., 1992; Jaffe et al., 2001]. Based on observations of several continental tracers, wind selection is also effective for the Trinidad Head data.

[11] For Lassen, we also used isentropic back-trajectories to segregate the data, since trajectories reflect the air parcel history in a way that local wind speed and direction cannot. We computed 10-day isentropic back-trajectories twice each day (0 and 12 GMT) during all seasons for the entire period of the data record (1988–2002) using the European Center for Medium range Weather Forecasting (ECMWF) data and previously published methods [Harris et al., 1992]. Because of prevailing westerly winds, most trajectories arrive at Lassen from the North Pacific. An air mass that took longer than 24 hours to reach the site from the Pacific coast

(defined by $125^\circ W$ longitude) or crossed over California south of $39^\circ N$ latitude was classified as “local”. An air mass that arrived at the Lassen site having spent less than 24 hours over land was classified as “marine”. Each air mass type was associated with a 12-hour average O_3 concentration, centered on either 0 or 12 GMT. For spring, summer, fall and winter, the marine dataset contains 39, 28, 31, and 33% of the full dataset. (For this paper, we define the seasons as the three-month periods beginning on the first of March, June, September and December.)

[12] For the two aircraft studies we selected data for the same time of the year (flights from April 20 to May 11), same region (30° to $40^\circ N$ latitude and 105° to $135^\circ W$ longitude), and same altitude range (from 0–8 km over the ocean and 2–8 km over land). In addition, we eliminated data with carbon monoxide levels above 150 ppbv (to avoid any strong transport of North American emissions to the flight altitude) or O_3 levels above 100 ppbv (to avoid influence from strong stratospheric intrusions).

3. Results

[13] Figure 1 shows the springtime, background O_3 mixing ratios measured at Lassen from 1988–2002 and the results from the two aircraft campaigns. Figure 2 shows the results from five MBL sites along the Pacific coast. Table 2 summarizes the trend analysis for the Lassen site. The analysis is done separately for the trajectory-selected, wind-selected, and full data sets for each season. In spring, a consistent, statistically significant, positive trend in background O_3 mixing ratios is found, regardless of which data set is used. The Lassen and the other MBL datasets all yield a consistent positive trend in background O_3 of approximately 0.5–0.8 ppbv/year along the Pacific Coast. While the aircraft mixing ratios are higher than the surface values, the increase from 1994 to 2002, 0.51 ppbv/year, is consistent with the surface trend.

[14] The spring background O_3 levels have a significant altitude dependence that is reflected in the year 2000 O_3 mixing ratios derived from the linear fits in Figures 1 and 2. The values increase from 40 ppbv at sea level to 46 ppbv (43 ppbv using trajectory-selected data) at Lassen (1.8 km) to approximately 55 ppbv for the 0 to 8 km altitudes covered by the aircraft studies. The mean background O_3 mixing ratios at Cheeka Peak, the highest altitude and highest latitude MBL site, are significantly higher than the other MBL sites. We calculated the trend both with and without the Cheeka Peak data, as shown in Figure 2. The linear regression fit is statistically significant in either case. Importantly, the trend determined from the four sea-level MBL locations (not including Cheeka Peak) is statistically significant, even if the data from any one of these sites is excluded.

Table 2. Trends for Lassen O_3 Data Using Full Dataset, Marine Trajectories Only, and the Wind-Selected Datasets

	Marine trajectory dataset			Wind selected dataset			Full dataset		
	Slope (ppbv/yr)	Year 2000 O_3 (ppbv)	r^2	Slope (ppbv/yr)	Year 2000 O_3 (ppbv)	r^2	Slope (ppbv/yr)	Year 2000 O_3 (ppbv)	r^2
Spring	0.60 ± 0.35	43.0 ± 2.3	0.51	0.46 ± 0.39	45.8 ± 2.4	0.36	0.60 ± 0.30	43.0 ± 2.0	0.58
Summer	0.79 ± 0.42	45.5 ± 2.7	0.56	0.69 ± 0.48	55.2 ± 3.2	0.42	0.69 ± 0.38	46.1 ± 2.5	0.54
Fall	0.45 ± 0.36	36.9 ± 2.5	0.38	0.16 ± 0.52	44.8 ± 3.6	0.04	0.49 ± 0.38	39.7 ± 2.6	0.40
Winter	0.38 ± 0.34	38.4 ± 2.3	0.31	0.33 ± 0.38	40.4 ± 2.6	0.23	0.40 ± 0.28	38.9 ± 1.9	0.42

Full and marine trajectory analysis extends from January 1988 to August 2002. Wind selected analysis begins in June 1988, when wind data became available. Uncertainties are given as 95% confidence intervals.

[15] Even though we have selected the data most representative of background air, diurnal cycles in the O₃ mixing ratios are observed at some of these sites. These cycles are attributed to a combination of in-situ photochemical O₃ production in daytime and surface deposition of O₃ in a shallower boundary layer at night. In spring, the peak-to-peak magnitude of the diurnal cycle at all sites is relatively small, between 4 and 7 ppbv. It is important to note that the long-term trends at Lassen are present in both daytime and nighttime data.

[16] The mean background O₃ levels along the west coast of the U.S. are spatially quite uniform during spring. For the four years with data from both Cheeka Peak and Lassen (separated by nearly 900 km and Lassen located well inland), the mean ratio of the wind-selected background mixing ratios is 1.00, with a standard deviation of 0.02. Similarly, for the four years with data from both Point Reyes NS and Redwood NP, (coastal sites separated by 400 km) the mean ratio of the derived values is 1.01 with a standard deviation of 0.08. Interannual variations are strongly correlated in both site comparisons: $r^2 = 0.97$ for Cheeka Peak-Lassen and 0.53 for Point Reyes-Redwood. This spatial homogeneity indicates that we are indeed successful in quantifying the springtime background O₃ levels characteristic of eastern North Pacific air masses reaching the U.S. west coast.

[17] Table 2 also presents the trend analysis for summer, fall and winter for the Lassen data set. Positive trends in O₃ mixing ratios are found in all seasons. Using the full data set, the trends are statistically significant in all seasons, and they are statistically significant in summer regardless of which data set is used. Whether the trends in other seasons reflect changes in background O₃ or changes in regional photochemistry is not certain. In particular, O₃ levels at Lassen in summer are clearly affected by local in-situ production to a much greater extent than in spring. This is indicated by the larger diurnal cycle in the wind-selected data in summer (21 ppbv peak-to-peak) compared to spring (6 ppbv). While the summer trend is interesting, at this time there is insufficient data to ascertain the exact cause.

[18] Several arguments indicate it is unlikely that local photochemical production of O₃ has made a significant contribution to the positive temporal trend in springtime background O₃. First, we have selected measurements from sites and meteorological conditions that minimize influence from North American emissions. Second, photochemical production of O₃ is slow in Northern California in spring as evidenced by the small diurnal cycles in observed O₃ levels. Third, the trend is uniform at all sites including the aircraft data that probed well out over the Pacific Ocean. Fourth, tabulated inventories (available online at <http://www.arb.ca.gov>) [Alexis et al., 2001] suggest that emissions of O₃ precursors have decreased significantly in the state of California over the past 15 years. Based on this analysis, we conclude that background O₃ in the eastern North Pacific during spring has increased.

4. Conclusion

[19] Over the past eighteen years the springtime background O₃ mixing ratios have increased by approximately 10 ppbv, i.e. 30%, along the U.S. west coast. This positive trend in the background O₃ mixing ratio is most likely due

to changes in emissions of O₃ precursors in Asia. This result is similar to the pattern seen over Europe, where background O₃ increased during the period when U.S. NO_x emissions were rising [Naja et al., 2003]. As energy use grows in developing regions of the world, atmospheric emissions of O₃ precursors will likely increase as well. These results provide new evidence that air pollution problems are not restricted by national boundaries, and that global cooperation will be required to address many critical environmental problems in the future.

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