AGU PUBLICATIONS

Geophysical Research Letters

RESEARCH LETTER
10.1002/2017GL074960

Key Points:
- Over the past decades, a long-term increase in baseline ozone has been observed at the North American West Coast; that increase has ended
- The end of increasing ozone concentrations transported into the U.S. eases one difficulty to meeting the U.S. ozone air quality standard
- Global models poorly reproduce observed baseline ozone; they do not accurately calculate North American background ozone and its trends

Supporting Information:
- Supporting Information S1

Correspondence to:
D. D. Parrish,
david.d.parrish@noaa.gov

Citation:
Parrish, D. D., Petropavlovskikh, I., & Oltmans, S. J. (2017). Reversal of long-term trend in baseline ozone concentrations at the North American West Coast. *Geophysical Research Letters, 44*, 10,675–10,681. https://doi.org/10.1002/2017GL074960

Received 14 JUL 2017
Accepted 24 SEP 2017
Accepted article online 9 OCT 2017
Published online 18 OCT 2017

Reversal of Long-Term Trend in Baseline Ozone Concentrations at the North American West Coast

D. D. Parrish[1,2], I. Petropavlovskikh[1,3], and S. J. Oltmans[1,3]

[1]Cooperative Institute for Research in Environmental Sciences, University of Colorado Boulder, Boulder, CO, USA, [2]NOAA ESRL Chemical Sciences Division, Boulder, CO, USA, [3]NOAA ESRL Global Monitoring Division, Boulder, CO, USA

Abstract Changes in baseline (here understood as representative of continental to hemispheric scales) tropospheric ozone concentrations that have occurred over western North American and eastern North Pacific are analyzed based on data from three measurement records: (1) sites in the U.S. Pacific coast marine boundary layer, (2) an inland, higher altitude site at Lassen Volcanic National Park, CA, and (3) springtime airborne measurements in the free troposphere between 3 and 8 km altitude. Consistent with previously published results, we find increasing ozone prior to the year 2000, but that rate of increase has slowed and now reversed in these data sets in all seasons. The past ozone increase has been identified as a significant difficulty to overcome in achieving U.S. air quality goals; this difficulty has now eased. Global models only poorly reproduce the observed baseline ozone and trends; policy guidance from such models must be considered very cautiously.

Plain Language Summary In U.S. urban and rural areas, ozone transported into the country from the Pacific (i.e., baseline ozone) makes substantial contributions to exceedances of the ozone national ambient air quality standard. Over past decades, baseline ozone concentrations increased, which made achievement of ozone air quality goals increasingly difficult. However, that increase ended in the early to mid-2000s, and the baseline concentrations have begun to decrease, thus easing this particular difficulty. Global modes are relied upon to quantify the influence of this ozone transport, but they poorly reproduce observed baseline ozone concentrations.

1. Introduction

Ozone is a secondary pollutant that, in the U.S., is subject to control under the 1970 Clean Air Act. In response to increasing evidence for human health effects from ozone at progressively lower concentrations (Office of Air and Radiation, EPA, 2014) the National Ambient Air Quality Standard (NAAQS) was lowered to 70 ppb in 2015. Contributions to ambient ozone can be divided into two categories: the concentrations that would exist in the absence of any North American emissions of anthropogenic ozone precursors, which are primarily hydrocarbons and oxides of nitrogen, and the ozone enhancements produced by photochemical processing of the North America anthropogenic emissions of ozone precursors. The former has been called "North American background ozone" (e.g., Fiore et al., 2014). Transport of ozone into the U.S. provides the majority of North American background ozone, especially in the western U.S. This transported contribution is modified by ozone destruction mechanisms (e.g., deposition to surfaces, especially vegetation) and photochemical production from natural U.S. precursor emissions (e.g., NO_x from lightning and biogenic hydrocarbons from forests). Here we refer to this transported contribution to North American background ozone as "baseline" ozone, which flows into the U.S. from all natural and anthropogenic upwind sources. Baseline ozone mixing ratios can be directly measured at surface sites or airborne platforms along the West Coast in air masses not influenced by recent North American continental influences; it is such measurements that we evaluate in this paper. Cooper et al. (2015) thoroughly discuss baseline and background ozone.

An increase in baseline ozone over the past three decades has been identified from measurements along the North American Pacific coast (Jaffe et al., 2003) and further characterized and discussed in several papers (Cooper et al., 2012, 2010; Parrish et al., 2004, 2014, 2012, 2009). From an air quality perspective, an increase in the transported component of ambient ozone concentrations is of concern because that increase may offset air quality improvements that would otherwise be expected to result from reductions in ozone precursor emissions (e.g., Jacob et al., 1999). Importantly, preliminary indications of a slowing of the increase and perhaps reversal of the observed baseline ozone trend were identified and discussed (Parrish et al., 2014,

2012). An analysis of the ozonesonde record at Trinidad Head on the California coast (Oltmans et al., 2008) also shows that ozone in the 850–700 hPa (1.5–3.0 km altitude) layer peaked in the early 2000s.

Here we reanalyze the measurement records that originally established the past increase in baseline ozone at the U.S. West Coast and include the most recent measurements (early 2017 for two data sets). We show that the past increase has indeed ended, generally in the early to mid-2000s, and that these baseline ozone mixing ratios have since been decreasing. This noteworthy change in baseline ozone trends is expected to ease the achievement of U.S. air quality goals.

2. Data Sets and Methods

Only very limited data are available for quantifying long-term changes in baseline ozone mixing ratios at the North American West Coast. In this work we extend to recent years the three such data sets that have been discussed in several published studies. Sections 2.1–2.3 describe these three data sets, with references given to the previously published analyses; the supporting information gives a summary of the measurement dates and sites and references to the archives where these data are available. Section 2.4 describes the methods employed to quantify the long-term changes in these data sets.

2.1. Pacific Marine Boundary Layer Data

A time series of seasonal average ozone mixing ratios in the Pacific marine boundary layer (MBL) at the U.S. West Coast was derived from measurements at five coastal sites. The majority of these data were collected at the Trinidad Head California monitoring site operated by the Global Monitoring Division of NOAA's Earth System Research Laboratory beginning in 2002. By including measurements from four other coastal sites (Parrish et al., 2009), these results covered 1988–2006 in all seasons, with springtime 1985 data available from one site (Parrish et al., 1992). There were no statistically significant differences between seasonal average ozone mixing ratios obtained from separate sites (Parrish et al., 2009), so all seasonal averages were considered as a single time series. This work showed that filtering the measurement data to include only periods of high onshore winds effectively eliminated continental influences that could modify marine ozone mixing ratios. At each site, all hourly data that fell in a selected wind window were averaged to 3 month seasonal periods (March, April, May; June, July, August; September, October, November; and December, January, February). For this work, we have extended the Pacific MBL data set by calculating seasonal averages of the Trinidad Head ozone data through winter 2017 (i.e., December 2016 and January–February 2017), using the wind filter employed by Parrish et al. (2009). An earlier extension of this data record through 2010 (Parrish et al., 2014, 2012) is identical to that presented here.

2.2. Lassen Volcanic NP Data

Lassen Volcanic National Park in California is the only elevated site in western North America that receives relatively undisturbed air inflow from the Pacific Ocean and where ozone measurements have been made over decadal time scales. Measurements were begun there in 1988. Unfortunately, this site lies approximately 240 km inland from the Pacific coast. Jaffe et al. (2003) first analyzed the long-term, seasonal average trends in this data set over the 1988 to 2002 period. They showed that filtering the data to isolate marine air, either based on back trajectory air parcel calculations or wind sector filtering, yielded trends not statistically different from the trends including all hourly data. Parrish et al. (2012) extended this time series through 2010 based on seasonal averages of all hourly data without any filtering, and here we further extend this record in the same manner through winter 2017.

2.3. North American Free Troposphere Data

Cooper et al. (2010) compiled springtime (April and May) ozone measurements in the free troposphere (3–8 km altitude) from all available platforms (research and commercial aircraft, ozonesondes, and lidar) over the eastern North Pacific and across western North America. This effort yielded a continuous data record for 1995–2008, with a single earlier year (1984). Cooper et al. (2010) compared the trends from the total data set with those derived from a subset of the data that used a particle dispersion model to filter out data with a recent, strong influence from the North American boundary layer; no statistically significant differences were found between these trends. Cooper et al. (2012) extended this data set with no filtering for air mass origin through 2011, and Lin et al. (2015) further extended this record through 2014, again with no filtering of the data; this latter data set with temporal coverage through 2014 is considered here. Cooper et al. (2012, 2010)

analyzed five percentiles of the data; here we consider only the medians to be approximately consistent with the seasonal averages of the Pacific marine boundary layer and Lassen Volcanic NP data sets.

Lin et al. (2015) compared North American free troposphere data set with ozone mixing ratios calculated with the GFDL-AM3 global chemistry-climate model nudged to reanalysis winds. They find that spatial sampling biases in the measurements may have influenced the calculated trends; these sampling biases must be recognized in considering the present results.

2.4. Analysis Approach

The conceptual model that is the basis of our analysis assumes that temporal variations of the seasonal averages are driven by two factors. First, there is an underlying, relatively smoothly varying, continuous long-term trend reflecting changes in the sources and sinks of tropospheric ozone (e.g., increasing or decreasing ozone precursor emissions, land-use changes that affect surface deposition, and changing climate) and perhaps long-term changes in transport patterns; we will refer to these long-term changes as the trend. Second are more chaotic, shorter duration changes due to interannual to decadal variability in circulation regimes (i.e., internal climate "noise"; see Lin et al., 2015 for a discussion). Many studies have used an ordinary linear least squares regression to at least approximately separate these two factors (e.g., Cooper et al., 2012, 2010; Lin et al., 2015, 2017, and references cited therein). The slope of the linear regression in units of ppb yr^{-1} is assumed to represent the trend in the data, while the scatter about that regression line is assumed to represent the internal climate noise. An important shortcoming of this approach is that in many cases a linear change only poorly represents the trend. In such a situation the linear regression slope gives an estimate for the average temporal change in the measured ozone mixing ratios over the time span of the measurement record, but the derived line does not accurately describe the trend. Likewise, in such cases the deviations of the measurements from the derived line represent a fraction of the long-term changes, in addition to the climate noise.We utilize a more general approach to quantify the underlying trends in the temporal series of seasonal average ozone measurements investigated here—a nonlinear least squares regression fit of a polynomial to the series, in this application the quadratic polynomial in equation (1):

$$[O_3] = a + bt + ct^2, \tag{1}$$

where a, b, and c are constant coefficients that quantify the underlying long-term trends in the time series. The nonlinear least squares regression fit returns 95% confidence limits for each of the three coefficients.

Equation (1) is a specific realization of a general approach; a polynomial fit to the data is equivalent to deriving a power series expansion of the underlying, relatively smoothly varying, continuous long-term trend. Any function can be fit to any desired precision if enough terms are included in the power series expansion (i.e., if more terms are included in the polynomial of equation (1)). However, the temporal series we are considering have significant internal climate noise about the trend, which limits the number of statistically significant terms of the power series that can be determined from a finite temporal series. If the absolute value of the final coefficient is larger than its 95% confidence limit, then that term is considered to be statistically significant. In the analyses in this paper, three and no more than three coefficients that are unambiguously statistically significant can be derived, and these we will discuss.

To most precisely determine the coefficients from the nonlinear regression, the time reference (i.e., the time origin) must be near the center of the data series. Here that reference is selected as the year 2000 (i.e., t in equation (1) is equal to year 2000). The first coefficient (a, with units ppb O_3) is the intercept of the fitted curve at this reference time; it gives us information regarding the absolute magnitude of the ozone mixing ratio. The second coefficient (b, with units ppb O_3 yr^{-1}) is the slope of the fitted curve at the reference time; it gives the best estimate the time rate of change of ozone in the year 2000. Finally, the third coefficient (c, with units ppb O_3 yr^{-2}) is equal to one half of the (constant) time rate of change of the slope of the fitted curve. It is important to note that this third term is quite important for quantifying ozone trends; the temporal series of seasonal ozone mixing ratios that we examine here generally have trends with positive slopes in the early parts of the record and negative slopes in the later parts, so that the overall change is small, but nevertheless, the trends are statistically significant.

Parrish et al. (2014, 2012, 2009) and Logan et al. (2012) utilized identical or closely related approaches to that described above. Most of the trends investigated had three statistically significant coefficients as illustrated in

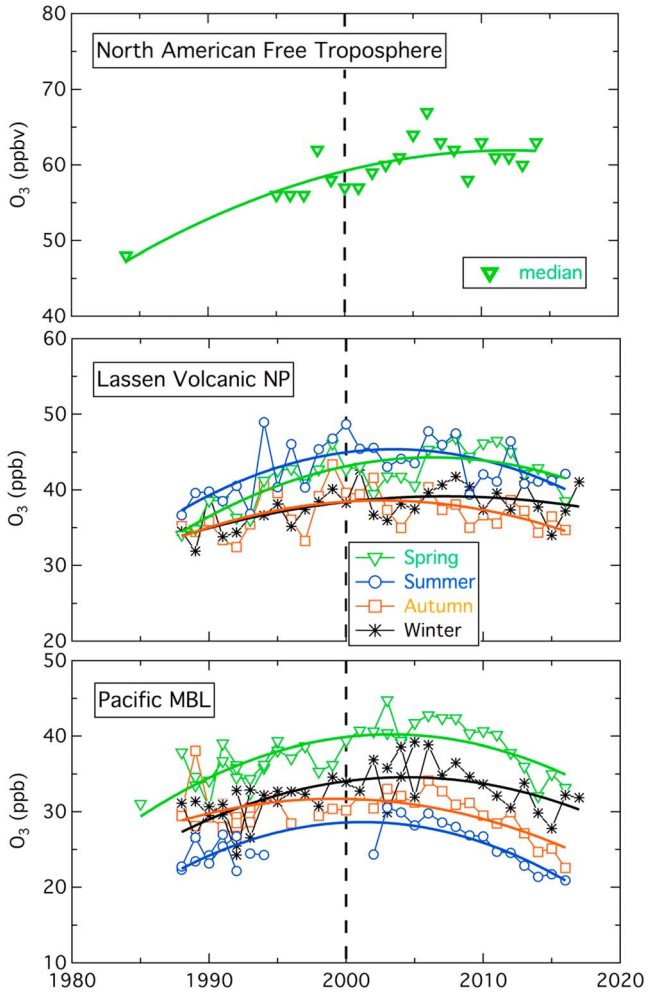

Figure 1. Seasonal O_3 averages measured in the vicinity of the northern U.S. Pacific coast. The solid lines give the least squares regression of equation (1) for each seasonal data set described in section 2. Colors and symbols identify the seasons as indicated in the annotation. Note that the O_3 mixing ratios differ on the three abscissas, but each spans a total range of 40 ppb.

equation (1), but Parrish et al. (2014) showed examples where four or even five polynomial terms were statistically significant.

3. Results

The temporal series of the seasonal average ozone mixing ratios discussed in section 2 are illustrated in Figure 1, along with the corresponding least squares regression fits to equation (1). Table 1 gives the coefficients (with 95% confidence limits) derived from these regression fits. Consistent with previous discussions of the long-term increase in baseline ozone at the North American West Coast (Cooper et al., 2012, 2010; Jaffe et al., 2003; Parrish et al., 2004, 2014, 2012, 2009), the positive *b* coefficients indicate that the derived long-term trends were increasing in year 2000 (except for autumn in the Pacific MBL). The *c* coefficients are all negative and statistically significant, indicating that, on average, the slopes of the long-term trends have been decreasing in all seasons over the period of the data records. Parrish et al. (2012) also investigated regression fits of equation (1) to these data sets, and the coefficients they derived are generally statistically consistent with those reported here in Table 1, but the confidence limits were significantly larger due to the shorter data records (only through 2010) available at that time. Consistent with the results in Table 1, Parrish et al. (2012) nearly always found negative *c* coefficients in all seasons in all data sets, but most were not statistically significantly different from zero (see figures in supporting information of Parrish et al., 2012). It is now clear that trends in seasonal average baseline ozone mixing ratios at the U.S. West Coast are not well approximated by constant linear changes. The time rate of change of these series of ozone mixing ratios (i.e., the slopes) has been decreasing at an approximately constant rate, so that a majority of these temporal ozone mixing ratio series reached maxima and are now decreasing.

Equation (1) can be manipulated to solve for the year that the maximum of the continuous long-term seasonal average ozone mixing ratio was reached, and that year can be calculated by substituting the coefficient values from Table 1. These years are included in the table for each season and data set; Figure 2 compares the resulting ozone maxima for each season and data set. In the Pacific MBL and at Lassen Volcanic NP the best

Table 1
Coefficients of the Regressions of the Measured Seasonal Average Time Series to Equation (1), Root-Mean-Square Deviation (RMSD), and the Year of the Maximum of the Fits Shown in Figure 1

Season	*a* (ppb)	*b* (ppb yr^{-1})	*c* (ppb yr^{-2})	RMSD (ppb)	r^2	$Year_{max}$
			Pacific marine boundary layer			
Spring	39.9 ± 1.1	0.21 ± 0.09	−0.033 ± 0.012	2.4	0.55	2003 ± 2
Summer	28.6 ± 1.3	0.09 ± 0.08	−0.035 ± 0.012	1.8	0.63	2001 ± 1
Autumn	31.7 ± 1.1	−0.04 ± 0.09	−0.023 ± 0.012	2.2	0.36	1999 ± 2
Winter	34.1 ± 1.3	0.24 ± 0.11	−0.027 ± 0.013	2.7	0.40	2004 ± 3
			Lassen volcanic NP			
Spring	43.1 ± 1.2	0.37 ± 0.11	−0.029 ± 0.013	2.2	0.65	2006 ± 3
Summer	45.0 ± 1.6	0.23 ± 0.14	−0.034 ± 0.017	2.8	0.43	2003 ± 3
Autumn	38.5 ± 1.3	0.12 ± 0.12	−0.022 ± 0.014	2.3	0.29	2003 ± 2
Winter	38.4 ± 1.2	0.20 ± 0.11	−0.014 ± 0.012	2.2	0.34	2007 ± 4
			North American free troposphere (median)			
April, May	59.2 ± 1.5	0.45 ± 0.15	−0.019 ± 0.015	2.4	0.69	2012 ± 11

Note. Also included are the squares of the correlation coefficients between the measurements and the regression fits.

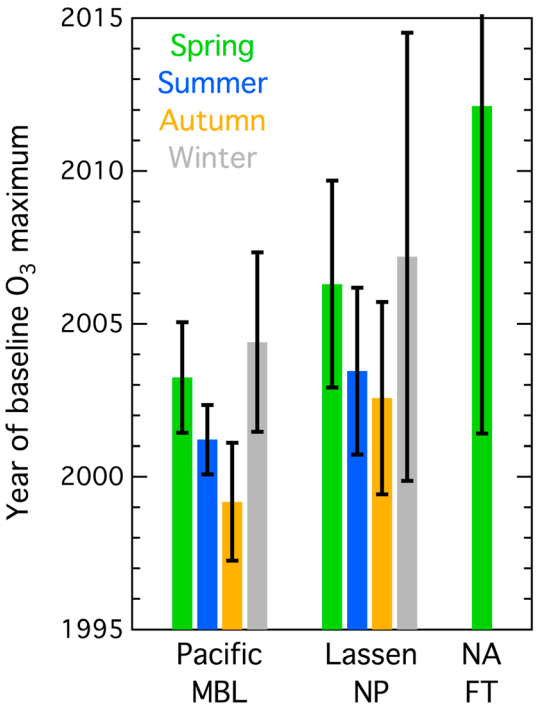

Figure 2. Year of maximum seasonal average baseline O_3 mixing ratios in each of the three North American data sets. Error bars give 95% confidence intervals for these determinations.

estimates for the years of the maxima fall between 1999 and 2008, with the same sequence of seasonal maxima (autumn first, followed in order by summer, spring, and winter). Although the maxima appear to occur later at Lassen Volcanic NP than in the Pacific MBL, the maxima agree within the 95% confidence interval for the differences between the two sites. The best estimate of the year of the maximum of the springtime North American free troposphere data set (2012) is later than the spring maxima of the other two data sets, but this difference is not statistically significant due to the large confidence limits of the North American free troposphere maximum.

The two additional statistics included in Table 1 give indications of the internal climate noise superimposed on the long-term trends. The root-mean-square deviation (RMSD) of the individual seasonal averages from the fits to equation (1) shows that the internal climate noise accounts for 1.8 to 2.8 ppb scatter about the fits. We also calculate the square of the correlation coefficient (r^2) for the linear regressions between the seasonal averages calculated from the respective polynomial fits and those measured. These r^2 values approximate the fraction of the variability in each seasonal time series that is captured by those polynomial fits; these values are between 0.3 and 0.7.

4. Discussion and Conclusions

Reducing the Nation's emissions of ozone precursors is the only effective tool available to improve local and regional air quality over the U.S. Emission reduction efforts over multiple decades have yielded dramatic improvement in ozone air quality (e.g., Parrish & Stockwell, 2015), but many regions still do not meet the NAAQS. The extent of further reductions necessary for a given region to reach the standard is not quantitatively known, but the reversal of the long-term increase in baseline ozone entering the U.S. from the Pacific will certainly ease the difficulty of achieving further reductions in ozone concentrations. Here we have shown that this reversal has occurred, but we have not established its cause. Recent analyses of satellite data (Liu et al., 2017) indicate that the decade-long increase in NO_x emissions in China has ended and that those emissions are now decreasing; this emission change may be at least partially responsible for the observed baseline ozone decrease.

Most published characterizations of the absolute mixing ratios and trends of North American background ozone rely on calculations by global models (e.g.,Fiore et al., 2014; Lin et al., 2015, 2017). Unfortunately, global models only poorly reproduce observed baseline ozone (Derwent et al., 2016; Parrish et al., 2014), so the accuracy of North American background ozone mixing ratios calculated by such models must be considered cautiously. Fiore et al. (2014) and Lin et al. (2015, 2017) used the GFDL-AM3 chemistry-climate model nudged to reanalysis winds to provide the most extensive characterization of background ozone over the U.S. Parrish et al. (2014) investigated the GFDL-CM3 global model, which is closely related to the GFDL-AM3 model, except that it utilizes free running meteorology. Here it is informative to revisit the performance of the GFDL-CM3 model for the three data sets considered above; Figure S1 and Table S1 of the supporting information summarize the model results in the same format as that for the observations illustrated in Figure 1 and Table 1 above. Large differences between the observed and modeled baseline ozone mixing ratios and trends are apparent. The *a* parameters (reflecting absolute ozone mixing ratios in the year 2000) for the model results are 11 to 18 ppb (i.e., 21 to 64%) higher than for the observations. Lin et al. (2012) discuss a similar bias in the GFDL-AM3 with nudged meteorology. Positive biases of similar magnitude are also seen in other global models (e.g., Yan et al., 2016). Derwent et al. (2016) discuss the biases in annual average ozone at Trinidad Head (the primary MBL site considered in this work) for the 14 chemistry-climate models that participated in the Atmospheric Chemistry Coupled Climate Model Intercomparison Project (ACCMIP). To our knowledge, the cause(s) of these biases remain undiagnosed.

Current state-of-the-art chemistry-climate models also greatly underestimate the rate of change of the slope of the temporal trends of baseline ozone (reflected by the *c* parameters) (Parrish et al., 2014). Model results

give much smaller (generally by a factor of ~5) values for this parameter compared to observations. The rapid change of the slopes of the temporal trends in the observations, but not in the model results, can potentially confound any comparison of linear trends between model results and observations. The *b* parameters (equal to the temporal trend slope in the year 2000) do allow a direct comparison; these parameters are generally smaller for the model results than found in the observations (deltas of −0.39 to 0.08 ppb yr^{-1} with only autumn in the Pacific MBL exhibiting a model slope higher than the observational result). Slope comparisons for other years or time periods must carefully consider the differing rate of change of these slopes between models and observations. Parrish et al. (2014) and Staehelin et al. (2017) find that model results capture only ~50% of baseline ozone changes observed over the five decades before the year 2000. This finding is consistent with the closely related finding that the ACCMIP models fail to reproduce preindustrial observations (Stevenson et al., 2013). The model results do agree with the observations that maxima have or will be reached in the baseline ozone mixing ratios in all seasons in all data sets. However, as expected from the model-measurement differences in the derived *b* and *c* parameters, the years of the maxima differ between the models and the observations, with the model maxima 7 years earlier to 6 years later, except in winter when the model predicted maxima are much later (~30 years) than observed. It is clear that the GFDL-CM3 global model with free running meteorology only poorly describes baseline ozone mixing ratios and their trends at the North American West Coast. Since this baseline ozone is the dominant contributor to North American background ozone, at least in the western U.S., these global models cannot be expected to accurately calculate North American background ozone and its trends in this region.

Lin et al. (2012, 2015, 2017) used the GFDL-AM3 model to quantify the internal climate noise and to examine its influence on trend determinations. This model with nudged meteorology does reproduce much of the variability about the long-term trends quantified in the observational record. For example, Lin et al. (2015) find that a large fraction of the variance in observational data sets is due to internal climate noise rather than the long-term trends. The r^2 values given in Table 1 provide an estimate of the fraction of the total variability in the respective data sets due to the long-term trends (i.e., 29 to 69%), which leaves a large fraction of the variability to be accounted for by internal climate noise or other causes. Lin et al. (2015) also argue that the airborne data that comprise the North American free troposphere data set discussed here overestimate the magnitude of the long-term changes in springtime free troposphere ozone due to spatial sampling biases combined with interannual variability in transport regimes. These issues may account for the apparently steeper increase in the North American free troposphere data in Figure 1 compared to the two surface measurement data sets. As more years of data are added to the springtime free troposphere ozone record, these uncertainties are expected to decrease. A notable feature of Figure 1 is that in spring in the free troposphere and at Lassen Volcanic NP the ozone decrease has been slower to begin than in other data sets. This is even clearer at Lassen Volcanic NP if only April and May (the two months included in the free troposphere data set) are considered. Gratz et al. (2014) report a trend analysis for April–May 2004–2013 ozone at an additional site representative of the free troposphere (Mount Bachelor Observatory in Oregon). This relatively short data record exhibits larger internal climate noise than the other data sets and does not yet indicate that a maximum has been reached in springtime baseline ozone mixing ratios. We have analyzed these data for all seasons through two additional years (to 2015) using the same approach discussed in section 2.4; the results are statistically consistent (within their large confidence limits) with the analysis of seasonal ozone trends discussed in this work.

Acknowledgments

The authors acknowledge support from NOAA's Atmospheric Chemistry, Carbon Cycle, and Climate (AC4) Program. The National Park Service provided the recent Lassen NP ozone data (https://www.nature.nps.gov/air/monitoring/network.cfm). We are grateful to Dan Jaffe for providing us with the Mount Bachelor Observatory ozone data. All of the data utilized in this paper are available from public archives that are referenced in this paper or from the authors of papers referenced herein. Disclosure: David Parrish also works as an atmospheric chemistry consultant (David D. Parrish, LLC); he has had contracts funded by several state and federal agencies, although they did not support the work reported in this paper. The scientific results and conclusions, as well as any views or opinions expressed herein, are those of the author(s) and do not necessarily reflect the views of NOAA or the Department of Commerce.

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