Long-term Baseline Ozone Changes in the Western US: A Synthesis of Analyses

David D. Parrish[1,2], Richard G Derwent[3], and Ian C. Faloona[2,4]

[1] David.D.Parrish, LLC, Boulder, Colorado, USA

[2] Air Quality Research Center, University of California, Davis, California USA

[3] rdscientific, Newbury, Berkshire, UK

[4] Department of Land, Air, & Water Resources, University of California, Davis, California, USA

Corresponding author: David D. Parrish, (david.d.parrish.llc@gmail.com)

Key Points:

- Reported trends in tropospheric ozone concentrations transported into the Western US vary between -2.8 to +7.0 ppb/decade
- All reported trends agree with an overall non-linear change – ozone increasing before the mid-2000s and slowly decreasing thereafter
- About 1/3 of the variance in reported trends is due to autocorrelation in the data, which was not adequately considered in prior analyses

Abstract

Quantification of the magnitude and long-term changes of ozone concentrations transported into the US is important for effective air quality policy development. We synthesize multiple published trend analyses of western US baseline ozone, and show that all results are consistent with an overall, non-linear change – rapid increase during the 1980s that slowed in the 1990s, maximized in the mid-2000s, and was followed by a slow decrease thereafter. This non-linear change accounts for ~2/3 of the variance in the published linear trend analyses; we attribute the other 1/3 to unquantified autocorrelation in the analyzed data sets. Recent systematic changes in baseline ozone at the US West Coast have been relatively small - the standard deviation of the 2-year means over the 1990-2017 period is 1.5 ppb. International efforts to reduce anthropogenic precursor emissions from all northern mid-latitude sources could possibly reduce baseline ozone concentrations, thereby improving US ozone air quality.

Plain Language Summary

Ozone is an air pollutant with significant human and ecological health impacts. Air masses transported into the western US from over the Pacific Ocean carry ozone concentrations that are, on average, a large fraction of the US health standard, so quantifying these trans-boundary concentrations are important for developing a complete picture of US air quality. Published analyses of temporal trends of these transported ozone concentrations vary widely, from early reports of increases to more recent reports of decreases. We show that the long-term ozone changes have been nonlinear, with concentration increases before the mid-2000s, followed by decreases thereafter. Superimposed on the overall changes is significant interannual variability that makes accurate determination of systematic trends over decade-scale time periods uncertain. The recent decreases in transported ozone concentrations is good news for US air quality, as it eases the difficulty of achieving the ozone air quality standard.

1 Introduction

Air masses from the Pacific marine environment enter the continental atmosphere over the western US carrying ozone concentrations determined by natural and anthropogenic sources and sinks in upwind regions. These transported ozone concentrations are large enough to significantly impact air quality in urban and rural US locations; fully understanding this impact requires characterization of the temporal and spatial distribution of those ozone concentrations. Over the past two decades, a number of observational-based studies have quantified the average ozone concentration changes at specific western US locations thought to represent changes in the transported marine air; Table 1 lists 28 of these quantifications. Reported average trends over different time periods vary widely, from relatively large increases to smaller magnitude decreases. Our goal in this study is to synthesize these disparate results, and to develop a consistent picture of the overall, decadal-scale temporal change in the transported ozone concentrations over the past 3 to 4 decades at the US west coast.

A conceptual picture provides a useful framework for understanding the temporal variation of ozone at northern mid-latitudes. On average, prevailing westerly winds define a circulating air stream repeatedly passing over all continents and oceans. The average net lifetime of ozone at these latitudes (~100 days) is longer than the circum-global transport time (~30 days). Overall, the long lifetime and zonal transport imply that a mean ozone concentration is established on a time scale of weeks to months, and that this mean concentration is similar throughout northern mid-latitudes; we roughly estimate that this similarity is within ± 10% from the top of the planetary boundary layer to ~9 km at all longitudes (e.g., see Figure 5 of Parrish et al., 2020). This picture also implies a relatively smooth and systematic seasonal cycle of ozone in baseline air masses. A zonally similar mean does not imply a lack of ozone variability, as ozone varies about that mean on a wide spectrum of shorter and longer time scales, including decadal climate variability (e.g., Lin et al., 2014), sporadic events such as wildfires (Lin et al., 2017), and heatwaves and droughts (Lin et al., 2020). The first four sections of the Supporting Information describe this conceptual picture and ozone variability in more detail.

The subject of this study is decadal and longer scale ozone changes, which are caused by long-term changes in precursor emissions and the changing climate; quantifying these changes must account for the variability of ozone on the wide spectrum of shorter time scales, variability that tends to obscure the long-term changes of interest. Importantly, our guiding conceptual picture implies that these long-term changes must be zonally similar, since it is the zonally similar average ozone concentration that must change; a recent analysis of baseline ozone concentrations at the west coasts of North America and Europe (Parrish et al., 2020) document this expected zonal similarity.

Several terms appear in the literature in reference to the transported ozone concentrations. A common general term is background ozone. However, it is important to note that presently observed concentrations do not represent natural ozone concentrations, i.e., those that existed before industrial development, since anthropogenic emissions of ozone precursors have increased ozone concentrations throughout northern mid-latitudes. For clarity, in this work we adopt the term “baseline” (e.g., see discussion in Chapter 1 of HTAP, 2010) to refer to ozone concentrations measured at western US locations that receive transported marine air without significant perturbation from recent local or regional North American influences. It is the long-term change in these baseline concentrations that we seek to quantify.

In addition to the linear trend analyses included in Table 1, two published analyses utilized non-linear approaches to quantify long-term changes of baseline ozone concentrations at northern mid-latitudes; both reached similar conclusions. Logan et al. (2012) analyzed several European baseline ozone data sets that extended through 2009, and showed that ozone increased by 6.5-10 ppb in 1978-1989 and 2.5-4.5 ppb in the 1990s, with that increase ending and a maximum reached in the 2000s, followed by decreasing concentrations, at least in summer. Parrish et al. (2020) analyzed those same data sets, which by then extended through 2018, plus additional European and North American data sets; in total 8 baseline data sets from surface sites, balloon-borne sondes and aircraft over western Europe and western North America were considered. These measurements covered altitudes from sea level to 9 km. Again, an initial, relatively rapid increase was observed, with ozone concentrations reaching a maximum in the mid-2000s, followed by decreasing concentrations. An important conclusion of these analyses is that, within statistical confidence limits, the same non-linear long-term baseline ozone change has occurred throughout northern mid-latitudes at all altitudes. The goal of this paper is to compare and contrast published linear trend and non-linear long-term change analyses of multi-decadal ozone time series collected at the surface and in the free troposphere over the western US, and to synthesize those analyses to provide an accurate and complete geophysical quantification of long-term changes in baseline ozone at the US West Coast.

2 Materials and Methods

As discussed above, a relatively large number of analyses of long-term baseline ozone changes have been published based upon ozone time series collected in the continental western US. This work is based upon the results of those analyses; no new data sets are analyzed.

Any analysis aiming to quantify the overall long-term change in tropospheric ozone at northern mid-latitudes must effectively deal with two issues: the non-linearity of the long-term changes that have been documented in previous work, and the substantial interannual variability in mean ozone concentrations that tends to obscure the long-term changes. All linear trend analyses return a single parameter value that quantifies the trend; this is effectively an average slope of the long-term change over the span of the analyzed time series. Hence, by its very nature linear trend analysis is ill-suited to quantify non-linear, long-term changes.

Without a priori knowledge of the functional form of the ozone concentration changes, long-term change analysis is generally based either on linear trend analyses or on fits of the first few terms of a power series to measured ozone concentrations. A power series fit does not assume any particular functional form for the time evolution; it is quite flexible, as it provides a quantitative description of the average continuous, long-term change in any series of observations (Parrish et al., 2019). The power series fit is obtained through a regression fit of a polynomial to the measurements, with retention of only the statistically significant terms to avoid over fitting the time series. In this work no more than the first three terms are considered,

$$O_3 = a + b\mathrm{t} + c\mathrm{t}^2, \quad (1)$$

because no more than three statistically significant terms (i.e., those with 95% confidence intervals not containing zero) are encountered in any fits in this study. Fits with only the first two terms statistically significant are equivalent to linear regressions. A statistically significant third term indicates the average long-term change is non-linear, i.e. d^2O_3/dt^2 is non-zero, but it does not indicate that the overall change is necessarily parabolic. Equation 1 does not account for seasonal variations; these variations are eliminated by fitting to annual means, seasonal means, or deseasonalized monthly means (sometimes called monthly residuals or monthly anomalies). Ozone concentrations are consistently quantified as mixing ratios, with units of 10^{-9} mole O_3 per mole air, denoted as ppb.

The time origin is chosen as the year 2000 (i.e., t in Equation 1 equals the year - 2000) to ensure precise determination of the coefficients in Equation 1. The first coefficient (a, with units ppb O_3) is then the intercept of the fitted curve at the year 2000, and quantifies the absolute concentration at that year. The second coefficient (b, with units ppb O_3 year^{-1}) is the slope of the fitted curve at that year, and gives the best estimate of the time rate of change of O_3 in 2000. The third coefficient (c, with units ppb O_3 year^{-2}) gives the constant curvature of the fit. For non-linear fits, a negative value is generally derived for c; such curves indicate ozone concentrations increasing early in the data record, reaching a maximum, and then decreasing at later times. The year of that maximum is

$$\mathrm{year}_{max} = 2000 - b/2c. \quad (2)$$

Parameter values taken from published analyses are generally given with specified 95% confidence limits. We also specify 95% confidence limits in this work. However, it is important to recognize that confidence limits reported in the literature are generally derived from the variability of the data points about the fitted line or curve without a full analysis of the autocorrelation in those data. As a consequence, the quoted confidence limits are generally underestimated to an unknown extent. This issue is important to the present discussion, when comparing and contrasting results from different analyses.

Within the baseline troposphere, average ozone concentrations do exhibit some systematic spatial variability, despite the general zonal uniformity at northern mid-latitudes; in particular baseline ozone concentrations generally increase with altitude (e.g., Oltmans et al., 2008). To remove this systematic variability when comparing long-term changes derived from data sets with different mean concentrations, fits of Equation 1 are normalized to zero at the year 2000 by subtracting the corresponding values of the *a* parameter. The normalization of a linear fit to a quadratic fit is discussed in the following section.

Polynomial fits have been used previously to quantify long-term changes in ozone concentrations (e.g., Logan et al., 2012; Parrish et al., 2012; 2017; 2020; Derwent et al., 2018). Such fits, as well as linear fits, are not based on a physical model of the observed temporal changes, so they cannot be reliably extrapolated to times outside the period of observations. For ease of presentation and discussion, the values of the *b* and *c* parameters are given as ppb O_3 decade^{-1} and ppb O_3 decade^{-2}, respectively. The Section S5 of the Supporting Information discusses the relation of polynomial fits and linear trend analysis in more detail, including their respective advantages and disadvantages.

3 Results and Discussion

The results of Parrish et al. (2020) are reproduced in Figure 1a: deseasonalized, normalized monthly means from each of eight data sets considered (gray points), 2-year averages of those monthly means (black symbols with error bars indicating standard deviations), and a quantification of the average long-term baseline ozone change (black curve). This curve is the least-squares fit of a quadratic polynomial (i.e., Equation 1) to the monthly means. Table 2 gives the parameters of the fit; Equation 2 indicates that a maximum average baseline ozone concentration was reached in the year 2005.7 ± 2.5. Figure 1a also includes quadratic polynomial fits to time series from a US Pacific marine boundary layer (MBL) data set and from one higher altitude (1.8 km) site further inland operated by the National Park Service at Lassen Volcanic National Park. Parrish et al. (2017) analyzed these data sets to demonstrate that the long-term trend in baseline ozone concentrations at the US West Coast had reversed from an early increase to a later decrease, with a maximum reached in early to mid-2000s; Figure 1a extends the analysis of those data sets through 2017. These are also two of the eight data sets analyzed by Parrish et al. (2020). There are apparent differences between the three curves, most prominently a more rapid recent decrease in the Pacific MBL data; however, Table 2 shows that the parameters from both the Lassen Volcanic NP and the Pacific MBL fits agree with those of the northern mid-latitude quadratic fit of Parrish et al. (2020) within their indicated confidence limits. Thus, there is no statistically significant difference between the three quadratic fits.

Previously published linear trend analyses of ozone changes within the western US are compared with the results of Parrish et al. (2020) in Figure 1b and Table 1 includes reported trends from 28 separate analyses. A selected sample of those trend results are represented by straight line segments with slopes equal to the reported trends and with lengths equal to the time spans of the analyzed data sets. Each straight line segment is normalized to the non-linear analysis results by minimizing the sum of the square of the deviations between the line segment and the 2-year averages (black symbols in Figure 1b) that fall within the time span of the corresponding data set. A common general feature characterizes these results – the earlier the start and end times of the trend analysis, the larger the quantified trend. This feature follows from the slowing of the increase in baseline ozone indicated by the black curve in Figure 1. The three earlier analyses (Jaffe et al., 2003; Parrish et al., 2009; and Cooper et al., 2010) consider data predominately from before the baseline ozone maximum was reached, and therefore report the larger trends. The multiple analyses of Cooper et al. (2020) and the two analyses of Gaudel et al. (2020) cover later time periods that include the ozone maximum with extended periods on either side; they therefore report small trends, some positive and some negative. Cooper et al. (2020) report three analyses, with progressively later starting times for each of 4 data sets; the derived trends become progressively more negative for the later starting times. These features of the linear trend analyses are all consistent with the overall behavior of the quadratic analysis indicated by the black curve.

The 28 referenced trend analyses considered data sets covering a total of 34 years (1984-2017) and derived widely varying trends (-2.8 to +7.0 ppb/decade). The nonlinearity of the long-term change accounts for much of these differences, but interannual variability about that long-term change also contributes to differences in the results. The analysis of Cooper et al. (2010) gave the largest trend (+7.0 ppb/decade); Lin et al. (2015) show that this result was an overestimate, as were 5 related analyses (Cooper et al., 2012; Lin et al., 2015), due to substantial influences from interannual variability. None of the 28 trend analyses accounts for uncertainties introduced into the results from the autocorrelation in data sets associated with interannual variability, although some address shorter-term, month-to-month autocorrelation (e.g., Gaudel et al., 2020). Section S6 of the Supporting Information discuss illustrates longer-term autocorrelation in two example data sets. The analysis based on the non-linear, least-squares fit included in Figure 1 (Parrish et al., 2020) effectively addresses both non-linearity and the longer-term autocorrelation in the longer, 40-year (1978-2017) data set. The resulting quadratic polynomial fit is derived from deseasonalized monthly means, but a fit to the 2-year averages of those monthly means gives nearly identical parameter values, but with significantly larger confidence limits; these larger confidence limits are included in Table 1. The high degree of temporal and spatial averaging in the 2-year means greatly reduces the influence of the autocorrelation associated with interannual variability.

We conclude that the black curve in Figure 1 provides a realistic and accurate quantification of the decadal-scale baseline ozone changes over the western US. The results of all published trend analyses are generally consistent with this non-linear fit over the shorter time periods of the trend analyses. The result of each linear trend analysis can be quantitatively compared with the average trend quantified by the quadratic curve over the time period of the trend analysis, which is equal to the slope of a straight line segment connecting the two points on the quadratic fit at the beginning (t_1) and end (t_2) times of the period included in the trend analysis:

$$\text{slope} = b + c^*(t_1 + t_2). \qquad (3)$$

Table 1 compares the slopes calculated from Equation 3 with the published trends, and Figure 2a shows their overall relationship. The quadratic fit accounts for ~67% of the variance in the 28 linear trend analysis results. We attribute the remaining ~33% of the variance to the influence of interannual variability and to any spatial differences between trends at the measurement locations, which include surface sites in marine and continental environments, as well as data sets from the lower and mid free troposphere.

It is possible to independently determine a quadratic description of the overall long-term ozone changes from the reported linear trends. Equation 3 indicates that a plot of the derived trends as a function of the centers of the time periods of the respective trend determinations will define a straight line with a slope of 2*c and a y-intercept of b of a quadratic curve as given by Equation 1. Figure 2b shows that plot, which includes a linear regression to the 28 trend determinations. Note that the x-intercept corresponds to the time that the trend is zero, i.e. the $year_{max}$ given by Equation 2. Table 2 compares the parameters from this quadratic determination with the three discussed previously; generally there is agreement within the indicated confidence limits, but reasons for exceptions to this agreement are discussed below.

The error bars illustrated in Figure 2a indicate the 95% confidence limits reported for the respective trends. The fraction of these error bars not overlapping the 1:1 line (~50%) is much larger than the expected 5%. (In Table 1 confidence limits are also included for the slopes derived from the quadratic curve through a propagation of error calculation based on the confidence limits of the quadratic parameters indicated in Table 2; inclusion of these confidence limits does not significantly increase the number of points in Figure 2a that overlap the 1:1 line.) Similarly, the parameter values compared in Table 1 do not agree in all cases within the derived confidence limits. We attribute this disagreement primarily to underestimation of the confidence limits derived in the trend analyses due to inadequate treatment of the autocorrelation in the data sets resulting from interannual variability. The trends derived in the six earlier analyses of springtime ozone mixing ratios in the free troposphere over western North America (Cooper et al., 2010; 2012; Lin et al., 2015) all are particularly influenced by interannual variability, as discussed by Lin et al. (2015). Increasing all confidence limits for the trends in Table 1 and the lower three rows of Table 2 by 50% brings the fraction of the error bars in Figure 2 overlapping the 1:1 line into close agreement with expectations, and eliminates the disagreements in Table 2. The scatter of the trend analyses about the fits in Figure 2 emphasizes the importance of careful consideration of the impact of autocorrelation in time series of ozone measurements caused by interannual variability.

4 Summary and Conclusions

The long net lifetime of ozone in the prevailing westerly winds at northern mid-latitudes implies that a common long-term change in mean baseline ozone concentrations must have occurred throughout this zone. Parrish et al. (2020) document this similarity and quantify the long-term change with a fit of a quadratic polynomial to monthly and biennial means of multiple data sets. Linear trends reported for different time periods in 28 published analyses of western US baseline ozone data sets vary between -2.8 to +7.0 ppb/decade. The quadratic fit of Parrish et al. (2020) accounts for about two-thirds of the variance in the trend results, with the remaining one-third attributed to interannual variability, which adds uncertainty to the trend determinations. All reported trend analyses for western US baseline ozone data sets are consistent with the picture conveyed by that quadratic fit - ozone increasing at the beginning of measurement records with the rate of that increase progressively slowing, and ozone reaching a maximum in the mid-2000s and decreasing thereafter. The quadratic fit provides an excellent fit to the twenty 2-year means over the 1978-2017 period, capturing about 89% of their variance with a root-mean-square deviation between the fit and the means of 1.3 ppb; that fit also captures about 67% of the variance in the 28 trend analyses, and agrees well with a quadratic fit derived from the trend analysis results themselves.

Baseline ozone transported into the US constitutes a large fraction of the 70 ppb ozone National Ambient Air Quality Standard (NAAQS); thus changes in baseline concentrations affect the difficulty of achieving the NAAQS in US nonattainment areas. During the 1980s and 1990s those baseline concentrations were increasing, making attainment of US air quality goals progressively more difficult and partially offsetting the air quality improvement that resulted from emission controls (Jacob et al., 1999). In the mid-2000s baseline ozone concentrations maximized and then slowly decreased at an average rate of 0.9 ± 0.8 ppb decade^{-1} over the 2000-2018 period (Parrish et al., 2020). This small decrease, if continued, would gradually lessen the difficulty of achieving US air quality goals. However, despite the changes evident in Figure 1, since 1990 average baseline ozone concentrations entering the western US exhibit little overall change; the standard deviation of the fourteen 2-year means over the 1990-2017 period is only 1.5 ppb. Improvement in US ozone air quality has come primarily from continued precursor emission controls that reduce local and regional photochemical ozone production; this improvement can continue and possibly be augmented by international efforts to reduce anthropogenic precursor emissions from all sources at northern mid-latitudes, thereby reducing the hemisphere-wide transported baseline ozone concentrations.

Acknowledgments, Samples, and Data

The authors are grateful for the extensive ozone trend analyses that have been published in the scientific literature; all analysis results on which this paper is based are reported in the references included in Table 1 and Parrish et al. (2020). The Pacific MBL and Lassen Volcanic NP data in Figure S3 are from Table S1 of Parrish et al. (2021) and the U.S.National Park Service (https://ardrequest.air-resource.com/data.aspx, last accessed 18 February 2020), respectively. This work was not supported by any funding agency, and the authors have no conflicts of interests.

References

Cooper, O. R., et al. (2010). Increasing springtime ozone mixing ratios in the free troposphere over western North America, *Nature*, *463*(7279), 344–348, doi:10.1038/nature08708.

Cooper, O. R., Gao, R.-S., Tarasick, D., Leblanc, T. & Sweeney, C. (2012). Long-term ozone trends at rural ozone monitoring sites across the United States, 1990–2010, *J. Geophys. Res.*, *117*, D22307, doi:10.1029/2012JD018261.

Cooper, O. R., et al. (2014). Global distribution and trends of tropospheric ozone: An observation-based review. *Elem Sci Anth* 2: 29. DOI: https://doi.org/10.12952/journal.elementa.000029

Cooper, O. R., et al. (2020). Multi-decadal surface ozone trends at globally distributed remote locations, *Elem. Sci. Anth.*, *8*, 23. doi.org/10.1525/elementa.420

Gaudel, A., et al. (2020). Aircraft observations since the 1990s reveal increases of tropospheric ozone at multiple locations across the Northern Hemisphere, *Sci. Adv.*, *6*, eaba8272.

HTAP (2010). Hemispheric transport of air pollution 2010, part A: Ozone and particulate matter, air pollution studies no. 17, edited by: Dentener F., Keating T., & Akimoto H., United Nations, New York and Geneva.

Jacob, D. J., Logan, J. A., & Murti, P. P. (1999). Effect of rising Asian emissions on surface ozone in the United States, *Geophys. Res. Lett.*, *26*, 2175–2178, doi:10.1029/1999GL900450.

Jaffe, D., Price, H., Parrish, D. D., Goldstein, A., & Harris, J. (2003). Increasing background ozone during spring on the west coast of North America, *Geophysical Research Letters*, *30*(12), 1613. doi.org/10.1029/2003GL017024

Lin, M., Horowitz, L. W., Oltmans, S. J., Fiore, A. M., & Fan, S. (2014). Tropospheric ozone trends at Mauna Loa observatory tied to decadal climate variability. *Nature Geoscience*, *7*(2), 136–143. https://doi.org/10.1038/ngeo2066

Lin, M., Horowitz, L. W., Cooper, O. R., Tarasick, D., Conley, S., Iraci, L. T., Johnson, B., Leblanc, T., Petropavlovskikh, I. & Yates, E. L. (2015). Revisiting the evidence of increasing springtime ozone mixing ratios in the free troposphere over western North America, *Geophys. Res. Lett.*, *42*, doi:10.1002/2015GL065311

Lin, M., Horowitz, L. W., Oltmans, S. J., Fiore, A. M. & Fan, S. (2017). US surface ozone trends and extremes from 1980 to 2014: quantifying the roles of rising Asian emissions, domestic controls, wildfires, and climate, *Atmos. Chem. Phys.*, *17*, 2943–2970.

Lin, M., Horowitz, L. W., Xie, Y., Paulot, F., Malyshev, S., Shevliakova, E., et al. (2020). Vegetation feedbacks during drought exacerbate ozone air pollution extremes in Europe. *Nature Climate Change*, *10*(5), 444–451. https://doi.org/10.1038/s41558-020-0743-y

Logan, J. A., Staehelin, J., Megretskaia, I. A., Cammas, J.-P., Thouret, V., Claude, H., et al. (2012). Changes in ozone over Europe: Analysis of ozone measurements from sondes, regular aircraft (MOZAIC) and alpine surface sites. *Journal of Geophysical Research*, *117*, D09301. https://doi.org/10.1029/2011JD016952

Oltmans, S. J., Lefohn, A. S., Harris, J. M., & Shadwick, D. S. (2008), Background ozone levels of air entering the west coast of the U.S. and assessment of longer-term changes, *Atmos. Environ.*, *42*, 6020–6038, doi:10.1016/j.atmosenv.2008.03.034.

Parrish, D. D., Millet, D. B., & Goldstein, A. H. (2009). Increasing ozone in marine boundary layer air inflow at the west coasts of North America and Europe, *Atmospheric Chemistry and Physics*, 9, 1303–1323.

Parrish, D. D., Law, K. S., Staehelin, J., Derwent, R., Cooper, O. R., Tanimoto, H., et al. (2012). Long-term changes in lower tropospheric baseline ozone concentrations at northern mid-latitudes. *Atmospheric Chemistry and Physics*, *12*, 11,485–11,504. https://doi.org/10.5194/acp-12-11485-2012

Parrish, D. D., Petropavlovskikh, I., & Oltmans, S. J. (2017). Reversal of long-term trend in baseline ozone concentrations at the North American West Coast. *Geophys. Res. Lett.*, 44. https://doi.org/10.1002/2017GL074960

Parrish, D. D., Derwent, R. G., O'Doherty, S. & Simmonds, P. G. (2019). Flexible approach for quantifying average long-term changes and seasonal cycles of tropospheric trace species, *Atmos. Meas. Tech.*, 12, 3383–3394, https://doi.org/10.5194/amt-12-3383-2019

Parrish, D. D., et al. (2020), Zonal similarity of long-term changes and seasonal cycles of baseline ozone at northern mid-latitudes. *J. Geophys. Res.: Atmos.*, doi: 10.1029/2019JD031908

Parrish, D. D., et al. (2021), Anthropogenic Reversal of the Natural Ozone Gradient between Northern and Southern Mid-latitudes. *Atmos. Chem. Phys. Disc.*, https://doi.org/10.5194/acp-2020-1198

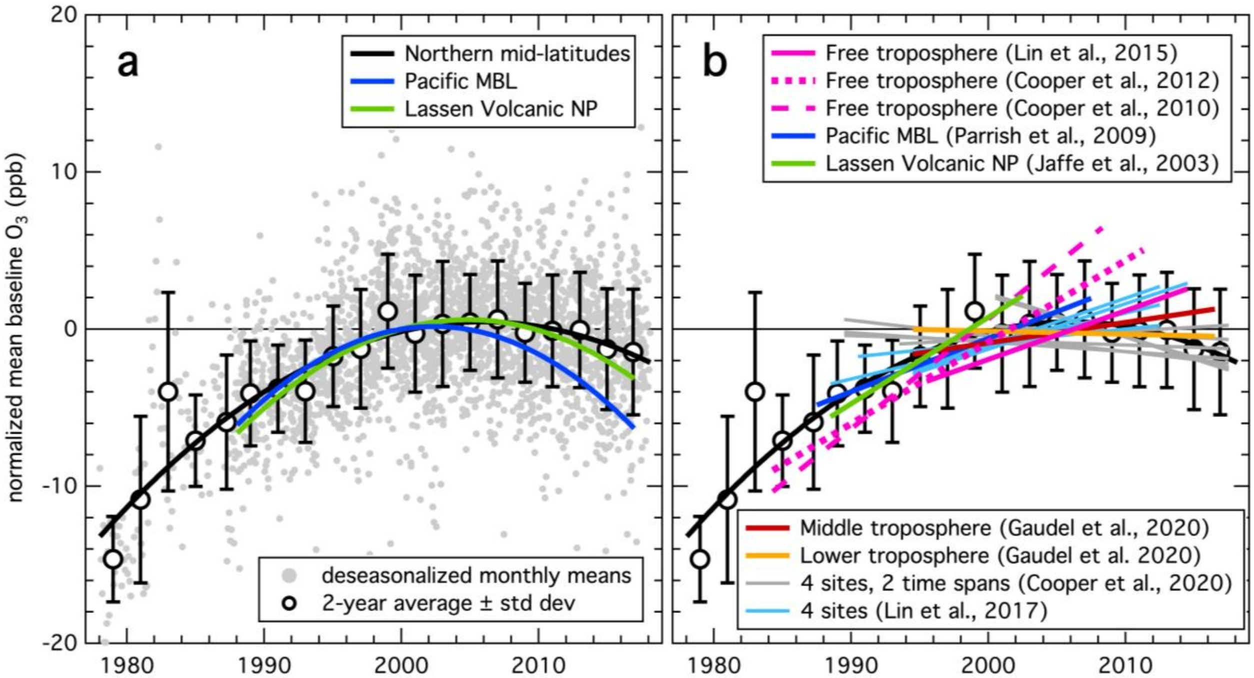

Figure 1. Long-term changes in baseline ozone at northern mid-latitudes. **(a)** Normalized, deseasonalized monthly mean data (gray points) from eight baseline data sets collected at the surface and in the free troposphere in western North America and western Europe. The symbols with error bars are 2-year means with standard deviations of the gray points, the black solid curve is a quadratic polynomial fit to the gray points. The colored curves are quadratic fits from analyses of two western US data sets (Parrish et al., 2020). **(b)** Quadratic fit and 2-year means from **(a)** compared to line segments representing the annotated trend determinations from western US data sets covering varying time periods included in Table 1.

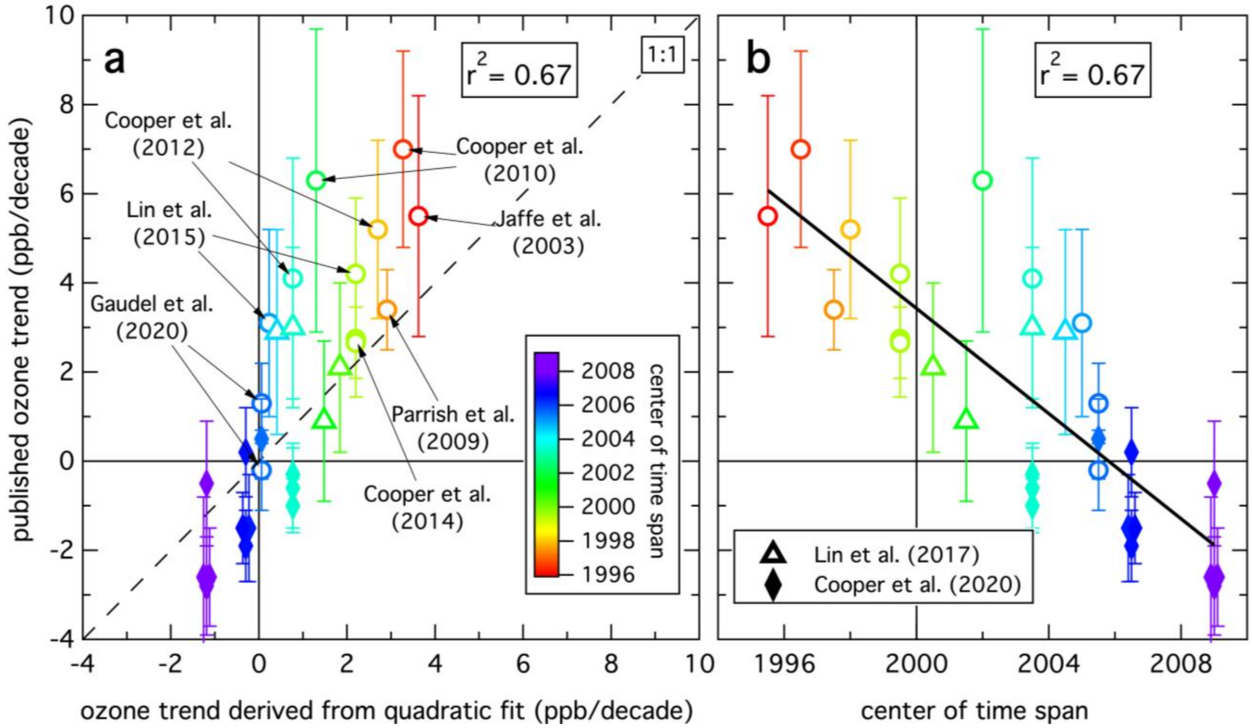

Figure 2. **(a)** Correlation between published ozone trends (Table 1) and those calculated for the same time periods from the long-term northern mid-latitude baseline ozone change quantified by the quadratic fit for northern mid-latitudes (Parrish et al., 2020). Symbols color-coded according to center of time spans of the the reported trends. Error bars indicate the reported 95% confidence limits of those trends. For clarity, some Cooper et al. (2020) symbols are slightly offset along the x-axis. **(b)** Correlation between published ozone trends and center of the time spans of the trend determinations. Symbols are in the same format as in (a). Solid line indicates the linear regression with the square of the correlation coefficient annotated, and t = 0 reference indicated by vertical line.

Table 1. Slopes derived from published trend analyses (with 95% confidence limits) compared with slopes calculated from the quadratic fit to the northern mid-latitude analysis over the same time periods. Studies included report results based on mean or median annual or springtime seasonal data.

Location	*b* (slope) (ppb decade^{-1})	slope from quadratic fit (ppb decade^{-1})	Years of data	Reference
Mid-troposphere	1.3 ± 0.9	0.1 ± 1.1	1994–2016	Gaudel et al. (2020)
Lower troposphere	-0.2 ± 0.9	0.1 ± 1.1	1994–2016	“
Centennial, Wyoming	-0.6 ± 0.9	0.8 ± 0.9	1989–2017	Cooper et al. (2020)
Great Basin NP	0.5 ± 0.9	0.1 ± 1.1	1993–2017	“
Gothic, Colorado	-1.0 ± 0.6	0.8 ± 0.9	1989–2017	“
Grand Canyon NP	-0.3 ± 0.7	0.8 ± 0.9	1989–2017	“
Centennial, Wyoming	-1.5 ± 1.2	-0.3 ± 1.2	1995–2017	“
Great Basin NP	0.2 ± 1.0	-0.3 ± 1.2	1995–2017	“
Gothic, Colorado	-1.9 ± 0.8	-0.3 ± 1.2	1995–2017	“
Grand Canyon NP	-1.5 ± 0.8	-0.3 ± 1.2	1995–2017	“
Centennial, Wyoming	-2.6 ± 1.8	-1.2 ± 1.4	2000–2017	“
Great Basin NP	-0.5 ± 1.4	-1.2 ± 1.4	2000–2017	“
Gothic, Colorado	-2.8 ± 1.1	-1.2 ± 1.4	2000–2017	“
Grand Canyon NP	-2.6 ± 1.1	-1.2 ± 1.4	2000–2017	“
Great Basin NP [1]	2.9 ± 2.3	0.4 ± 1.0	1994–2014	Lin et al. (2017)
Yellowstone NP [1]	2.1 ± 1.9	1.8 ± 0.7	1988–2012	“
Pinedale Wyoming [1]	0.9 ± 1.8	1.5 ± 0.7	1990–2012	“
Mesa Verde NP [1]	3.0 ± 1.8	0.8 ± 0.9	1994–2012	“
Mid-troposphere [2]	3.1 ± 2.1	0.2 ± 1.0	1995–2014	Lin et al. (2015)
Mid-troposphere [2]	4.2 ± 1.7	2.2 ± 0.6	1984–2014	“
Pacific MBL	2.7 ± 0.8	2.2 ± 0.6	1988–2010	Cooper et al. (2014)
Lassen Volcanic NP	2.7 ± 1.3	2.2 ± 0.6	1988–2010	“
Mid-troposphere [2]	4.1 ± 2.7	0.8 ± 0.9	1995–2011	Cooper et al. (2012)
Mid-troposphere [2]	5.2 ± 2.0	2.7 ± 0.6	1984–2011	“
Mid-troposphere [2]	6.3 ± 3.4	1.3 ± 0.8	1995–2008	Cooper et al. (2010)
Mid-troposphere [2]	7.0 ± 2.2	3.3 ± 0.6	1984–2008	“
Pacific MBL	3.4 ± 0.9	2.9 ± 0.5	1987–2007	Parrish et al. (2009)
Lassen Volcanic NP [3]	5.5 ± 2.7	3.6 ± 0.6	1988–2002	Jaffe et al. (2003)

[1] Lin et al. (2017) results are taken from their Figure 13 for stations west of the Front Range of the Rocky Mountains.

[2] Results given for 50th percentiles of the springtime seasonal data sets.

[3] Jaffe et al. (2003) results are 4 season average from full seasonal data sets in their Table 2.

Table 2. Parameter values (with 95% confidence limits) derived from quadratic fits for northern mid-latitudes and for two data sets collected in the western US (Parrish et al., 2020). Intercept and slope are given for the year 2000.

Location	*a* (intercept)[1] (ppb)	*b* (slope) (ppb/decade)	*c* (curvature) (ppb/decade^2)	year$_{max}$	Years of data
Northern mid-latitudes	---	2.0 ± 0.6	-1.8 ± 0.6	2005.7 ± 2.5	1978–2017
Lassen Volcanic NP	41.0 ± 0.7	2.6 ± 0.7	-2.4 ± 0.8	2005.4 ± 2.2	1987–2017
Pacific MBL	32.9 ± 1.1	1.4 ± 1.0	-2.5 ± 1.1	2002.8 ± 2.2	1987–2017
derived from trends	---	3.4 ± 0.9	-2.9 ± 0.8	2005.8 ± 2.2	1984–2017

[1] A value for *a* is not returned for the fit to the normalized monthly means and the linear fit to the published trends.