

Interactive comment on “Increasing ozone concentrations in marine boundary layer air inflow at the west coasts of North America and Europe” by D. D. Parrish et al.

Anonymous Referee #3

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General Comments.

Parrish and co-authors present an analysis of surface ozone data from sites located on or near the west coast of North America. Their goal is to demonstrate that there has been an increase in ozone in surface air entering the west coast of the United States since 1987, substantiating their own earlier studies of trends in spring. The major problem they face is that only one location has continuous data since 1990, Olympic National Park (NP), and it is the least suitable of the sites for extracting “background ozone”, as it is located next to a city of 20,000 and is about 100 km from the west coast. When they filter the Olympic NP data, they remove 95% of the measurements. Two

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sites have a few years of data starting in 1987, Point Reyes and Redwood NP; 90% of the Redwood data is filtered out. Trinidad Head has data for the end of the period, since 2002. Taking data from all sites together, the filtered data show an increase of 0.45 ppb/yr in winter and spring, 0.25 ppb/yr in summer, and no significant trend in autumn. The trends are 1-1.5%/yr.

Much of the paper is devoted to the approach used to filter the data to extract what the authors call "ozone in the marine boundary layer" (MBL). They are trying to remove any data impacted by surface deposition over land or by photochemical sources and sinks associated with U.S. emissions. They ultimately use wind speed and wind sector to filter the ozone data. They justify this approach by analyzing from 5 weeks of measurements in spring 2002 at Trinidad Head, when measurements of ozone, radon (Rn), MTBE (a short lived soluble hydrocarbon in gasoline) and CO₂ were available. Continental influence is shown by high Rn, MBTE, and CO₂, all of which show decreasing ozone as these species increase above some threshold (because of surface deposition and photochemistry). They deem as marine air the region where ozone is approximately constant at low values of Rn, MBTE, and CO₂. In this regime, ozone is 30-52 ppb, and similar values (27-52 ppb) are found for NW winds greater than 3 m/sec. With this selection on wind speed and direction, they obtained almost the same mean ozone value (3% less) as using the trace gas data selection, while if they used only wind direction, the mean ozone was 6% less than that given by the tracer data. They then use the wind speed and sector to select data from other locations without trace gas data. They further use the small diurnal variation of ozone in the 5 week period with their wind selection criteria to argue that it is valid.

The measurement locations are over 1000 km apart. The authors use the small mean differences between simultaneous data at pairs of sites to show there does not seem to be a latitudinal gradient (although there are substantial differences for individual years, see Figure 6). They also show the wind speed distribution varies considerably among sites (Figure 5b), and that is it the lack of data at higher wind speeds that eliminates

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most of the Olympic data. Figure 5a shows that for wind speeds of 3-5 m/sec, the mean ozone at Olympic for 1991-2004 is the same as that at Trinidad Head for 2003-2006, implying little trend for these winds speeds, but the authors do not comment on this.

The authors use a threshold of 3 m/sec (along with NW winds) to filter all the sites except Olympic, for which they use 2 m/sec (to avoid eliminating even more than 95% of the data). They need to comment on whether this introduces biases when forming seasonal means. Are there enough data left each month to form meaningful seasonal averages? The authors should give the requirements for a month or season of data to be included in their analysis, in terms of the number of hours of data available, and whether data was required in each of the three months in a season. Or they should state that they did not have any such requirement, if this is the case.

In Section 4, the authors choose to compute trends based on seasonal averages of the filtered data, a very simple approach. They comment that this avoids "the deseasonalization" process employed in some work, which introduces unneeded complexity and potentially can confound the trend analysis if different seasons exhibit different temporal trends. This is a strange statement, as there is no reason why a more sophisticated approach has to "confound" the analysis when different seasons have different trends. There are well-known statistical approaches to determine trends in seasonally varying data, by fitting the seasonal cycle and seasonal (or monthly) trends simultaneously. These methods are standard practice for stratospheric ozone trend analysis, and also have the advantage of giving a robust methodology for estimating for annual trends and their errors from the seasonal trends. Such methods are described in the WMO Scientific Assessment of Ozone Depletion: 1998, Chapter 4.

The key result of this paper is shown in Figure 7, which plots the seasonal means for each site for each year. This figure must be improved by increasing its size so it spans the page (vertically and horizontally) and by adding the standard deviation for each season. A similar plot must be included that shows the time series for all the data at each site. This is shown only for Olympic NP in spring in Figure 8. The trend analysis

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on the seasonal means shows that there have been statistically significant increases in the filtered ozone data in winter, spring, and summer.

On p. 13878 the authors report annual mean trends based on 12 month running means. As Figure 7 shows there are no summer data for 1994-2001, the authors need to clarify how they calculated 12 month running means for the entire record (shown in Figure 12).

On p. 13876, the authors use minimum values of ozone during southwest flow events to argue that there has been an increase in minimum ozone values during such events. I would expect that such minimum values would depend on how deep into the tropics the origin of the air mass was. Also there is no reason to expect that such air masses would be influenced by export of Asian emissions. One might imagine that any increase in minimum values seen in southwest flow would be a result of changes in dynamics, rather than changes in photochemical production of ozone. The authors quote Oltmans et al. (2008) as saying such events occur in the Trinidad Head record since 2002 at least once each spring, and that the ozone concentrations drop to near 30 ppbv. In fact Oltmans et al. show a figure of one event in 2007 with mean values dropping to 30 ppb, but hourly values as low as 15 ppb, and they say there is one event each spring; however they do not give the minimum values in the other events. Oltmans et al. also note that there were no such events during the ITCT-2K2 campaign in spring 2002, which is interesting as Parrish et al. use the minimum values observed during ITCT-2K to indicate an increase in ozone minima. Parrish et al. need to revise this discussion to focus more on issue of interannual variability in dynamics confounding the trend in minimum values, as southwest flow events are rare. Alternatively, drop this section (5.3). I see no evidence presented in this paper to support their conclusion "A striking feature of these events has been the generally rapid increase of these minimum ozone concentration", as they have not conducted a detailed analysis of such events, which requires a back-trajectory analysis.

At the end of the paper, the authors argue that their filtered data provide a test of

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hindcasts by global chemistry models. In fact, such models use fairly coarse spatial resolution, especially for 20 year runs (200-500 km grids), so would not necessarily be reliable for filtering the model results using surface wind and direction, especially if (as argued by the authors on p. 13859) they do not capture orographic effects along the coast. By providing the monthly means of the unfiltered as well as filtered data for each site, the authors would enable modelers to use their data in the way they think best.

Overall, the authors make their point in this paper, that there has been an increase in their filtered data on the west coast of the U.S. A few things however left me uneasy about this study and paper. One is the reliance on data from a clearly unsuitable site, Olympic NP, a nowhere near the west coast. Based on the Google map location of the Visitors Center (where the data are taken), flow from the NW sector has passed over the city of Port Angeles and a highway before reaching the site, and only 2.4-7.5% of data are retained, depending on season. This is mitigated by the fact that the other sites give ozone mixing ratios similar to the Olympic data during the brief periods of overlap.

My second concern is the negativity about other work. One example is the comments on trajectory based selection approaches, the other is the comments on the paper by Oltmans et al. (2008), one of which was addressed above. The authors note that there is mostly flow from the NNW because of orographic effects (p. 13859). They also argue that because the orography is not resolved by the meteorological data used to drive trajectory models, that such trajectory analyses are therefore not useful for selecting marine air at the coastal sites. I do not think that the use of surface wind data is necessarily any better or worse than the use of trajectories, as the local surface wind at the time of the ozone measurement does not give any information on the history of the air parcel. For example air could have circulated over the continent as the air parcel moved around the Pacific High, before descending into the boundary layer. Figure 2 shows quite clearly a huge range in ozone values, likely because of very different air mass histories before the air parcel arrived at the surface site.

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The authors criticize the work of Oltmans et al. (2006, 2008, hereinafter Oltmans.06 and Oltmans.08) in several places. They should be more careful of their facts when doing so. For example, they state (p.13853, l.18-20) that Oltmans.08 suggest Yreka, CA, is "useful for characterizing the marine background ozone flowing into North America". I could find no such statement in Oltmans.08. Oltmans.08 state that Trinidad Head is a great site for this purpose - no argument here; they state in their introduction: "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." They conclude "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 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." They are just saying that this site does not see an effect of changing background ozone. It seems that Parrish et al. would agree with this, as they feel the site is influenced by other factors. In fact Oltmans.08 state in their Abstract "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."

On p. 13861, l. 27, Parrish et al. state "It should be noted that simply selecting a northwest wind direction as proposed by Oltmans.08 is not adequate for isolating marine air." I could find no such statement in Oltmans.08. Oltmans.08 plot wind roses of ozone for April and August at Trinidad Head, with day and night ozone values shown, but do not discuss selecting on wind direction. In fact, Section 3.2 "Air flow and selection of background measurements" of Oltmans.08 does not even discuss the selection criteria, which is a bit odd. Their Fig. 11 comparing Trinidad Head to the Channel

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Island site, and Fig. 18 comparing individual years at Trinidad Head both use daytime data (12-18 LST), so it looks like they did not use wind speed at all, except in their discussion. In fact, Fig. 4 of Parrish et al. of the diurnal cycle shows that selecting just data from 12-18 LST is a pretty similar their selection criteria during these hours, but they could check this.

Clearly Parrish et al. need to read Oltmans.08 more carefully, and not mis-quote it. They need to check every attribution to Oltmans.08 for accuracy. I did not check their detailed comments on Oltmans.06, but given the misleading statements about Oltmans.08, they need to check that also.

Detailed comments:

1. The title should not include Europe. The authors do not analyze data from Europe, they merely compare their results from North America to published results from Europe.

Do not refer to ozone "concentrations" when the ozone is always given as mixing ratios.

p.13849, l.7. The text states "In about 1970 emissions from Europe and North America began to stabilize and since have decreased (RETRO, 2007)." This is misleading, as it does not make clear when the decrease started. The EPA Trends data (<http://www.epa.gov/ttn/chief/trends/index.html>) shows that NOx emissions were essentially stable from 1970 to 1995, but have decreased almost 30% from 1995 to 2005. Regulation of emissions started later in Europe than in the U.S., and emissions there increased until about 1990, then started to decrease. So the statement about European emissions is just wrong.

p. 13850, l.25. The text states that the large region upwind of the West coast of N. America "provides the opportunity for the ozone distribution to homogenize to the extent possible .." Ozone is pretty variable on the west coast, as shown by Figures 2 and 3, and by the Trinidad Head sonde data (see Oltmans et al., 2008). So this is a strange statement. In fact the entire discussion (lines 25 on) about "regional integration" is a bit

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simplistic when the data defined later as background show a range of 30-52 ppb. Also for the authors to state that west coast data provide ozone without . . . "transport of air masses with widely varying history" is not correct, as shown by Figure 7 of Oltmans et al. (2008). Careful analysis of aircraft data has shown that the ozone production is very different for air masses that take the most northerly route across the Pacific in spring, and those that take a more southerly route (e.g., papers by Heald et al., 2004, JGR; Hudman et al., 2004, JGR). Clearly they cannot really expect ozone to be "homogenized".

p. 13851, l. 13. Which two marine environments? The paper is about North America.

p. 13853, l. 10. Add the distance from the west coast to Table 1 for each site.

p. 13854, l. 27 to l12 of the next page. Drop the discussion of the virtues of island sites, since none of the data in the paper are from island sites.

p. 13859, l. 21 on. The authors compare the CO₂ with wind selected data with the average of flask data for the same period. They should agree, and the criteria for the flask data must also be based on wind sector, and perhaps speed; just find out if this is the case from the GMD group.

p. 13874, l. 16-19. The comments about the data adjusted to 2000 being lower than Oltmans.08 plot for 2002-2007 is inaccurate, Ozone in 2000 looks very like that in 2004, hardly surprising given the obvious interannual variability.

In several places, the authors use the term "background" ozone. They need to define what they mean by this.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 13847, 2008.

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