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Atmospheric Chemistry and Physics Discussions

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.

D. D. Parrish et al.

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The authors are grateful to the referees for the time and thought that they put into their reviews and comments regarding our paper [Parrish et al., 2008]. The revised manuscript is now significantly improved as a result of our response to some of their comments. We rebut other of their concerns below; this rebuttal helped us to review and refine our analysis and further improve the paper.

General Comments:

1) Contradictory conclusions have been reached by two pairs of papers regarding long-term trends in background ozone concentrations reaching the west coast of North America. Two papers conclude that significant recent increases had occurred and were continuing at the time of writing:

Jaffe et al. [2003] in their paper entitled "Increasing background ozone during spring on the west coast of North America" state that "O_3 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."

Parrish et al. [2004] in their paper entitled "Changes in the photochemical environment of the temperate North Pacific troposphere in response to increased Asian emissions" state that they "... provide substantial evidence that a marked change in the photochemical environment in the springtime troposphere of the North Pacific is responsible for this increased O_3."

In contrast two other papers argue that no significant changes had occurred, at least recently:

Oltmans et al. [2006] in their paper entitled "Long-term changes in tropospheric ozone" state that 1) "For the approximately 15-year data records at each site, only Olympic shows a single month (February increase) with a significant change indicating that for these more remote sites there have not been important recent changes, related to either local or distant sources." (in reference to several National Park sites in the western US). 2) "The Yreka record suggests that longer-range effects may not be playing a major role, however, since no spring increases are noted." and 3) "It appears from the records considered here that there has been a leveling off of recent increases in the tropospheric ozone burden, particularly at mid latitudes of the N.H. that have been most affected by anthropogenic changes."

Oltmans et al. [2008] in their paper entitled "Background ozone levels of air entering the west coast of the U.S. and assessment of longer-term changes" state that "This at least suggests that for the months of strongest transport across the Pacific (winter and spring) that for this site there likely has not been significant impact of changing background ozone amounts reaching the west coast of the U.S." (in reference to the

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Yreka, California data record.)

These two groups of statements cannot both be correct. It is important that these contradictory conclusions be resolved so that observed ozone trends can be used as tests of models of the tropospheric ozone budget, and indeed to improve our understanding of this budget. In the paper under review [Parrish et al., 2008], hereinafter referred to as PMG.08, one of our goals is to provide a critical review of existing data sets and the methods that have led to these contradictory statements. Many scientific controversies can be resolved by additional experimental studies. However, the present contradictions involve interpretation of historical data sets, and it is obviously impossible to now increase the historical ozone data record. Thus, resolution of the contradictory findings necessarily requires direct, objective statements regarding the suitability of the underlying data sets and the validity of the analyses applied to these data sets in both pairs of papers. In this process we have strived to avoid a strident tone and unnecessary disparagement of any work. We have revised our PMG.08 to maintain as courteous a tone as possible throughout. While we cannot claim to have presented the "last word" in this discussion, we have presented as definitive an analysis as we can devise based on the available data sets. This review strongly supports the conclusions of the former two papers. (It should be noted that the authors of PMG.08 are also coauthors of the former two papers.) The three referees of this paper challenge the definitiveness of the analysis of PMG.08 to a greater or lesser degree. These general comments are intended to rebut those substantive challenges as clearly, definitively and objectively as justified by the available data and analyses. In this response, we refer to figures and tables in PMG.08 by number, and additional figures presented here are labeled by R1, etc. These additional figures are posted online at: http://esrl.noaa.gov/csd/pubs/acpd_8_13847to13901_2008/refereeresponsefigs.pdf

2) On pages S8486-S8487 Referee 1 notes that our analysis includes the Olympic National Park data set (among 4 other marine boundary layer sites), and that we extensively criticize the Yreka data set, upon which Oltmans et al. [2006, 2008] focus their attention. Referee 1 asserts that both of these sites are equally questionable. However, this assertion is not correct. PMG.08 demonstrate that the Olympic NP data, when properly filtered for marine air, give results consistent with three other marine boundary layer (MBL) sites. This consistency is found for absolute ozone concentrations measured in the same season and year (Fig. 6 and associated discussion), long-term temporal trends (Fig. 7 and associated discussion), and seasonal cycles (Fig. 10 and associated discussion). In contrast the Yreka site is located inland in California in a valley behind the coastal mountain ranges; hence it never directly receives marine air without continental influences, and these continental influences are very pronounced at Yreka due to large local emission sources that strongly affect the measured ozone concentrations. As discussed in PMG.08, and illustrated in Fig. R1 here, ozone concentrations at the Yreka site exhibit a very different seasonal cycle compared to that found at the MBL sites. The Yreka seasonal cycle shows a wintertime minimum with much lower ozone concentrations (presumably due to reaction with local NOx emissions) and a summertime maximum with significantly higher ozone concentrations (presumably at least partly due to photochemical ozone production from local emissions of ozone precursors). The MBL ozone concentrations exhibit a springtime maximum and a summertime minimum. As the referee notes the results of the trend analysis of PMG.08 is similar to those of the analysis of Oltmans et al., 2008, but the referee fails to note the critical point that these Yreka trends are not characteristic of ozone concentrations in the marine air inflow.

3) On pages S7017 and S7021 Referee 3 also expresses concerns about the use of data from the Olympic National Park site. The most significant criticism is that the site is about 100 km from the west coast. While true, this does not indicate that the site is isolated from direct inflow of marine air from the MBL. As Fig. 1 shows, the site is located on the shore of the Juan de Fuca Strait, which exposes it to direct airflow from the Pacific. Fig. R2, to be included in the revised PMG.08 as Fig. 3b, shows an example wind rose for Olympic NP in the format of Fig. 3a. The high winds occur from the west-northwest, which is the orientation of the Juan de Fuca Strait. Hence, the ozone concentrations selected by the wind criteria do represent direct Pacific airflow through the Juan de Fuca Strait to the site. Although the site location is certainly not optimal for sampling marine air, the Olympic NP data are nevertheless useful for investigating MBL ozone concentrations. Some of this discussion has been added to our revised manuscript.

4) All three referees (Referee 1 on pg. S8488, Referee 2 on pg. S8600, and Referee 3 on pg. S7019) express concerns about the small fraction of data from Olympic NP that meet our criteria for MBL ozone concentration analysis. For example, during spring 2003 (the data in Fig. R2) only 72 hours meet the wind selection criteria. This corresponds to 3.2% of the total hours in the 3-month period and 3.6% of the time covered by the available data. However, the diurnal land-sea breeze cycle plays a significant role in determining wind speeds. The high, northwest winds occur predominately for a few hours in the afternoon. Those 72 hours of selected data represent 17 different days in all three months of the season. It would be desirable to have data from all days available, but the 17 days with suitable data nevertheless provide a reasonably representative sample of days, a sample that compares favorably with the once per week schedule often employed for ozone sonde launches from the few intensive sites conducting such launches. Some of this discussion has been added to our revised manuscript. Most importantly is the comparison in Fig. 6 between the Olympic NP measurements and those from the other sites. That comparison, which includes 22 separate seasonal averages with measurements at Olympic NP and another site, demonstrates that the Olympic NP data have no significant average bias (-0.23 $\pm$ 0.78 ppbv) where the 1 σ confidence limit is indicated. These considerations justify the lack of any specification of a minimum number of available days to include a specific seasonal average.

5) PMG.08 conclude that large-scale trajectory calculations are not adequate for avoiding time periods when continental effects significantly affect observed ozone concentrations at the MBL sites studied. PMG.08 further conclude that selection according

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to criteria based on local wind speed and direction is superior and necessary (at least when measurements of continental tracers are unavailable). All three referees (Referee 1 on pg. S8487, Referee 2 on pg. S8599, and Referee 3 on pg. S7021) express doubts about these conclusions. The line of reasoning that leads to these conclusions is quite simple: First, when air approaches the site from the land, ozone concentrations are substantially reduced by deposition to continental surfaces and reaction with continental NOx emissions. Second, the land-sea breeze circulation and other orographically driven circulations frequently bring air from the land to the MBL sites, predominately during nighttime and early morning. Third, local winds provide an effective tool for screening out the air directly transported from the land. Fourth, the land-sea breeze circulation and other orographically driven circulations have spatial scales on the order of 10s of km, while the meteorological fields upon which the large-scale trajectories are based have spatial resolutions on the order of 100s km; hence, large-scale trajectories cannot possibly provide discrimination against the effects of the smaller scale circulations. This reasoning is unchallenged by any of the doubts expressed by the referees. PMG.08 do not argue that the large-scale trajectories are not useful; they do provide important indications of the history of sampled air masses over the previous several days. The Pt. Arena study [Parrish et al, 1992], which provided one of the data sets analyzed by PMG.08, provides a useful example of these issues. The large scale trajectories presented in that work indicate strong marine air inflow throughout the study period, but only 61% of the data satisfied the wind selection criteria. The rejected measurements often showed substantially depleted ozone concentrations due to local continental effects.

6) PMG.08 rely on very simple trend analysis, and argue that this analysis is the preferred method for determining the temporal trends of ozone in each season, and is more appropriate than the approach of Oltmans et al. [2006, 2008]. All three referees (Referee 1 on pg. S8487, Referee 2 on pg. S8600, and Referee 3 on pg. S7019) express doubts about these arguments, and some suggest that a more sophisticated approach may be preferred. Referee 3 suggests Chapter 4 of the WMO Scientific

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Assessment of Ozone Depletion: 1998 as a useful guide. Although the WMO work deals with trends in stratospheric ozone and the present work deals with tropospheric ozone, we do find this material helpful. The first section of that chapter discusses eight "necessary or desirable terms" in the statistical model (referred to below as the WMO elements). Below we evaluate the PMG.08 approach with respect these WMO elements and briefly identify them in bold print.

To define each temporal trend, PMG.08 seek the average yearly increase in ozone concentration in that season over the period of the data record, and the intercept in a reference year, arbitrarily chosen as 2000. We apply a simple linear regression analysis to 3-month seasonal averages of the marine data selected by the wind criteria. Only two parameters are extracted from each seasonal data set. The **seasonal means** and **seasonal trends** are properly treated. Notably, our approach allows the derived trends to vary by season. Methods that attempt to "deseasonalize" a data record with a seasonal term that does not vary over the time span of the data record are handicapped in this regard. The only **ancillary variable** we treated is wind speed, but this is the only one justified, because (as discussed below) the residuals of the data about the derived trend are only as large as expected from the 1 σ confidence limits of the seasonal average determinations. **Autoregression** of the residuals about the trend are not treated, but inspection of Figure 7 shows that autoregression of the residuals between successive years is not discernable. In Table 5 the $\chi\nu$ statistic does not show a strong seasonal cycle, so different **weighting according to season** is not necessary. **Dynamical variables** are not considered, and the WMO reference warns that "use of these variables in regression models where the primary purpose is to estimate ozone time trends leads to difficulties in interpretation of the resulting trends in ozone." **Standard errors for seasonal trends** are calculated properly; PMG.08 do use unweighted fits, but since the $\chi\nu$ statistic closely approximates the 1 σ confidence limits of the seasonal averages, the standard errors of the parameters derived from the trends analysis are indeed robust. The PMG.08 approach is in accord with each of the first seven WMO elements, and the eighth (**model variations**) is not relevant here.

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7) One major goal of PMG.08 is to carefully characterize a feature of the tropospheric ozone distribution that will challenge our knowledge of that distribution as it is incorporated into global chemical transport models. The temporal trend of ozone in the marine environments, including the MBL, off the west coasts of the northern mid latitude continents is such a feature. It is an important feature, because it can be easily extracted from model output. Referee 1 (on pgs. S8488-S8489) and Referee 3 (on pgs. S7020-S7021) both argue that "real" or "unfiltered" data would provide better comparisons. We disagree. The global models, operating on grid scales on the order of 100 km, cannot resolve the manifold subgrid scale processes that affect the measured ozone concentration at any given continental site. An excellent example of comparing global model ozone and CO output with observations at the Trinidad Head site, while filtering out local influences from the subgrid scale processes of local emissions and deposition, is described in Goldstein et al. [2004]. The CO and O_3 GEOS CHEM and MOZART model temporal variability matches the hourly observations well when local scale influences are removed using either wind direction and speed or chemical tracers to filter the observations. Any means of simplifying the comparison, such as using measurements to quantify the temporal trend of ozone concentrations in marine environments, will lead to much more robust comparisons. These comparisons can be conducted in several ways. For example, model results do not have to be taken from a single grid cell where a site is located, but can be averaged over a number of grid cells near North America, but far enough off shore to be isolated from the continental effects. Alternatively, such models have the ability to examine inflow ozone by tagging ozone from different sources.

Point by Point Responses to Reviewer Comments:

Referee 1:

- Pg. S8486, para. 1-3: Response given in General Comments 1, 2, and 6.

- Pg. S8487, para. 2: The Lassen Volcanic NP site does receive direct inflow of marine

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air under some conditions because it is located at an elevation considerably higher than the intervening coastal mountain ranges. In contrast, although closer to the coast, Yreka is located in a valley behind the coastal ranges and thus cannot receive direct inflow of marine air.

- Pg. S8487, para. 3: Oltmans et al. [2008] use several approaches for investigating ozone in marine air. They do use calculated back trajectories in some places (e.g. discussion in Section 3.2). They also use an "off the ocean" wind direction (see discussion in Section 3.1 including the wind roses in Fig. 5 and the diurnal cycles in Fig. 6) and the selection of daytime data (see discussion in Section 2, and the discussion of Figs. 9 and 11 and the trend analysis shown in Figs 18-20) as representing marine air. We have modified our wording to more accurately describe the methods of Oltmans et al. [2008]. The remainder of this comment is addressed in General Comment 5.

- Pg. S8488, para. 1,2: Response given in General Comment 4.

- Pg. S8488, para. 3: Response given in General Comment 5.

- Pg. S8488, para. 4: Jaffe et al. [2003] and Parrish et al. [2004] based their analysis on aircraft data and measurements from an elevated site as well as MBL data. PMG.08 give detailed discussion of only MBL data. The sentence questioned by the referee is included to assure the reader that, at least so far, there is no reason to question the earlier analysis of aircraft and elevated site data by Jaffe et al. [2003] and Parrish et al. [2004] even though they are not discussed in this paper. This comment is retained.

- Pg. S8488, para. 5: The point of Fig. 12b is to put the present results into the widest possible temporal perspective. The many other sites to which the referee refers are not included for several reasons including that they do not span as long a time period, and they are not representative of large regions as are the earlier and the present data sets. We agree that transport models are capable of simulating many features. There are, though, real questions regarding the accuracy of those simulations, including that of the long-term trends. The remainder of this comment is addressed in General Comment

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7.

Referee 2:

- Pg. S8599, para. 1: Response given in General Comment 5.

- Pg. S8599, para. 2: As PMG.08 discuss in the paragraph beginning at the bottom of pg. 13868, the wind selection criteria do not strongly affect the trend derived from either the Olympic NP or the other four MBL sites. Figure 8 shows this for Olympic NP and Fig. R3 illustrates this for the combined data from the other four sites. Figure R3 is now included as a second panel of Fig. 8 in our revised paper. The referee's argument in this comment is simply not correct.

- Pg. S8600, last comment: Response given in General Comment 4. We do not wish to include the wind speed in the statistical model, because our goal is to determine not only the trend, but also the best estimate for the absolute ozone concentration so that we can quantitatively compare the North American concentrations with the European concentrations in Figures 7 and 11-12.

Referee 3:

- Pg. S7019, para. 1: Response given in General Comment 4. We have included no requirement on either the number of hours of available data or on the availability of data in each month of the season, beyond the exclusion of seasons when the wind or ozone instruments were not operational for more than one-half of the season.

- Pg. S7019, para. 2: Response given in General Comment 6.

- Pg. S7019, para. 3: Figure 7 has not been changed. A nice feature of on-line journals such as ACP is that the figure can be sized as desired on the reader's computer monitor. The standard deviations are not added, since they have no relevance except how they affect the confidence limit of each seasonal average. As far as we can tell, there are no significant variations in these confidence limits. A second panel has been added to figure 8 showing the time series for all the data at the four other MBL sites in

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spring.

- Pg. S7020, para. 1: Table 5 shows that there are indeed summer data for Olympic NP, but they are not adequate to quantify a summertime temporal trend and hence they are not included in Figure 7. We have added these data to the seasonal cycle in Figure 10, and have clarified how we calculated 12 month running means.

- Pg. S7020, para. 2: The southwest flow events are thoroughly discussed in Parrish et al. [1992, 2004a], not in PMG.08. A southwest flow event similar to the one described by Oltmans et al., [2008] was indeed observed during ITCT 2K2, and is discussed by Parrish et al. [2004a] (see Fig. 5 in that paper). The analysis in that paper does not need further justification here. The quote from PMG.08 questioned by the referee is amply supported in Parrish et al. [2004a]. The referee asserts that during the example event discussed by Oltmans et al. [2008], the ozone concentration dropped to hourly values as low as 15 ppbv. However, this is an excellent example of the necessity of examining local wind speed and direction to select marine air. Figure R4 shows that even though the long-range trajectories illustrated by Oltmans et al. [2008] (Fig. 10) indicate marine air flow impacting the site, the local winds show that local continental influence accounts for the 15 ppbv noted by the referee. Such low ozone values are no longer observed in marine air inflow at the west coast of North America, at least in spring.

- Pg. S7020, para. begining at the bottom of the page: Response given in General Comment 7.

- Pg. S7021, para. 1: Response given in General Comment 3.

- Pg. S7021, para. 2: Response given in General Comments 1 and 5. Also the discussion above regarding this referee's comment regarding southwest flow events shows the need for local wind analysis.

- Pg. S7022, para. 1: Response given in General Comment 3. We do make the state-

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ment that Oltmans et al. [2008] suggest that the Yreka site is useful for characterizing the marine background ozone flowing into North America, but we do not imply that it is an exact quote. We do believe that it is an accurate characterization of that paper, since it is the only trend analysis that they perform that supports their conclusion that "there likely has not been significant impact of changing background ozone amounts reaching the west coast of the U.S." and they do title their paper "Background ozone levels of air entering the west coast of the U.S. and assessment of longer-term changes"

- Pg. S7022, para. 2: Response given in response to Referee 1 Pg. S8487, para. 3 comment. Again, we do not imply that we give an exact quote from Oltmans et al. [2008], but we do believe that we give an accurate characterization of their approach. The referee is correct that selecting daytime data (12-18 LST in many places, 11-19 LST in Fig. 9 and associated analysis) as done by Oltmans et al. [2008] often gives similar results to the wind selection criteria. We attribute this similarity to the land-sea breeze circulation that tends to give stronger onshore winds in these daytime periods. However, we believe the wind criteria selection is superior because it focuses upon the mechanism responsible for bringing continentally influenced air to the site. The selection by wind criteria includes periods of strong onshore airflow outside of the afternoon time window, and eliminates offshore flow that frequently occurs even during the afternoons.

- Pg. S7023, para. 1: We have carefully read both Oltmans et al. [2006, 2008]. Any direct quotes from those papers are enclosed in quotation marks. Where we give statements describing the approaches and techniques used in those papers we believe that we have been accurate.

Detailed Comments: - Title of paper - The similarity of the trends seen in the MBL at the west coasts of Europe and North America is one of the major results reported in our paper. Thus, we have not changed the title of the paper. It is true that we did not analyze the European data ourselves. This was not necessary since Derwent et al. [2008] had already presented an analysis of those data comparable to our own

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approach.

- Use of the term "concentrations" - We prefer to use "concentrations" rather than the more ambiguous term "levels". We take the chemical definition of concentration as "the amount of a specified substance in a unit amount of another substance". Thus "mixing ratio" is one acceptable quantitative way to express "concentration". We have not changed this usage.

- Description of time evolution of emissions (p.13849, l.7) - In the first paragraph of the Introduction (to which the referee refers) we give a very concise summary of the temporal changes in emissions from the three major source regions (North America, Europe and Asia) at northern mid-latitudes. The description is accurate, even if it is a very simplified description of a complex issue. In particular, the referee makes the unreferenced assertion that European emissions continued to increase until about 1990. According the RETRO emission inventory, which we judge to be the most accurate retrospective analysis of European emissions and upon which we based our summary, European emissions began decreasing about the same time (in the 1970s) as USA emissions (e.g. see figures on pg. 45 of "Emission data sets and methodologies for estimating emissions: Work Package 1, Deliverable D1-6" available from http://retro.enes.org/reports/D1-6_final.pdf). Consequently, since our statement regarding European emissions is in agreement with the RETRO inventory, we have made no change.

- Variability of ozone in marine environments? (p. 13850, l. 25) - We recognize that the marine ozone concentrations are variable. However, air that has spent long periods in a marine environment has not been recently influenced by the processes that contribute to the much greater variability of ozone concentrations in continental environments. This lower variability allows long-term trends to be quantified more accurately and precisely. The discussion has been modified to make this point more clearly.

- Two marine environments? (p. 13851, l. 13) - This paper discusses both North

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America and Europe. No change made.

- Include distance from coast for each site (p. 13853, l. 10) - As discussed above in General Comment 2, the distance to the west coast is not necessarily a good measure of exposure to marine air. This distance has not been added to Table 1. An interested reader can consult a map to determine this distance, since the precise latitude and longitude are given in the table.

- Discussion of island sites (p. 13854, l. 27 to l. 12 of the next page) - The discussion of island sites is necessary to indicate why detailed analysis is required for data sets collected at coastal sites. No change has been made.

- Comparison of wind-selected and flask CO_2 measurements (p. 13859, l. 21) - We agree that the comparison of CO_2 data sets should agree. The fact that they do agree is a useful test that the wind selection criteria used in this work is consistent and as effective as the criteria used in filling the sampling flasks. No change has been made.

- Seasonal cycle adjusted to 2000 lower than Oltmans et al [2008] result (13874, l. 16-19) - The referee is correct that the year with the lowest observed ozone (2004) of the years presented by Oltmans is similar to our results detrended to the year 2000. The discussion has been clarified.

- Definition of "background" ozone term - In the introduction we now define this term.

References:

Goldstein, A.H., D.B. Millet, M. McKay, L. Jaegle, L. Horowitz, O. Cooper, R. Hudman, D.J. Jacob, S. Oltmans, and A. Clark, Impact of Asian Emissions on Observations at Trinidad Head, California, during ITCT 2K2, J. Geophys. Res., 109, D23, D23S17, 10.1029/2003JD004406, 2004.

Parrish, D. D., D. B. Millet, and A. H. Goldstein, Increasing ozone concentrations in marine boundary layer air inflow at the west coasts of North America and Europe, Atmos. Chem. Phys. Discuss., 8, 13847-13901, 2008.

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