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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Long-term changes in lower tropospheric baseline ozone concentrations at northern mid-latitudes" *by* D. D. Parrish et al.

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The authors are grateful for the time and thought that Dr. Jennifer Logan put into her review and comments [*Logan*, 2012] regarding our paper [*Parrish et al.*, 2012]. We incorporate most of those comments into our revised manuscript, which has led to substantial improvements. Other suggested changes are not made for the reasons discussed below. Our responses to all comments follow. The original comments from *Logan* [2012] are in *italics* and our responses in plain text.

Parrish et al. present a review of trends in ozone at selected surface sites in the northern mid-latitudes, expanding on work presented in the HTAP report (2010), and in previous publications by the authors (Parrish et al., 2009; Cooper et al., 2010; Tanimoto et al., 2009; Gilge et al., 2010), and by others (see below). Thus there is not a lot

that is new in this work, and its main purpose is to present an analysis of data from various sites in one paper, sometime updating the published records by a year or two. In terms of scientific results, the paper is adequate for publication in Atmos. Chem. Phys. as a summary of the authors' own work on trends at the selected sites, except for the analysis of data for Japan (discussed below). However, in its present form it does not meet conventional standards for a scientific paper, and it requires significant improvement before being acceptable for publication.

We find this comment especially useful. *Parrish et al.* [2012] is not simply a review, but rather a synthesis of the absolute ozone concentrations at northern mid-latitudes, the changes in these concentrations, and the changes in the rate of change of the ozone concentrations. Importantly, the discussion of the high degree of similarity of historical ozone changes throughout the northern midlatitudes is new. The specific ozone changes have been discussed in previous publications, but generally the emphasis has been that the "picture of long-term tropospheric ozone changes is a varied one in terms of both the sign and magnitude of trends ..." [[*Oltmans et al.*, 2006]. The synthesis that we present here is derived from treating all of the selected data sets with a consistent analysis approach, and is our major focus. It is clear from Dr. Logan's comment that we have not adequately communicated this focus and the important new results. In our revised manuscript we sharpen the description of this synthesis in the Abstract, Introduction and Conclusions sections. Dr. Logan's suggestions regarding analysis of the Japanese data are discussed where specifically commented upon below.

The first major problem with the paper is that it does not give a summary of previously published work on trends in tropospheric ozone in the Introduction, so the reader can see the context for the current work, and see what may be new. There is a body of literature on changes in tropospheric ozone, often using the same data as in this work. As far as I can tell, the overall results in this paper are not new: ozone doubled from the 1950s to around 1990 in Europe; there are seasonal differences in the ozone trends; ozone has leveled off and started to decrease in summer over Europe; ozone

has increased in spring over western North America. The leveling off at Mt. Lassen in California is new, as far as I know. The lack of references to prior work on ozone trends continues throughout the paper (except for occasional references to papers by the co-authors). However, the paper contains a reasonable amount of citations the literature on all other areas (model studies, trends in precursors, etc), so the lack of citations to the prior literature on ozone trends, the focus of the paper, is all the more strange, and unacceptable.

The paper requires a paragraph early in the introduction that states, at a minimum:

1. Ozone doubled in the Swiss Alps from the 1950s to the early 1990s (Staehelin et al., 1994). The results of Feister and Warmbt (1987) on the increase at Arkona should also be discussed. 2. Ozone sonde data show increases in ozone over Europe from the 1970s to the 1990s, although the details of the increase differ among the three stations (Logan, 1994; Logan et al., 1999). 3. Ozone at Mace Head increased from 1987 to the late 1990s with no increase thereafter (Derwent et al., 2007). 4. Data from alpine sites in Europe show ozone increased from 1978 until around 2000 and then stabilized (several papers summarized in the Introduction to Logan et al. (2012), as well as the latter paper). The synthesis of data for central Europe by Logan et al. (2012) shows that ozone has decreased since 1998 in sonde, MOZAIC, and alpine site data, with the largest decrease in summer, and no increase in ozone in summer since 1990. The alpine data show increases in the decades before 2000, except summer. The paper also shows similar behavior at Mace Head and the alpine sites. 5. Studies by Parrish et al. (2009) and Cooper et al. (2010) found increases in spring in ozone on the west coast of the U.S., but had to rely on much sparser data records than available for Central Europe. Earlier work on trends in this region by Oltmans et al. and Jaffe et al. should also be acknowledged. While these authors are cited, their results are not discussed. 6. Tanimoto et al. (2009) showed large increases in spring at Happo, the only mountain site in Japan, that are larger than increases at sea-level sites by factors of 2-3.

Instead of this, the paper merely says: p. 13885. l. 9-10 "During the latter half of the 20th century O_3 concentrations increased markedly at northern midlatitudes. This increase has been documented by a variety of observational studies, . . ." (Note lack of citations).

l. 23-25. "Nevertheless, it does appear that concentrations were lower up to the 1950s with mixing ratios (strictly speaking mole fractions) around 10-20 ppbv, for example, over Europe [Volz and Kley, 1988; Staehelin et al., 1994]."

p. 13886. l. 7-8. "Several recent summaries of changes in tropospheric O_3 have been published [e.g. Vingarzan, 2004; Oltmans et al., 2006]." Note, no comment on the results in these papers.

Until this paper summarizes the results of previous work on trends in the Introduction, it is not acceptable for publication. The summary should start where the well-known increase in tropospheric ozone is first mentioned, p. 13885, line 10. The paper must also comment on previous work at appropriate points throughout the paper and in the Conclusions.

The lack of information on previous work on ozone changes is particularly noticeable given the detail that is included on factors that may influence ozone changes on p 13885-6.

Our goal is not to review trends in tropospheric ozone in general. Several such reviews are available, and we cite two in the Introduction [*Vingarzan*, 2004; *Oltmans et al.,* 2006] to provide the interested reader with further context for our work. Nevertheless, we have added some additional historical context as suggested by Dr. Logan. This does provide ample context for the specific work that we present in this paper, and at appropriate points in the discussion of the results nearly all of the references suggested by Dr. Logan are cited and discussed.

Comments on the results section.

The authors include figures for all the sites as seasonal time series in a Supplement, while showing figures in the main body of the paper for only a few sites: Hohenpeissenberg and Mace Head for Europe, a composite of sites along the coast (2 year update of Parrish et al., 2009) and Mt. Lassen for the west coast of the U.S., and Mt. Happo for Japan. They also compare time series from various sites for spring (Figure 5). I recommend they add a figure like Figure 5 for other seasons, at least for summer, into the paper (include Figure S12). At present these figures are in the supplement. This paper would be much improved by including a focus on summer, the most photochemically active season, as well as on spring. Most exceedances of ozone air quality standards are in summer, with serious effects on human health, crop yields, and vegetation in general.

The changes in tropospheric ozone have many effects. Our goal is not to survey these effects in general. Figure 5 of the paper is intended to exemplify the analysis from which the major results (presented in Figures 6-8) were derived. We selected spring for this example, since it is the season of strongest long-range transport, and since we are focusing on baseline ozone trends, this is an important consideration. A second example from another season is not necessary.

The paper also shows comparison of trends, for varying time periods depending on the start date of the record (Figures 7 and 8). This figure should include the differing time periods for which the trends were computed. It would very be useful also to compare trends for identical time periods, which would restrict the analysis to 1991-2009. This is an interesting period, as trends in emissions in Asia are diverging from those in Europe and North America. I recommend that the authors compute such trends, and make plots similar to Figures 7 and 8 for this period. For Japan they should use data from Ryori, Japan, which has data for 1990 onwards.

Table 1 gives the time periods for which the trends were computed. Figures 7 and 8 are already packed with information, and repeating the time periods in the figure would add unnecessary clutter. Figures 1 and 2 included here show Figures 7 and 8

revised based upon data for 1990-2010 only. In other respects, the analysis is exactly the same as described in *Parrish et al.* [2012]. The major difference is the poorer precision with which the trends and accelerations can be determined when the data sets with longer records are limited to only 20 years. The absolute (Fig. 7a revised) and relative trends (Fig. 7b revised) determined for some of the European sites are systematically smaller compared to those derived from the full data sets, as expected from the negative accelerations of the trends noted in this paper. However the precision with which those trends can be determined over only two decades of data is so poor that in only a few cases (Arkona-Zingst in summer, and Arosa in summer and winter) are these differences statistically significant at the 95% confidence limits. Interestingly, the average relative trends over the European and North American sites for 1990-2010 (0.89 ± 0.17, 0.47 ± 0.22, 0.64 ± 0.18 and 1.03 ± 0.19 % /year in spring, summer, autumn and winter, respectively) compared to the full data records (1.08 ± 0.09, 0.89 ± 0.08, 0.79 ± 0.12 and 1.22 ± 0.12 % /year) are statistically significantly lower in summer (ratio is 0.64) but not in the other three seasons, although they are generally somewhat lower (on average by 17%). The accelerations in Fig. 8 revised are generally more negative for the European sites during 1990-2010. However, the differences are statistically significant only at Arkona-Zingst, and now fewer of these negative accelerations are statistically significant.

Given the relatively small differences in the analyses for the two different selections of time periods, in our judgment the advantages of determining these parameters with greater precision outweigh any advantage from restricting the analysis to shorter, but consistent time periods. The revised figures are shown here, but will not be included in our revised paper.

As discussed fully below, the data from Ryori, Japan are not included.

The choice to focus on Hohenpeissenberg is rather odd, as it is the least likely to be a baseline station, compared with the high altitude sites of Zugsptize or Jungfraujoch. The site is only 300 m above the surrounding countryside, it is within 50 km of the

center of Munich, and within 10 km of several towns with a populations of 10,000-20,000. It is well within the boundary layer. The diurnal variation at the site is not discussed, and should be. The reason for showing this site should be given, as it can only be considered as a regional central European site, rather than a baseline site.

Dr. Logan makes a good point, but we really do not focus on Hohenpeissenberg to any greater extent than any of the other data sets. We chose that site for the first example in Fig. 2 to illustrate the analysis because it has a long, high quality data record. As Dr. Logan points out, this site may have stronger local and regional effects due to its location, but as discussed in the paper, the Hohenpeissenberg data have been filtered to remove very local contamination. Further, the close correspondence of its ozone changes, seasonal cycles, etc. to the higher altitude sites and the analysis discussed in Section 4.4 indicate that these local and regional effects do not strongly affect our analysis or results.

Linear trends are run for 1970 to 2000, and the text comments that a linear increase was observed for the first 30 years, yet it is obvious from Figure 1 that the increase in summer had stopped by 1990. One can see by eye that there is no increase in summer after 1988, and Figure S12 shows the decrease at all the European sites except Mace Head from at least the mid-1990s if not earlier. This must be discussed.

Dr. Logan actually misquotes our paper here. We do state "The linear regressions indicate that over at least the first approximately 30 years of the data record, an increase in O_3 was observed in all four seasons." We stand by this statement. We are careful to describe in Section 4 of *Parrish et al.* [2012] our purpose in using linear regressions: "The slope, in units of ppbv/yr, quantifies the growth rate of O_3 and is the best measure of the average annual increase in O_3 mixing ratio over the period of the data record, ... The utilization of a linear fit does not assume that the temporal change was necessarily linear over the data record, and does not imply that a trend will continue linearly into the future." Thus, it is clear that we do not state that a linear increase was observed over the first 30 years.

In the paragraph beginning on pg. 13897 we do discuss the slowing and reversal of the increase in O_3 at Hohenpeissenberg. Our quadratic fit finds that the summertime maximum was reached in 1996 $\pm$ 8 yrs, which is (barely) in statistical agreement with the Dr. Logan's 1988 estimate by eye. And in several places in the paper we discuss the slowing and in some cases the reversal of increasing O_3 at European stations. The abstract of the revised manuscript will read: "At most European sites and some North American sites the rate of increase has slowed over the last decade (possibly longer), to the extent that at present O_3 is decreasing at some sites in some seasons, particularly in summer. "

Logan et al. (2012) noted the ozone maxima in July 1994, August 2003, and July 2006 caused by heatwaves. We also commented on the slow-down in the growth of ozone at alpine sites from the 1980s to the 1990s. It is not a new result that the ozone increase over Europe has stopped, as discussed by Logan et al. (2012) and papers cited therein. This should be made clear, as it is seen at all the other alpine sites.

We agree that others have noted on the slow-down in the growth of ozone. We have added the suggested reference to our revised manuscript.

Ozone changes at sites in Japan.

The authors did not pick the most suitable sites for analysis, and more careful analysis of the Japanese surface data is needed. Logan et al. (2012) showed that it is easy to identify problems with particular data sets by examining time series of monthly mean differences of sites within a few hundred kilometers of each other. I include such a plot (Figure 1) for the EANET sites used here, Rishiri Island, Sado Island, and Cape Tappi, as well as data from Oki (EANET) and Ryori. The site locations are shown in Figure 2. (The Ryori data are available at http://ds.data.jma.go.jp/gmd/wdcgg, and the EANET data for 2000-2009 at http://www.eanet.cc). The data for the other sites before 2000 are not publically available. Parrish et al. averaged ozone at Rishiri, Tappi, and Sado.

The problems evident with the Cape Tappi data in Figure 1 of *Logan* [2012] were

avoided in our analysis, because the data after May 2008 were not included. We thank Dr. Logan for identifying the Ryori data set for us; we had not considered that record previously. We have reproduced the plots in Figure 1 of *Logan* [2012] for Ryori from the monthly data downloaded from the suggested web site. The results of the analysis of this data set are not statistically significantly different from the Japanese MBL data set that we included, although the trends are generally smaller. Also, the longer Ryori data set gives significantly improved precision of derived average ozone trends. However, Figure 3 here shows two-month long time series of hourly average O_3 and carbon monoxide (CO) data from Ryori. The elevated CO concentrations indicate that the Ryori site is strongly influenced by local and likely regional Japanese pollution. Both seasons show elevated levels of CO with O_3 titration during some episodes in winter, and O_3 production in summer (and even during one interesting episode in February). Without a careful selection of meteorological conditions, we cannot take the derived O_3 changes as characteristic of baseline conditions. Such an analysis is beyond the scope of this paper, so we have not included the Ryori data.

Other comments:

Abstract. The text states that "the rate of increase has slowed" at most European sites. This is a strange way of saying the increase has stopped, or in some cases, turned around to a decrease. Clarify.

Importantly the rate of increase has not stopped at all sites in all seasons, so the wording suggested by Dr. Logan is not appropriate. However, we have clarified this statement to read "At most European sites and some North American sites the rate of increase has slowed over the last decade (possibly longer), to the extent that at present O_3 is decreasing at some sites in some seasons, particularly in summer.

p. 13884. l. 3. Give a more recent reference as well for the source of ozone from STE.

We have added the Collins et al., 2003 reference.

l. 10. Levy's paper is not about ozone, so drop the citation here, and include the Chameides and Walker (1973) reference here.

Suggestion accepted.

l. 17-20 Insert a reference for the lifetime of ozone towards chemical loss, such as Fusco and Logan (2003), which shows this in their Figure 5. The ozone lifetime is considerably longer than the quoted 20-30 days in the middle and upper troposphere.

Suggestion accepted, and the longer lifetimes are now noted.

p. 13885 The cartoon in Figure 1 showing processes affecting transport of ozone and PM across the Pacific was fine in the HTAP report, intended for a broader (and perhaps less informed) audience, but it is not needed for this article in ACP. And this article is not focused on transport across the Pacific.

We intend this figure to generally represent intercontinental transport processes, as is clear from the figure caption. Neither the figure nor the paper focuses on transport across the Pacific. We believe that the figure does serve the purpose of emphasizing that this paper focuses on large scale, baseline O_3 changes rather than on regional changes.

l. 15 Variability in the stratospheric flux of ozone has been proposed as a cause of the increase in tropospheric ozone in the 1990s (after the minimum attributed to the aerosol loading from Pinatubo) in the cited papers, as well as by Tarasick et al. (2005) and Terao et al. (2008). It has never been suggested that this has influenced the doubling of ozone since the 1950s.

Agreed. We have clarified this statement in our revised manuscript.

p. 13888-13889. The paper argues that it is about "baseline" sites, but the authors can compare "baseline" and "nonbaseline" ozone at only 3 of their 11 sites. They should remove "baseline" from the title of their paper.

We do conduct what comparisons we can between baseline and unfiltered data sets at 8 of the eleven sites. This issue is addressed in Section 4.4. Nevertheless, since we do consider data that are not filtered for baseline conditions, we have removed "baseline" from the title of the paper.

They also argue that for global models a comparison to baseline selected data is useful for evaluation of simulated trends, but ignore the fact that this is never done (in the literature to date), as it is far from straightforward to duplicate the methods used to separate the data into baseline and non-baseline, in part because of the larger spatial scales at which global models are run.

It may be far from straightforward to do baseline filtering in global models, but certainly it would be quite useful to disentangle changes in local influences from changes in baseline O_3 in models to determine whether the model can reproduce the baseline changes.

line 17. Downwind of the Asian continent, surely. Japan is in Asia.

We have repaired this sentence as suggested.

p. 13891 l. 9-10, 17-18. It is interesting that the authors are so negative about the sonde data, when they describe the early Arkona data as being from "well calibrated, well characterized" wet chemical methods. In fact the sonde and Arkona techniques both relied on the oxidation reduction reaction of KI with ozone, and SO_2 interferes quantitatively (negatively) with the measurement of ozone. Wintertime concentrations of SO_2 may have been high enough to interfere with the ozone measurements in the early years, and the published paper only comments on annual mean SO_2 being low in 1969-1971. It is interesting that there was essentially no increase in ozone in the 1970-1980s following the installation of a filter for SO_2, until the higher values in 1989.

We are not completely negative regarding sonde data, as they have served as the basis for many useful analyses. However, it is much less challenging to develop a consis-

tent temporal record from surface measurements where the wet chemical method can be implemented into one "well calibrated, well characterized" instrument that is continually operated with consistent reagents by a consistent operator over many years. With sondes, generally a different, necessarily less expensive and necessarily less well-characterized instrument is used for each launch. Hence, we do believe that it is prudent to not rely on sonde data sets in the determination of the long-term changes in tropospheric ozone concentrations. This prudence is supported by numerous analyses showing inconsistencies between sonde and other data sets, including a least one by Dr. Logan, which we cite in *Parrish et al.* [2012].

Table 1 must include which measurement technique was used for which period, for all data sets used. The reader needs to know when earlier, and often less reliable, methods were used. For example, which technique was used at Mt Happo before 1998? The Tanimoto et al. analysis starts in 1998.

We have not investigated the particular techniques used for which periods at which sites. Instead we rely on data sets previously discussed in the literature. Most of these data sets have been extensively described in earlier papers, which we cite. In the revised manuscript, we describe the measurement technique used at Mt Happo before 1998.

l. 13. Derwent et al. (2007) did not use the dispersion model to filter the data from 1989 on, they used halocarbon and CO data up to 1997, and the dispersion model after that.

Thank you for this correction. Dr. Logan is correct regarding the Derwent et al. [2007] paper. Here we use data selected by the dispersion model for the entire period analyzed, but the period before 1997 has only recently been analyzed by the dispersion model and included here. The revised manuscript now has a more accurate description of this data selection procedure.

Section 4, on the Analysis approach, largely repeats the same points, and uses ex-

actly the same methods, as Parrish et al. (2009); the present text comes across as pedantic and repetitive, and should be shortened considerable but saying exactly the same approach was used as in the earlier paper. There are differing approaches to deriving seasonal trends in ozone, and the method adopted by the present authors is appropriate when there are no gaps in the time series, so that true seasonal means can be formed. The authors should say what they do when months of data are missing, especially if a season is represented by only one month. There are gaps in some of the datasets they used.

It is our experience that different researchers in the field have used contradictory methods. We prefer to give too much detail rather than failing to clearly describe our approach and the reasons for selecting that approach. The methods are described in a single paragraph, so this description is over long. A given seasonal average is included only if data are available for each of the three months of the season; this is now stated in the revised manuscript.

The authors should state how they obtain the confidence intervals for their quadratic fit, and state which statistical package they use, or if they wrote their own software.

Suggestion accepted; it is now stated in the revised manuscript that all of the regressions were calculated with Igor Pro, a technical graphing and data analysis package (http://www.wavemetrics.com/), which gives the parameters with 95% confidence limits through standard statistical methods.

p.13895 l. 5. It is unclear if the STE flux of ozone should change only gradually. The change in ozone in the lower stratosphere after the major eruptions of El Chichon and Pinatubo was not gradual, so even if the mass flux of air changes gradually, that of ozone may not have for short periods. And the air mass flux may not change gradually, but have interannual variability.

We agree with Dr. Logan's assertion. Our methods are not designed to capture such interannual variability, and we accept that limitation.

p. 13896 l. 3. The rate of change of the slope should not be referred to as the acceleration, as it can also be a deceleration. It should simply be referred to as the change in slope, or the quadratic term, with the units as given (ppb year^{-2}). To refer to "negative accelerations" is rather like referring to "negative increases" instead of "decreases" and sounds rather silly. This must be fixed throughout the paper, where "negative acceleration" appears quite often.

We have struggled with the terminology, since we are interested in quantifying both the average rate of change of O_3 and the average rate of change of that rate of change. The slope of a linear regression is the means we chose to quantify the rate of change of O_3, but for clarity we prefer to discuss the rate of change of O_3, rather than the slope. Similarly, we settled on the term "acceleration" as a term for the average rate of change of the rate of change of O_3. The term acceleration is familiar to most scientists who have had an elementary physics course, where it can have either a negative or positive magnitude. Importantly, the quadratic term is actually not equal to the acceleration, as they differ by a factor of 2 as indicated by Eq. 2 of the text. We believe that we have been clear and consistent in our terminology and its use.

l. 11-14. Of course the slope and its change (in original units) do not depend on the reference year chosen. This is hardly the place to be giving a tutorial about extremely simple statistics that were used.

In discussing the results presented in this paper with colleagues, the question of the effect of choice of reference year on derived parameters has risen several times. We do believe that it is useful to answer this question, which may also arise in the minds of the readers.

p. 13898. A quadratic fit is not likely to give the same year for maximum ozone as a data set with variability, so this point is rather belabored. There are many papers on the high ozone seen in August 2003 prior to the one cited.

We believe that the discussion is useful. An earlier reference will replace the one cited

in the revised manuscript.

In terms of the comment that the longest record will yield the most precise regression results, this is only because the increase is so large. The precision of a trend depends on the magnitude of the change, and the variability in the time series, as well as on its length. The statement as given (l. 23-24) sounds naïve. The Zingst-Arkona record has precise trends because they are so large compared to the variability in the record.

Here we believe that Dr. Logan is not completely correct. A measure of the precision is the magnitude of the confidence limits of the derived parameters. Those confidence limits decrease as the length of the data record increases, regardless of the magnitude of the change (other things, such as interannual variability, remaining constant). Ozone changes derived from the Zingst-Arkona record would be precise, as gauged by the $\pm$ ppbv yr $^{-1}$ confidence limits, even if they were very small compared to the variability in the record.

P. 13899 The discussion of the Mace Head data is a bit vague, "specific transport patterns", which begs the question of which ones, and why they only affected the late 1990s. The boreal fires that Derwent et al. mention were in August 1998, so cannot explain the high ozone prior to this, and they cannot explain the relatively high ozone in 1999 and 2000. Others have argued for transport anomalies in the extratropics after the 1997/98 El Nino, including enhanced STE, but these papers are not cited.

Our goal here is not to provide definitive explanations for the interannual variability of the Mace Head record; rather we wish to point out that the apparent slowing of the O_3 trend at that site may plausibly be due to a slowing of a long-term baseline trend, or to positive excursions of the interannual variability in the middle of the data record. No revision has been made to this discussion.

What is abundantly obvious from Figure 3 is that ozone at Mace Head does not appear to have increased since the late 1990s, and this is discussed in several previous papers.

The parameters of the regressions shown in Fig. S2 of the supplement indicate that the accelerations are significantly negative only in spring and winter. If 1998-2000 data are removed from the regressions, the statistical significance of the negative spring acceleration disappears, and a statistically insignificant positive acceleration appears in autumn. This is an excellent example of the difficulty of determining trends from data records of limited temporal extent. Given the limits of the current data record, the discussion in our paper is justified.

The text is unnecessarily confusing in saying that there are differences in seasonal cycle (and trends) between Mace Head and Arkona-Zingst. Why not just say that the former has a summer minimum, and the latter a summer maximum, as is well known and well understood (with citations of course). (Photochemical sink in summer for baseline Mace Head (low NOx), photochemical production giving a summer maximum over mainland Europe (with attendant emissions of precursors), also give reasons for differences in winter, etc.) The only proper way to compare trends between the two sites is to first compute trends for identical time periods, and compare them. This should be done.

While it is true that Mace Head has a summer minimum and Arkona-Zingst a broad spring-summer maximum, we do not believe that this phenomenon is well understood. The alpine sites in Europe (e.g. Zugspitze and Jungfraujoch) also have a similar broad spring-summer maximum, but this is not primarily due to photochemical production over mainland Europe (with attendant emissions of precursors), since this maximum reflects baseline ozone in the lower free troposphere transported into Europe. As air masses come ashore from the North Atlantic and are transported across Europe, the marine boundary layer structure present at Mace Head erodes, and continental vertical mixing processes bring free troposphere air into the continental boundary layer as shown in HTAP model results. Simultaneously at surface continental sites, photochemical production from continental emissions adds additional ozone, and ozone is lost by deposition within the continental boundary layer. The relative importance of

these processes toward establishing the seasonal maximum at Arkona-Zingst is not well understood. For our purposes in this paper, we do not need to sort out the relative influences of these processes. We have attempted to clarify our discussion in this paragraph without going into these complexities.

We disagree with Dr. Logan's assertion that the only proper way to compare trends between the two sites is to first compute trends for identical time periods, and compare them. We choose to select the longest possible time period to derive as precise a measure of the trend as possible. The revised figures 7 and 8 included in this response do compare the Mace Head and Arkona-Zingst trends over identical time periods.

p. 13900. l. 9-10. The alpine sites used in the supplement (and in other papers as noted above) do not cover a particularly large part of Europe, mostly just the Alps. Logan et al. (2012) show that there has been no increase in ozone in summer in central Europe since 1990, and this is apparent in the figures in this paper. However it is not mentioned explicitly, instead only commenting on the quadratic fit. The Zingst data also show this lack of increase since 1991 when that data start. Discuss this.

We now note that "At most European sites and some North American sites the rate of increase has slowed over the last decade (possibly longer), to the extent that at present O_3 is decreasing at some sites in some seasons, particularly in summer." The quadratic fits do provide a convenient, quantitative means to express this slowing increase and in some cases a reversal, a method that Dr. Logan has used in her recent JGR paper referenced in *Parrish et al.* [2012].

p. 13901-13902. A comparison is made of trends for filtered and unfiltered data for Jungfraujoch, but the authors did not do this the correct way, as they did not use the same time periods for each. In a record of 20 years, the addition of 2 years of data will inevitably change the trends, so they should have used 1990- 8 2008 for both filtered and unfiltered data for deriving trends with the quadratic fit (Figure S16 used 1990-2008 and 1990-2010 respectively), to show if filtering the data makes any difference.

As noted in the annotations in Fig. S16, the trends were derived for both data sets over the same time periods, as Dr. Logan suggests should be done (although the later years of unfiltered data are shown in the plot). Thus, no change is needed here.

p. 13903 There is discussion of the difference in trends for the filtered and unfiltered data from Mace Head. The reason for this (which the authors should point out) is that there is a jump in the offset between the two time series in 1997 (see Figure 1). It is not because there is a monotonic trend in the difference between the two. The change in the offset suggests that the filtering changed around 1997. This should be discussed. It seems highly unlikely that titration of ozone by NOx is an issue at Mace Head, unless the authors are discussing urban air in winter. Are they arguing that undiluted urban air reaches this remote location in winter? Clarify.

As has been clarified in previous discussion, the Mace Head data were filtered for baseline conditions using the trajectory dispersion model for the complete data set. Thus, there is no change in the filtering that could possibly account for the jump. The statistical significance of that apparent jump has not been established to our knowledge.

It is well established that titration of ozone by NOx is an issue at Mace Head in winter [*Derwent et al.*, 1994; 1998; *Simmonds et al.*, 1997] due to transport of polluted air masses to Mace Head from continental Europe. *Parrish et al.* [1998] also report transport of such air masses from the North American coast 1000 km downwind to Sable Island. In fact titration of ozone by NOx dominates the variability of ozone at both Mace Head and Sable Island in winter. No clarification is required.

p. 13906 l. 5-6. "Observational data" – one is redundant. Observations, or data, you do not need both. There is no ambiguity here that the paper is about observations. There is the same amount of data in all seasons except for the Cooper et al. study, so this should be made clear.

The phrase "observational data are most abundant," has been removed

Figure S12, comparing the time series in summer, should be moved to the main paper and discussed there. This is the most photochemically active season.

We disagree. We have kept the spring data to exemplify the analysis.

This statement should be dropped: "it must be realized that the lack of a statistically significant change (e.g. the Japanese MBL in winter in Fig. 7a) does not necessarily indicate that there has been no change; rather it may indicate that any long-term change that occurred over the period of the data record is too small to be discerned with strong statistical significance given the length of the data record and the interannual variability that is present in the data sets." This sounds like a plea for a trend in ozone when the trend analysis says there isn't one for the period analyzed.

Here we again disagree with Dr. Logan. Taking the particular example discussed, the trend derived from the Japanese MBL in winter is 0.29 $\pm$ 0.48 ppbv/yr, which is not statistically significant at the 95% confidence limit. However, this also indicates that there is only about a 14% chance that the trend is zero or negative. Further, the trend derived for the U.S. Pacific Coast MBL is 0.37 $\pm$ 0.14 ppbv/yr, which is not statistically significantly different from the Japanese MBL trend. Thus, to say that there is a trend at the U.S. Pacific Coast MBL, but that there isn't a trend in the Japanese MBL is misleading. In fact, there is about a 37% chance that the Japanese MBL trend is larger that the U.S. Pacific Coast MBL trend of 0.37 ppbv/yr, which is a greater chance than its being zero or negative. Hence, the questioned statement is important, and conveys a message that is useful to all those that work with and compare ozone trends.

The authors should use the Ryori data instead, which does happen to have a small, but significant, increase in winter, as I show above.

Thank you for suggesting the Ryori data; however as discussed above, the trend at Ryori cannot be taken to represent baseline conditions.

p. 13906 It has been known for decades that ozone increases with altitude above the

surface, based on profile data (see literature cited in Logan, 1999). This section is rather naïve. It is common practice to show profile trends in percent per unit time (see Logan et al., 1999, and references therein).

Indeed it is well known that ozone increases with altitude, at least above the boundary layer. The increase with elevation of surface site is a more complicated issue, since the sites are all within the boundary layer, but that increase is also well established as we reference. Our goal is not to point out a new finding, but rather to establish the method we use to normalize the ozone trends to the year 2000 intercept. *Logan et al.* [1999] do show trends in percent per unit time, but this (at least in general usage) implies an exponential rather than a linear change. These are significantly different. For example, a linear increase of 5% of an initial value per year corresponds to a factor of 2 increase in 20 years, while an exponential increase of 5% per year corresponds to a factor of 2.65 increase in 20 years. It is useful to clearly define the methods we use.

p. 13907

The authors average the trend results for nine sites in Europe and North America, but the errors on the mean trends are likely too small. The time series for the alpine sites in Europe are highly correlated, and the results for these sites should not be considered as independent measures of trends for the entire northern mid-latitudes. A more careful approach to computing errors on the mean trend is needed, allowing for correlation among sites within a region.

This is a valid point. If we assume that the three alpine sites in Europe provide only one independent measure, then we have seven independent measures instead of nine (at least in spring). This would increase the confidence limits on the averages by a factor of the square root of 9/7, or 13%. These larger confidence limits are included in the revised manuscript. It is reasonable to consider the other sites as independent measures of trends for the entire northern mid-latitudes, since they represent quite different environments: the eastern Atlantic MBL, the Baltic Sea MBL the continental

European BL, the eastern Pacific MBL, the western North American continental BL, and the free troposphere over western North America.

The text reads as if the trend was the same for 50 years (1950-2000), but the time series show that the leveling off started before 2000 at many sites.

We definitely do not wish to convey the idea that the trend was the same for 50 years. This is clear from our two mathematical models used to describe the changes. The linear fit (Eq. 1) implies a constant trend, but the quadratic fit (Eq. 2) implies that the trend was changing at a constant rate over the entire data record, not just since or before 2000. Again, our purpose in using linear regressions is to quantify the average annual increase in O_3 mixing ratio over the period of the data record, not to imply that the temporal change was necessarily linear over the data record.

Comments on the Supplement.

Figure S4. Using the very limited data from Arosa in the 1950s to compute trends when there is a gap of over 30 years is stretching things a lot, especially when the early data are duplicated as 5 identical points. As noted above, ahe doubling of ozone from the early to the later data is already documented in the paper by Staehelin et al. (1994). A similar comment (to S4) applies to Figure S6. The fit for 1934-2000 in Figure S6 clearly is only for summer, so the legend should be changed. The caption should state that the Jungfraujoch data are available at http://ds.data.jma.go.jp/gmd/wdcgg/.

Our primary goals are, first, to determine the average annual increase in O_3 mixing ratio over the period of the data record, and second to determine the average acceleration over the period of the data record. The presence of the 30-year gap in the measurements is of small consequence for these purposes. The gap may limit the precision of these determinations, but the long, even if discontinuous, data record yields high precision determinations. Technically the legend for Figure S6 is correct, since the fits are over all available data from 1934-2000; the plotted lines end in 1990 for the three seasons other than summer, so no confusion should arise.

Figure S8. The caption states "Nevertheless, the linear fit includes all years, because the precision of the derived parameters is significantly better, and the data near 2000 appear anomalously high." Subjective comments such as "the data near 2000 appear anomalously high" have no place in this paper as a rationale for anything, especially when the most obviously anomalous points in the figure are in 1994, and are not even mentioned. One cannot say which points are anomalous without doing a lot more analysis.

We agree with Dr. Logan's point. We have removed the subjective statement.

Figure S11. The data for 2000-2009 are available at the EANET web-site, and are the same as those used by Tanimoto et al. (2009).

This is now noted in the figure caption in the revised manuscript.

Figure S12. Move to the paper and discuss the summer time series as noted above.

As noted above, we prefer to limit the number of season examples to one.

Figure S12. left: what are the solid lines? right: The vertical distribution of trends should only be shown if the same time period is used for all sites.

We assume that this comment refers to Fig. S15. The solid lines indicate the linear regressions of the intercepts (left panel) and trends (right panel) with elevation. This is now indicated in the figure caption. Since derived trends show very little statistically significant difference as the analyzed time period varies, we prefer to work with the more precise trends derived from the full periods of the data sets.

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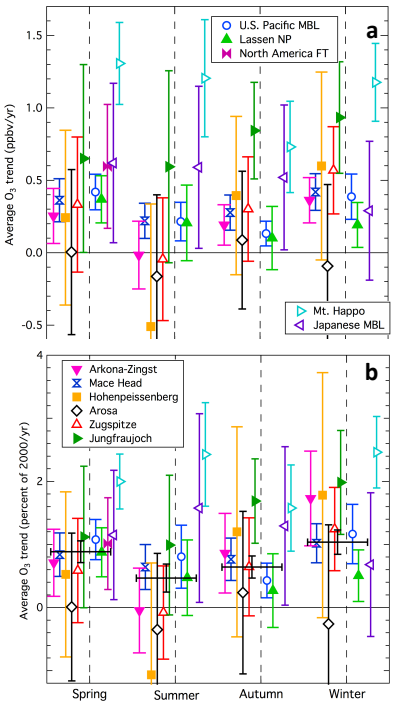

Fig. 1. Revision of Fig. 7 of Parrish et al. [2012]. This figure follows identical analysis that led to the original figure, except it includes 1990-2010 data only.

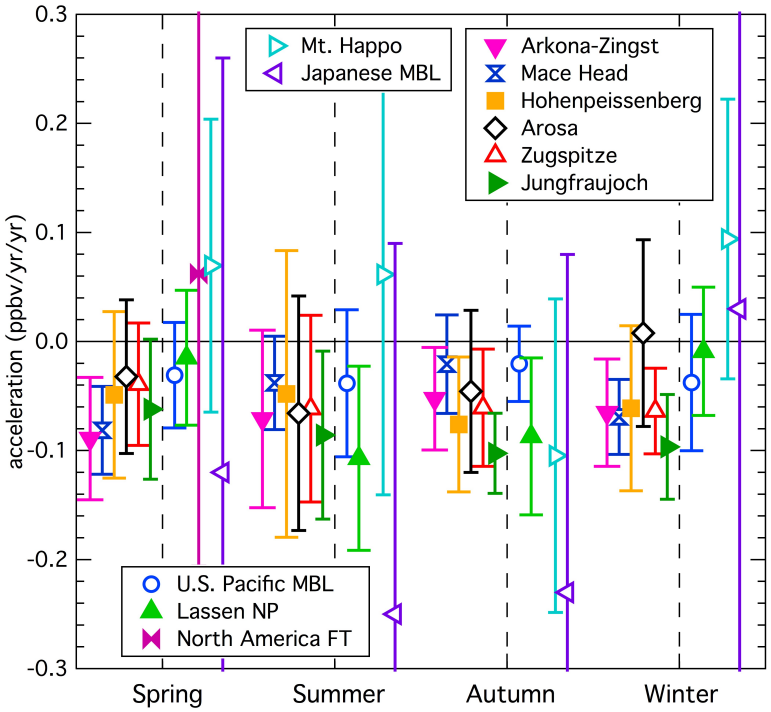

Fig. 2. Revision of Fig. 8 of Parrish et al. [2012]. This figure follows identical analysis that led to the original figure, except it includes 1990-2010 data only.

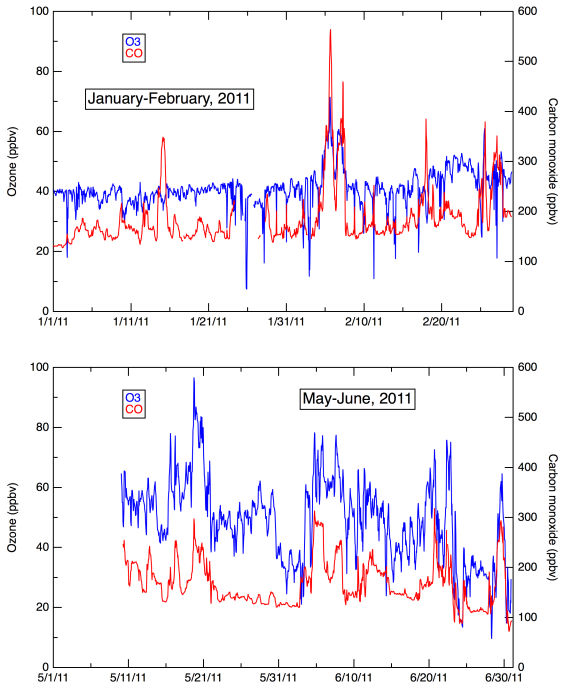

Fig. 3. Two-month time series of hourly average measurements of ozone (blue) and carbon monoxide (red) at Ryori, Japan during wintertime and spring-summer time periods.