

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

### **Anonymous Referee #3**

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Review of Parrish et al, acp-2012-273.

**General comments** The paper brings together long-term surface/free trop. ozone measurements from the Northern hemisphere and presents trend analyses of the data. The analyses are done in a straight-forward manner and the presentation is mostly done in a clear way. The main findings of the paper are of large interest to the scientific community. Thus, the manuscript is recommended for publication in ACP. Several modifications are suggested before publication, though. Significant parts of the underlying data and analyses are based on previous work published e.g. by Parrish (2009) and Cooper (2010) etc. More details of these studies would improve the clarity of the manuscript. Furthermore, parts of the observational data needs more discussions.

**Specific comments** p. 13890: A map showing the monitoring stations/areas including

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the underlying individual stations for the merged data sets would help.

p. 13891: Regarding the measurements at Arkona, the authors state: “These measurements were initially conducted by well-calibrated, well-characterized wet chemical methods”. Could the authors provide some more discussion of the general agreement between the old wet chemical method and today’s UV monitors? Is there any risk of systematic artefacts due to interference or other analytical pitfalls? The very low concentration levels in the mid 1980s may indicate analytical problems.

p. 13891: The authors refer to (and use) the measurements from the 1930s and 1950s (Arosa and Jungfrauoch). A short description of the analytical methods together with the expected precision and the general quality of these data should be given (Fig S4 and S6 indicates the importance of these data).

p. 13892: The authors state: “Parrish et al. (2009) have demonstrated that these measurements can be combined into a single record representative of MBL baseline O3 concentrations.” How was this done? It’s not obvious how it’s possible to combine time series from several stations spread over a large area into one single time series. Although this is documented in the original reference it would help the readability of the present manuscript to include some description here.

p. 13893: (Similar comment as above). The authors write: “To increase the robustness of the latter data set, measurements from the three northern MBL sites (Rishiri Island, Cape Tappi, and Sado Island, 38–45 N latitude) reported by Tanimoto et al. (2009) are combined into a single data set.” Again – how are these three more or less parallel time series “combined” into one? This should be explained.

p. 13893: Authors write: “For the US Pacific Coast MBL, Parrish et al. (2009) utilized a high, onshore wind window to select baseline conditions.” What does this mean? Was the screening of data based on local wind measurements at the individual sites – for certain sectors/wind speeds? How good metric is the local wind compared to the general advection (e.g. land/sea breeze effects etc)?

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p. 13896: Authors write: “The year 2000 was selected here because that year is well inside the time period covered by all data records; selection of a reference year near one end of the data record would degrade the confidence limits for the intercept determined in the linear regression”. That may be true, but for several sites (e.f. Hohenpeissenberg) the linear fit is applied only unto 2000, thus this year is indeed at the end of the record. What possible effect does this have for the results? Should the ref.year be changed to an earlier year for these sites?

p. 13897 and elsewhere: The authors find the strongest rate of increase in winter. This is somewhat surprising as one could expect the signal of anthropogenic precursors for ozone formation to be stronger in spring and summer. Some discussion of the possible reasons for the seasonal differences in long-term trends would be of interest.

p. 13901: Authors write: “After 2007 the traceability system and operational protocol for monitoring ambient O<sub>3</sub> in Japan was modified and the instruments of EANET were replaced in 2009, so a systematic error is suspected. Efforts are underway attempting to resolve this discrepancy; until a resolution is reached, the more recent data are not included in the analysis of O<sub>3</sub> changes at this site.”

The Japanese data are of particular interest in this manuscript because they indicate a very different trend than in Europe and the US. Unfortunately, the two most recent years of data (after the mentioned change in procedures) may indicate a major problem with the previous ozone data and it's not obvious that it's only reflecting a “systematic error” as the authors write. A systematic error could either be a fixed or a relative bias which would influence in the estimated trends in different ways. A non-systematic error (e.g. problems linked to procedures for calibration, maintenance etc) would, however, influence the measurements and trends in a more random manner. The Japanese historical data are an essential part of the manuscript and at the same time the most recent data undermines their reliability. How to deal with that? I think the authors need to give more details about these data, what type of change in procedures etc were adopted in 2007 and what implications that will have for the data and the estimated

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trends up to 2007.

p. 13902: The authors should give some more information about the North American FT data. Based on Fig S9 they seem to be based on data in 1984 and the years from the mid 1990s and on. Why this large time gap? Is the linear slope significant if 1984 is omitted? Although this presumably is explained in Cooper et al (2010) some key information should be included here as well.

p. 13907. Authors write: "The result is an increase of approximately  $1\%yr^{-1}$  relative to the respective year 2000 intercepts in each season, specifically  $1.08\pm 0.09$ ,  $0.89\pm 0.08$ ,  $0.79\pm 0.12$  and  $1.22\pm 0.12\%yr^{-1}$ ." It's somewhat unclear how these confidence levels were calculated.

p. 130909: The value of the data from Bermuda and Sable Island is questionable, and the authors also state that due to the scattered data coverage "neither of these data sets is ideal". I think these data don't bring much extra value to the manuscript and suggests taking them out unless the authors provide good reasons for having them in.

p. 13912: This paragraph is somewhat obscure and should be rephrased: "Approximately 85% of the European and North American error bars include the average seasonal O3 changes derived from the nine European and North American data sets. To put this in perspective, if the entire northern mid-latitude region in this longitude range had hypothetically experienced a uniform O3 change at all sites, with differing inter-annual variability superimposed, then 95% of the confidence limits would be expected to include the average seasonal O3 changes. The close correspondence between the actual and the hypothetical inter-site agreement indicates this high degree of uniformity in the average seasonal O3 increases."

p.13912: Authors write: "This slowing in the rate of increase has advanced to the point that O3 over Europe has recently begun decreasing." This could be read as a rather political statement. When looking at e.g. exceedances of ozone threshold values, it is presently difficult to see signs of reductions in Europe. I would suggest to rephrase

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this statement to clarify what is actually decreasing. Is it the baseline mean, the overall seasonal/annual mean etc?

Further remarks This manuscript is based on seasonal means. Have the authors looked at other ozone indicators, like percentiles, day-time values, daily max etc? Change in anthropogenic emissions would not only affect the seasonal means, but also the frequency distribution of hourly ozone (both the high and low percentiles). In general, the annual or seasonal mean values are less suited for process studies than e.g. the high percentiles (and low percentiles in winter) since the mean values are a mix of all kinds of atmospheric processes. For the present study the mean values may, however, be the most appropriate, as the focus is long-term changes in the background/baseline. Some words on this question would be good to include, though.

Do the authors have any explanation why the results for different seasons apparently vary in similar manner for several sites, i. e. that dips and peaks in the 3-months means seem to be reflected in both summer and autumn, summer and winter etc? Seems to indicate a season-to-season dependency?

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Interactive comment on Atmos. Chem. Phys. Discuss., 12, 13881, 2012.

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