

Atmos. Chem. Phys. Discuss., community comment CC1
<https://doi.org/10.5194/acp-2020-1198-CC1>, 2021
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Comment on acp-2020-1198 by Cooper, Tarasick, Galbally and Schultz

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Community comment on "Investigations on the anthropogenic reversal of the natural ozone gradient between northern and southern midlatitudes" by David D. Parrish et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2020-1198-CC1>, 2021

No Evidence for Anthropogenic Reversal of the Natural Ozone Gradient between Northern and Southern Mid-latitude

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The manuscript by Parrish et al. to which we refer with our comment misinterprets findings from the recent Tropospheric Ozone Assessment Report and draws conclusions that are not substantiated by the available data. Below we discuss the main issues that we have with the Parrish et al. study, as lead authors of the *TOAR-Observations* paper (Tarasick, Galbally et al., 2019).

Ozone increase since pre-industrial times:

Interest in measuring and quantifying long-term changes in atmospheric composition grew slowly following the activities of the 1957 International Geophysical Year, and was brought into international focus by a meeting in Stockholm in 1971: a "Study of Man's Impact on Climate" (SMIC 1971). Subsequently Angell and Korshover (1983) produced the first systematic account of trends in tropospheric ozone in the 800-400 hPa layer, finding a 12% increase in ozone between 1970 and 1981. The notion that tropospheric ozone has increased greatly since pre-industrial times, however, seems to have originated with Bojkov (1986), based on an analysis of Schönbein papers, and the publication of trends in several decades of surface ozone measurements made at some sites in Germany (e.g. Arkona), by Feister and Warmbt (1987). Although more quantitative than the Schönbein papers, the Arkona measurements also used a difficult and obsolete chemical method with a large and uncertain bias relative to modern UV methods, and an additional bias due to SO₂ interference. Nevertheless, with the then-recent recognition (Crutzen, 1973, 1974;

Liu et al., 1980) that photochemistry could be a major source of tropospheric ozone, this hypothesis seemed likely, and was further reinforced by Volz and Kley's (1988) analysis of the data of Albert-Lévy (1877) from Montsouris. Many other analyses concluded that ozone in pre-industrial times was as low as 1/5 of its present concentration (e.g. Marengo et al., 1994; Sandroni et al., 1992; Sandroni and Anfossi, 1994; Pavelin et al., 1999).

These conclusions conflicted with those of some contemporary observers in the early 20th century, who had noted that surface ozone was typically about 20-25 ppb in clean air (Fabry, 1950), and also that the very low Montsouris measurements were not characteristic of rural air (Hartley, 1881). Moreover, global atmospheric chemistry models had difficulty reproducing such a large historical increase from pre-industrial times (e.g. Wang and Jacob, 1998; Mickley et al., 2001; Lamarque et al., 2005; Young et al., 2013; Parrish et al., 2014; Young et al., 2018). For these reasons *TOAR-Observations* (Tarasick, Galbally et al., 2019) set out to examine in detail the quality and credibility of all known historical measurements of tropospheric ozone. This comprehensive reanalysis drew several conclusions:

- There is no unambiguous evidence in the measurement record back to 1896 of typical mid-latitude background surface ozone values below about 20 ppb
- As suggested by Fabry (1950), average values during the historical period for northern temperate regions were about 24 ppb
- There is robust evidence both from the TOAR analyses of surface, balloon borne and aircraft observations (Tarasick, Galbally et al., 2019) and from the recent analysis of oxygen isotopes in ice-core data by Yeung et al. (2019) for increases in the northern hemisphere (NH) of 30-70%, with large uncertainty, between the period of historic observations, and the present day
- The uncertainty in this estimated increase depends more on the modern region chosen for comparison than on the historical data, which are comparatively uniform. The representativeness of modern data thus seems to be the most important source of uncertainty in regional or global trend estimates.

What information can be gained from trends at a few sites in the center of Europe?

While accepting the re-analyzed historical data presented in *TOAR-Observations*, the Parrish et al. manuscript concludes (along with, apparently, the paper cited as Parrish et al. 2020a), that ozone has increased in the NH by $110 \pm 20\%$. Despite the surprisingly narrow confidence interval on this estimate, this conclusion is based on data from six sites: Jungfrauoch, Zugspitze, Arkona-Zingst, Mace Head, Hohenpeissenberg, and Arosa. Mace Head data were selected for "baseline" conditions, while the other sites contain available observations whose diurnal sampling is not explicitly stated. Arosa is a ski town at the bottom of a Swiss valley, Arkona-Zingst is a combined record from two sites on the north German coast, and Hohenpeissenberg is a hill in the middle of Germany. Jungfrauoch and Zugspitze are alpine sites and in daytime upslope winds bring air from the valleys to the summits. Cooper et al. (2020) showed that Jungfrauoch and Zugspitze are heavily influenced by the European boundary layer. The Jungfrauoch data before 1990 are very limited: 7 days in August 1933, and 5 days in August 1938. Older data from Arosa, Hohenpeissenberg and Zugspitze are from several wet chemical methods discussed by *TOAR-Observations*, and subject to SO₂ interference. Arkona data, as noted, are from the obsolete Cauer method and had significant biases relative to modern UV methods, rendering the Arkona-Zingst trend doubtful.

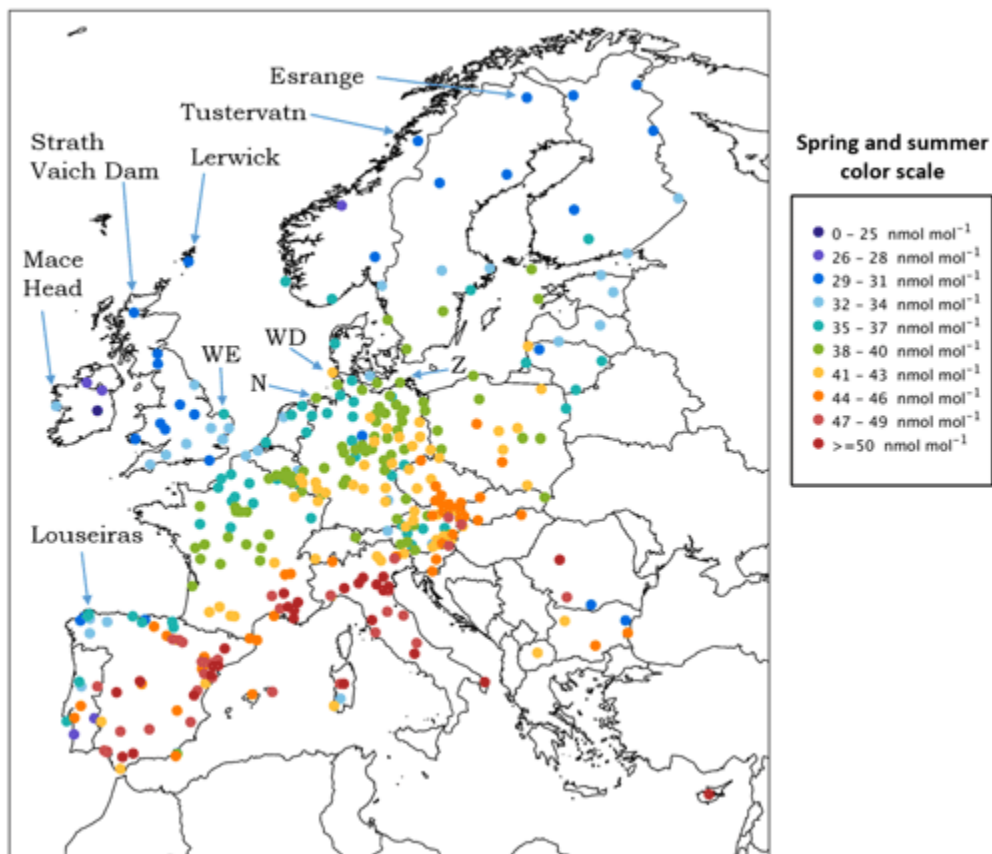


Figure 1. Observed summertime (JJA) daytime average ozone at rural sites (below 700 m) across Europe, 2010-2014. The sites used in the current manuscript are indicated, with Arkona-Zingst indicated by the letter Z, Westerland by WD, and Norderney by N, all on the North German coast. The UK site of Weybourne is also indicated (WE), and is discussed below. Note the increase of ozone from west to east, with even the coastal sites on the North Sea and the Baltic Sea showing more ozone than the westernmost sites. All data are from the TOAR database.

Parrish et al. (2021) (like Parrish et al. 2020a) are in effect claiming that data from this set of six sites, despite their issues, are more representative of changes in the NH than the averages used by *TOAR-Observations*. This conclusion is inconsistent with recent studies of the spatial representativeness of surface ozone (cf. Sofen et al., 2016). *TOAR-Observations* averaged all available "rural" data over several large regions for its comparisons. Stations were classified as rural based on a set of stringent criteria including low population density (see Schultz et al., 2017). As can be seen from Figure 1, there are a large number of such sites in Europe and these are distributed across the continent. More than 250 rural sites in Europe were used in *TOAR-Observations*). The use of just a few sites, as a superior subset, requires justification, which is not provided in the Parrish et al. manuscript. Such a justification would have to consider in detail the representativeness of the modern data, since, as noted above (point 4), the differences between the TOAR and Parrish et al. estimates of the NH change lie in the different choices of modern data. If properly carried out, this would be an advance on the TOAR analysis. Data representativeness is a key issue for both air quality and climate models (cf. Sofen et al., 2016). Spatial representativeness is typically the largest source of uncertainty in the use of ground-based data; surface ozone is normally undersampled, as over land surfaces it may vary on scales of kilometres (Spangl et al., 2007). Current state-of-the-art model evaluation procedures attempt to represent these small-scale variations, and the changes/trends at many different places.

As noted above, five out of six sites used by Parrish et al. to determine baseline ozone trends are actually situated in the polluted European boundary layer. Figure 2 provides a simple illustration of the differences in both the phase and the amplitude of the annual cycles as well as the residual mean value between baseline ozone at Mace Head, Ireland and ozone at rural sites used in the Parrish et al. analysis. In particular, we emphasize the differences in behaviour of the sites of Zingst on the northern German coast and Hohenpeissenberg in southern Germany (both used by Parrish et al., 2021) to that of Mace Head.

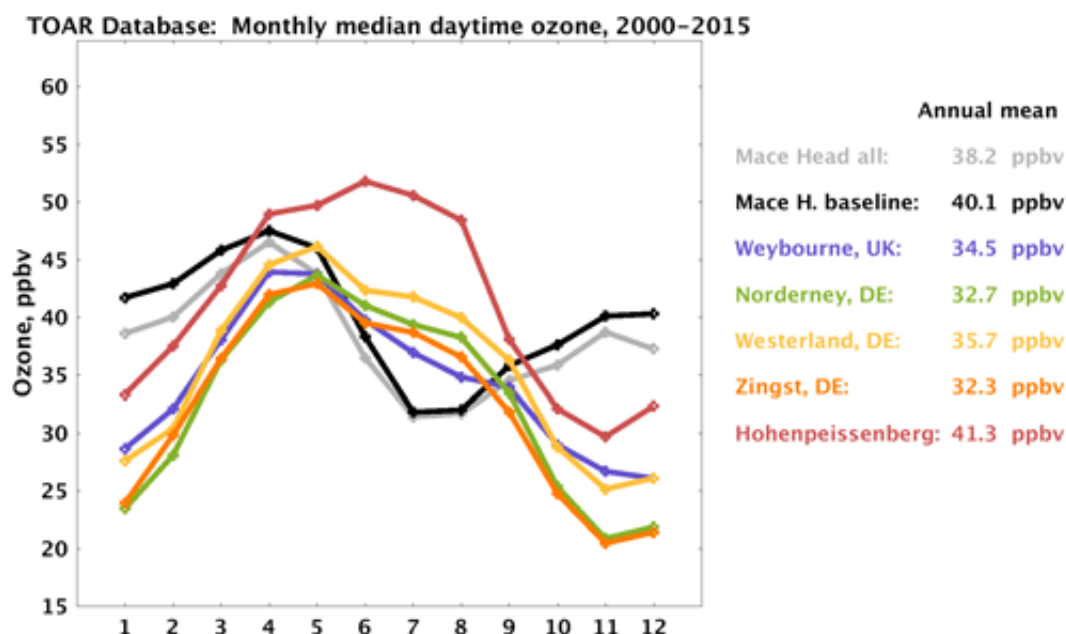


Figure 2. Monthly median ozone, averaged over the years 2000-2015, at six rural sites in Western Europe. All data are from the TOAR database, with the exception of the baseline values at Mace Head, which are from Derwent et al. 2018.

The grey line shows unfiltered ozone at Mace Head Ireland, while the black line shows ozone filtered for baseline conditions (from Derwent et al., 2018). The baseline data show a peak in spring and a distinct summer minimum. Over the course of a year the average ozone value is 40.1 ppbv. Moving eastward we show the rural research station of Weybourne on the east coast of England (adjacent to the North Sea). The impact of emissions and depositional processes across Ireland and the UK are evident, as wintertime ozone at Weybourne is much less than at Mace Head, while summertime ozone is greater. Annual mean ozone is 6 ppbv less than at Mace Head, indicating a net loss of ozone. Two other sites on the North Sea coast (Norderney and Westerland) behave similarly to Weybourne. Zingst on the northern German coast also behaves similarly to the North Sea sites; annual mean ozone is 8 ppbv less than at Mace Head, again indicating a net loss of ozone. Hohenpeissenberg is quite different, showing a broad summer peak and a wintertime minimum, typical of polluted inland sites. Pollution influences at Hohenpeissenberg can also be seen from Figure 3, which shows the diurnal cycle of ozone mixing ratios at this site for different seasons averaged over two decadal periods. In particular, the range of the diurnal cycle is maximum in summer when photochemistry dominates.

Hohenpeissenberg diurnal cycle of ozone mixing ratios

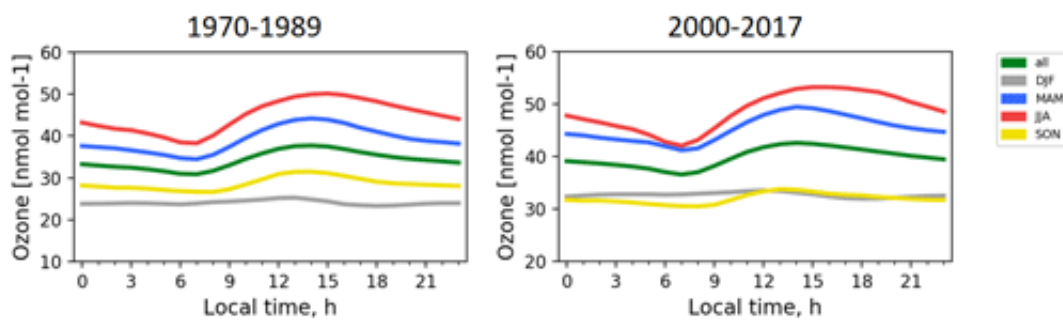


Figure 3. Diurnal cycles of ozone mixing ratios at Hohenpeissenberg for different seasons and averaged over two decadal periods. All data are from the TOAR database. Note the different y-axis scales on the two panels.

Clearly, the interior sites behave differently from Mace Head and should not be used to infer trends of baseline ozone.

Unsupported hypothesis:

The Parrish et al. paper is centered around the following hypothesis:

“...before the natural ozone distribution was perturbed by anthropogenic emissions of ozone precursors, the ozone gradient was reversed compared to that of today, with concentrations higher in the SH than the NH at mid-latitudes.”

This is an intriguing suggestion for which there is some discussion in the literature of 40 years ago. However, there are no observations from the pre-industrial era that can support this hypothesis. There aren't even any data prior to 1970 in the mid-latitudes of the SH. Older data in the SH (Galbally and Roy, 1980; 1981) like those in Figure 3, show values indistinguishable from the historic averages for the NH.

The authors extrapolate the Cape Grim data back in time using standard linear regression. This is a doubtful exercise as there is no rationale to explain why ozone changes at Cape Grim should have been linear over extended periods of time prior to the beginning of the instrumental record. Parrish et al. then use their estimate of the NH trend to argue (their Figure 3) that this trend implies that ozone was lower in the NH in the pre-industrial era.

They ignore the fact that their extrapolation of the NH data (their Figure 3) puts the NH average well below the actual measurements (points 1&2, above). Because the models do not show this hypothesized reverse gradient, the authors conclude that the models are flawed. This conclusion, we are told, is necessary because “present NH ozone concentrations are less than a factor of 2 greater than those in the SH”.

However, there is a simpler explanation for this alleged “inconsistency”, namely that ozone has NOT increased in the NH by 110%, but by a smaller amount, as found by *TOAR-Observations*. Although they are not cited, the recent analysis of ice-core data by Yeung et al. (2019), and the independent analysis of aircraft and balloon data in *TOAR-Observations* both support a smaller increase of tropospheric ozone. We therefore conclude that there is presently no evidence to suggest that pre-industrial model simulations overestimate historic ozone levels, and we see no reason to criticize the models.

Other Issues:

- Parrish et al. 2021 make the following claim about the Cape Grim ozone data: "The annual mean Cape Grim data were downloaded from the TOAR data archive (Schultz et al., 2017; <https://join.fz-juelich.de/access/>, last accessed 20 April 2020); they were selected for baseline conditions as described in the TOAR data header." This statement is incorrect. The TOAR Surface Ozone Database does not contain a baseline-selected subset of Cape Grim data, and there is nothing in the header information that would lead the user to such a conclusion.

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The following statements by Parrish et al. (2020) can give the impression that the conclusions of *TOAR-Observations* support the findings of Parrish et al. (2021):

"As part of the Tropospheric Ozone Assessment Report (<https://igacproject.org/activities/TOAR>), Tarasick et al. (2019) critically reviewed the record of historical ozone measurements throughout the global troposphere. Parrish et al. (2020a) have recently synthesized the HTAP and TOAR analyses."

"However, the accuracy of relative long-term ozone changes derived from these data is supported by the critical evaluation of Tarasick et al. (2019), which found no significant, systematic inaccuracy in the historical data analyzed by Parrish et al. (2012; 2014)."

As authors of *TOAR-Observations* we would like to clearly state that the conclusions of *TOAR-Observations* do not support the claim by Parrish et al. that baseline ozone has increased by a factor of 2.1 +/- 0.2.

Please also note the supplement to this comment:

<https://acp.copernicus.org/preprints/acp-2020-1198/acp-2020-1198-CC1-supplement.pdf>