Atmos. Chem. Phys. Discuss., 2, S583–S588, 2002
www.atmos-chem-phys.org/acpd/2/S583/

Atmospheric Chemistry and Physics Discussions

Interactive comment on "An exploration of ozone changes and their radiative forcing prior to the chlorofluorocarbon era" *by* D. T. Shindell and G. Faluvegi

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Received and published: 5 November 2002

We thank Johannes Staehelin for his constructive comments. Dr. Staehelin raises two main issues with the paper, namely the evaluation of the 1957-1975 column ozone measurements and the use of the nineteenth century Schönbein data. We address those topics first, followed by a point-by-point response to the other minor comments.

The reviewer suggests the use of NCEP/NCAR reanalysis 100hPa temperatures and 300hPa geopotential heights to screen for breaks in the ozone time series as deposited in the World Ozone Data Center. We agree that given the relatively strong correlation between ozone column amounts and meteorology on short timescales, appropriate correlations could be derived for each station and then a criterion developed to test for sudden deviations from that correlation. However, we believe that to execute such

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a project rigorously (i.e. finding the most appropriate correlation timescale, deriving criteria for deviations, investigating mismatches between the location of ozone observing stations and the coarser, gridded NCEP data, etc.) would require an enormous amount of time and effort (which is probably why the Data Center contains only uncorrected data), and is beyond the scope of our investigation. Our purpose has been both to demonstrate that the early data are potentially important and may be able to shed light on early ozone trends which are presently very poorly constrained, and to develop a framework in which all the available early data could be used together to improve the reliability of trend estimates. We feel that the present analysis has accomplished both these objectives, and that since the analysis relies on several independent data sources, this result would not be compromised by a revision to one of the early data sets. Indeed, the framework should be well suited to incorporation of re-analyses of early data such as the recent water vapor studies or the improved column ozone analysis suggested by the reviewer. We hope that either our group or another will in fact be able to perform such an analysis in the future, which we agree would be valuable and would present an improved result for the column trends over that shown here (we will now state this in the Appendix).

While our analysis is certainly not the final word on trends from the column data, we have gone to considerable length to ensure that the current work is objective. Our method avoids any arbitrariness in removing stations based on disagreement with nearby stations, potential very large SO2 contamination, etc., which we felt would be unjustified without a much more detailed analysis. Maximizing data coverage is also an important issue given the inhomogenaity of tropospheric ozone. The method we used does check the ozone data themselves for breaks, and omits sites with very large variability. It is also thoroughly documented in the Appendix, and is straightforward enough that anyone could reproduce it. We believe it is a significant improvement over the last published estimates of trends during this time, which used minimal screening and found much larger NH trends (see section 2 for references to the earlier papers, primarily from the 1970s, and the trends derived therein). We feel these are impor-

ACPD

2, S583–S588, 2002

tant advantages. That said, however, we agree that data quality remains a significant problem for the column ozone data. Most of the trends shown in Table 1 indeed do have a large uncertainty. As we note in the Appendix, 25 out of the 31 NH stations overlap with the best estimate NH troposphere trend using their 2-sigma uncertainties. In many cases this simply reflects the large magnitude of those uncertainties. Yet this may indicate that those uncertainties have been reasonably estimated. Since our trend from these many stations is calculated using an uncertainty weighted mean, the more reliable stations will have much greater weight. We feel that this reduces the impact of potentially less reliable stations, without sacrificing coverage or using arbitrary criteria. Of course this only accounts for random errors, not systematic ones. But those are extremely difficult to quantify. A thorough analysis of SO2 trends is another complex project that would improve the usefulness of these data, and would allow correction for a source of systematic biases, but is also beyond the scope of this paper. (We note the referee's comments regarding Uccle, which may have had decreasing SO2 pollution since the 1980s, but feel that without a systematic study of SO2 contamination, exclusion of just this one station would be somewhat arbitrary. In general, we hope that since legislation requiring cleaner emissions went into affect in many industrialized nations in the 1970s, our 1957-1975 period will not be greatly affected by changes in SO2 emissions). We sincerely hope that our paper will stimulate further work, such as that proposed by Dr. Staehelin, on the early data to improve trend estimates, which can then be used with other available data to better constrain the overall trend estimates following the method we have demonstrated.

The second main issue raised by this reviewer concerns the reliability of the nineteenth century surface ozone measurements. We thank the reviewer for his extremely detailed commentary on this issue, which we largely agree with. We are used to modeling studies comparing with these measurements, but having been convinced by the reviewer and a rereading of the relevant papers, we concur that the Schönbein papers are in fact poorly suited to quantitative analysis. Though the Montsouris arsenite method observations are quantitative, they are for only one location. So we will point out explicitly

ACPD

2, S583–S588, 2002

in the text that we do not believe that the preindustrial surface data are quantitatively reliable. In light of this, we feel that it is useful to see what the model gives if set to match these preindustrial values only so that we may compare with other model simulations, which frequently also compare with these data. Given that investigation of the preindustrial troposphere then becomes largely a modeling exercise, we will remove mention of this from the abstract and conclusion, and substantially reduce the discussion of this portion of the work, leaving only a brief comparison with other models.

For persons following this interactive discussion, we note that the Linvill paper discussed in the reviewer's comments on this issue is from 1980, not 1880 (and so we hope that the referee will see how fallible we all are, and forgive us misspelling his name).

A last point on this topic relates to two comments from the referee regarding the Schönbein data and the preindustrial emissions. He says that "If the data are suspicious it makes no sense to conclude that the emissions are not adequately known". We did not draw the latter conclusion from the data, but from the lack of constraining data on the early emissions. The referee continues: "I can not see any argument why the Schönbein measurements should be more reliable than the generally used emission estimates." We agree, in the sense that we feel that both are very unreliable. The preindustrial emissions from biomass burning, for example, are usually set to 10% of present day, but this value is completely arbitrary. Production of NOx from lightning is another example of a source that is unconstrained in preindustrial times.

Detailed comments:

Before number 1) The referee mentions relevant work regarding a compensating effect between stratospheric ozone decreases and tropospheric ozone increases. We agree that this work is relevant, and will add a reference to it in the text.

1) We'll add "20th century" to the abstract to make clear what time we're comparing with.

ACPD

2, S583–S588, 2002

2) A footnote to Table 2 says that our results are broadly consistent with those of Dvortsov and Solomon (2001), which is true. It is difficult to compare exactly, however, as the changes in water vapor are fairly different. Additionally, the radiative response to water vapor changes seems to vary significantly between models (see Oinas, V., A. A. Lacis, D. Rind, D. T. Shindell, and J. E. Hansen, Radiative cooling by stratospheric water vapor: big differences in GCM results, Geophys. Res. Lett., 28, 2791-2794, 2001), which may account for some of the differences.

3) We will add more description of the model runs to section 3.

4) We will add a reference for the aerosol loading as a function of time (Sato et al., 1993).

5) The referee asks which emissions were used for the 1950s and 1970s. Inventories of emissions for these time periods are not yet available, so we used the published 1990s emissions and preindustrial emissions, since changes in anthropogenic emissions are best quantified at these times, as noted in the text. We then scaled the model results to match the surface observations from the 1950s and 1970s (see text and Figure 2). Though not perfect, until emissions have been well studied and are made available to the modeling community, it is not possible to quantitatively constrain tropospheric ozone from GCM studies. (The recent work of van Aardenne et al, which we will reference, is a big step in this direction, though the data themselves were not yet available to modelers as of our last request).

6) See reply to first main issue above.

7) The uncertainty-weighted calculations for both the total column trend derived from the individual station trends and the overall multisource tropospheric and stratospheric trends were simply weighted according to the inverse square of its uncertainty, following standard statistical practice. The uncertainty on that mean is also derived from the sum of the inverse squares of the individual uncertainties (the square-root of one over that sum), again following standard practice. We will describe this more explicitly in the text.

ACPD

2, S583–S588, 2002

8-9) There are no comments 8 and 9.

10) Spelling corrected, and please accept our apologies.

11) The reviewer questions the scaling of the pre-1957 water vapor trend by modern estimates. We agree that this is quite uncertain, and will now point out that this is so. We performed the model runs for this early period both with and without these trends, in fact, since they were so uncertain. The main point, however, is that even with extrapolated trends in the greenhouse gases (including water), the ozone change during the period 1850-1975 is almost the same as that during 1957-1975 (see Table 2), so that there is little additional changes. We will rewrite the discussion of these results to emphasize that point.

12) For the model exercise to match the purported 'preindustrial surface ozone measurements', we simply prescribed that the model values match by removing an appropriately-scaled anthropogenic ozone distribution (Figure 1), and not by setting emissions. Like other models, ours fails to reproduce the 'observations' if we use the standard assumptions for preindustrial emissions. This will be described more clearly in the text.

13) See reply to second main issue above.

Interactive comment on Atmos. Chem. Phys. Discuss., 2, 1371, 2002.