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Atmospheric Measurement Techniques Discussions

Interactive comment on "Flexible approach for quantifying average long-term changes and seasonal cycles of tropospheric trace species" *by* David D. Parrish et al.

David D. Parrish et al.

david.d.parrish.llc@gmail.com

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We are grateful for the referee's careful reading and helpful comments. Following are the comments by the referee (*in italics*) and our response (in plain text):

1. The power series fit as denoted by Equation 1 is certainly more useful compared with the normally used linear fit method. Nevertheless, could the authors describe the possible physical meaning of the changes subjected to t^2, t^3, t^4, : : :? In short, why power series fit? To fit the nonlinear trend, we could also try other function forms to describe the atmospheric oscillation. For example, if we think the ozone change may partly or largely be driven by the temperature, we may think a form of equation to

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describe the temperature change with the time.

This is a very important comment, and applies to the use of Fourier series to describe the seasonal cycle, as well as to the power series to describe the long-term changes. In our original submission we briefly discussed this issue in the final paragraph of the Introduction.

From a formal mathematical perspective, these series simply provide a means to quantify the long-term changes and seasonal cycle in a time series of measurements with the fewest possible statistically significant parameters. There is no implication that the individual terms have a physical cause, so one cannot necessarily attribute physical significance to any of the terms. Both series are mathematical representations of the respective systematic temporal variations; their utility arises because these mathematical representations are flexible enough to capture the statistically significant temporal variations in the time series. Bowdalo et al. [2016] discuss this issue for Fourier series quantification of temporal variability, including higher frequency temporal variations.

However, this formal perspective does not necessarily imply that one or more terms do not have an underlying physical cause. For the Fourier series, the fundamental is attributed to the yearly seasonal cycle, and indeed the form of Equation 3 is designed to facilitate that attribution (i.e., χ spans one year's time period in radians from 0 to 2π). However, in the case of ozone in the marine boundary layer, Parrish et al., [2016] identify a physical process as the cause of the statistically significant second harmonic - the second harmonic of the photolysis rate of ozone, i.e., j(O^1D), which causes a second harmonic in the loss rate of ozone. However this identification required information and analysis beyond that in the time series of concentration measurements.

If we have reason to believe that one or more other factors drive some of the temporal variability in a time series of concentration measurements (e.g. ozone change driven by temperature change) we could certainly fit a functional form that represents those causes through consideration of additional physical variables. However, that is beyond the scope of the present work, as our goal is to develop a flexible analysis approach that does not require physical information beyond the time series of the concentration measurements.

Some of this discussion has been added to the final paragraph of the Introduction in the revised manuscript: "Without an underlying physical model, care must be exercised in the interpretation of the derived parameter values, and in the attribution of a physical cause to any of the terms in the series. Parrish et al. [2016] do present evidence of a direct physical cause of the statistically significant second harmonic of the seasonal cycle of ozone in the marine boundary layer (MBL) by showing that the photolysis rate of ozone, i.e. j(O^1D), which drives the loss of ozone in the MBL, also has a second harmonic of opposite phase to that of ozone's seasonal cycle. However this identification required information and analysis beyond that of the time series of concentration measurements alone."

2. The information of Figure 2 is quite limited which I think can be removed from the main text.

Thank you. Figure 2 has been removed, and the following figures renumbered.

3. The Fourier Transform for the detrended monthly change of ozone concentrations shown in Figure 3 is very interesting but the information from Figure 4 is basically the same with a different view angle. I suggest to merge the two figures as one figure and assigned with two panels.

Thank you. However, due to some unfortunate technical issues, combining Figures 3 and 4 would be quite difficult; they have been retained as separate, newly numbered Figures 2 and 3.

4. The section - 'The rate of change of the concentrations' is certainly very interesting, as the rate of change can be derived consequently as a differentiate of equation 4. Nevertheless, as long as the physical meaning is not clear, I suggest to remove this *part or to add more discussions on this part to abstract the possible meanings of the phenomenological analysis with its theoretical background.*

We agree that this is an interesting section. However, as discussed in the response to Comment 1) above, we cannot in general attribute a physical cause to the rate of change of concentrations. We have retained this section, and added sentence to the revised manuscript discussing the utility of quantifying the rate of change of concentrations: "Acceleration or deceleration in the rates of change of a species may contain information regarding changes in the magnitude of sources or sinks of the species, and thus may lead to improved physical understanding of the processes that determine the observed atmospheric concentrations."

Technical comments:

The time axis 1/1/99, 1/1/00, 1/1/10 better changed to be 1/1/1999, 1/1/2000, and 1/1/2010 or 1999, 2000, and 2010

Thank you. The time axes on all relevant figures have been changed to 1999, 2000, and 2010.

The legend of Figure 1.: the unit of parameter c, should it be ppb yr^{-2}

Thank you. This typo has been corrected.

Figure 5: it would be helpful to add a description of the violet line in the figure legend

Actually, the violet line was already briefly described in the figure legend (now legend for Figure 4); no change has been made here.

References

Bowdalo, D.R., Evans, M.J., and Sofen, E.D.: Spectral analysis of atmospheric composition: application to surface ozone model–measurement comparisons, Atmos. Chem. Phys., 16, 8295-8308, doi:10.5194/acp-16-8295-2016, 2016.

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