Interactive comment on “Spectral analysis of atmospheric composition: application to surface ozone model-measurement comparisons” by D. R. Bowdalo et al.

Anonymous Referee #2

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Description: This discussion paper describes application of spectral analysis, specifically the Lomb-Scargle Periodogram (LSP), to illuminate temporal patterns in observed and modeled hourly surface ozone, globally. Compared to the Fourier transform, the LSP is better equipped to handle missing data and has the ability to estimate at any frequency. The authors walk through the spectrum for one site, Cape Verde, in detail and then present global patterns in both the amplitude and phase for 710 sites and GEOS-Chem predictions. Seasonal and diurnal processes are responsible for about 50 percent of variability in ozone, on average, while the remaining variability stems from weather and “macroweather” (>10 days) timescales. In general, modeled ozone overestimates the seasonal amplitude and produces a phase that is 1-5 months compared to observations. The authors attribute these biases largely to uncertainties in emissions.

Relevance: Spectral analysis is a powerful method for detecting cyclical patterns in data, and the LSP seems especially apt for analysis of air quality data. The analysis encompasses both observed and predicted mixing ratios, facilitating inspection of model biases that is different from the usual time-domain perspective. Adopting this perspective on a global scale produces interesting insights into how the underlying processes vary at different sites across a spectrum of latitudes, climates, and anthropogenic influence.

Assessment: Comparison of spectral patterns in both observations and predictions is a strength of the work. The methods are exceptionally clearly explained, the figures are informative, and interpretation of the result is reasonable. Overall, the writing is well organized and very clear; the paper is interesting and a pleasure to read. Suggestions for improvement are listed below.

Specific comments

1. p. 3, lines 3-11: Without proper context, the authors seem to be implying that this is the first time the LSP has been applied to air quality data. It would be useful to cite other papers that have utilized the LSP to analyze air quality data, such as Dutton et al., Temporal patterns in daily measurements of inorganic and organic speciated PM2.5 in Denver, Atmos Environ. 2010; 44(7): 987–998.

2. p. 4, line 17: Specify the temporal resolution of the surface ozone data from Cape Verde, and describe the location in terms of where it is, its climate, and degree of human development. Why Cape Verde? Including a contrasting site with nearly opposite characteristics would provide a nice counterexample.

3. p. 5, lines 13-18: This paragraph seems like it would fit better in section 3.2, Annual and daily cycles, and some of it is redundant with material in that section.
4. p. 5, lines 13-14: “From Fig. 2 it is evident that there are significant peaks at the annual and half annual timescales, and at the daily, half daily, and third daily timescales.” If amplitudes above the 99th percentile are defined to be significant, then I do not see “significant peaks” at the half annual (\(~182\) days) or third daily (\(~0.3\) days) timescale. There are certainly peaks in amplitude above the red line at periods of 365 days, 1 day, and 0.5 days in Fig. 2.

5. p. 5, line 32: The distinction between “seasonal” vs. “annual” cycle is not clear throughout the paper, and this would be a good place to distinguish between the two terms rigorously, or to state that they mean the same thing. Yashayaev and Zveryaev, Climate of the seasonal cycle in the North Pacific and the North Atlantic oceans, Journal of Climatology 2001; 21(4): 401-417 did a nice job of defining the annual cycle as the first harmonic only and the seasonal cycle as the sum of the annual, half annual, and harmonics.

6. p. 6, line 28: The large variance from the seasonal cycle at the Antarctic and continental Southern Hemisphere sites may also be due to low anthropogenic influence, in addition to spatial homogeneousness.

7. p. 9, line 11: “Regions with significant annual cycles…” This is an example of where the distinction between annual and seasonal cycles is unclear.

**Technical corrections**

8. p. 8, line 6: “peak” should be “peaks”

9. Fig. 5: Define the abbreviations NA, EU, AS, and ROW.

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