

Review of " Baseline carbon monoxide and ozone in the Northeast U.S. over 2001 - 2010" by Y. Zhou et al.

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**Summary:**

This paper presents and analyzes a large data set that is a 10-year record of ozone and CO measurements at 7 sites in the northeast U.S. The authors have done a wide variety of analyses, described in long discussions, but in the end, they can draw no firm conclusions. The work as it stands has major shortcomings. I suggest that this paper be rejected. The following comments describe my concerns.

**Major issues:**

- 1) The title of the paper emphasizes my first concern. The term "baseline" concentration has a specific meaning, which I believe was originally defined in the 2009 HTAP Report entitled "*Hemispheric Transport Of Air Pollution 2010*:"

"A baseline concentration is an observation made at a site when it is not influenced by recent, locally emitted or produced pollution. These baseline sites are typically situated in locations with minimal and infrequent impact from local sources of anthropogenic pollution. Observations may be made continuously and subsequently sorted, or air samples taken only when meteorological conditions are such that the recorded concentrations are free from the local contamination. Time series of baseline concentrations provide the range and frequency of pollutant concentrations transported to the site from upwind locations. However the requirement that only recently emitted or produced local pollution be excluded means that baseline concentrations may contain traces of local pollutants that were emitted many days earlier and became well-mixed with other air masses. There is no strict definition of "recently" emitted or produced local sources of anthropogenic pollution."

The current paper selects data to analyze that is labeled "baseline" but in fact no means was identified to determine the influence of "locally emitted or produced pollution". I do not believe that it is possible to determine baseline concentrations of CO or ozone at any site in the northeastern US, since the region is surrounded by pollutant sources, and most of the North American continent lies upwind of that region. Furthermore, characterization of baseline concentrations requires defining the distribution of concentrations that are measured under baseline conditions. They may be low concentrations, but they may also be high concentrations. For example, baseline concentrations of CO may be elevated if anthropogenic emissions or wildfire emissions are transported to the site from a distant continent, and baseline concentrations of ozone may be greatly elevated if a stratospheric intrusion is transported to the site.

- 2) What the authors attempt to do in this paper is to determine monthly average concentrations of an approximate regional background of CO and O<sub>3</sub>. It may be that a useful paper could be written discussing such background concentrations, and how they vary with the many variables that the authors consider. However, even such an analysis is compromised because the authors are not successful in describing a regional background that is actually regional. This is clear from Figure 2 where time series of the results are plotted. The background concentrations of CO and ozone determined for each of the seven sites are not the same. There are certainly similarities, but there are also large, unexplained differences. In my view, the authors must reconsider their analysis to arrive at two time series: one a monthly average regional background CO concentration, and the other a monthly average regional background ozone concentration. It is not clear to me that this can be

achieved, but in Figure 2 there appears to be enough similarity between sites that it may be possible to derive time series that represent a regional background for the northeastern US.

- 3) It is clear that there are experimental problems in the data set. In the discussion the authors note unexplained differences between results at different sites. Some of these are certainly due to experimental problems. Before proceeding with any attempt to define a regional concentration, the authors must undertake a critical evaluation of their data sets to eliminate confounding experimental problems.
- 4) Pages 27263-27280 give an extended discussion of many topics. Much of this discussion is speculation regarding causes of trends, variability and correlation perceived in the data. For a useful paper to eventually arise from the authors' analysis, the speculation must be removed, and clear, well-reasoned discussion must be substituted. I suggest that the authors focus on a limited set of hypotheses that they feel they can discuss in a rigorous and complete manner.

**Minor issues:**

- 1) Figure 2 is confusing. The station labels in the annotations are not in accord with the figure caption. It is not clear which curve corresponds to which station.