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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Air quality and atmospheric deposition in the eastern US: 20 years of change" *by* J. E. Sickles II and D. S. Shadwick

Anonymous Referee #3

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This manuscript extends earlier analyses of measurements from the Clean Air Status and Trends Network (CASTNET) and the National Atmospheric Deposition Program (NADP), focused on 33 CASTNET sites located east of the Mississippi River plus one site in Arkansas. The measurement locations and methods of analysis are the same as described previously in Sickles and Shadwick (2007a; 2007b). As noted by the authors, one of the earlier papers examined two five-year periods (1990-1994 and 1995-2000), while the second additionally considered a third period (2001-2004). The present paper adds a fourth five-year period (2005-2009). CASTNET and NADP are major U.S. air quality monitoring efforts, potentially yielding important insights into changes in air

quality across large regions. The analysis provides a valuable broad-based assessment of changes in deposition and ambient air concentrations, and the likely relation of each to emission changes.

Given the previous work, many readers will expect the present paper to indicate what has been learned by analyzing an additional five years of data, such as by systematically comparing current conclusions with the findings from the previous studies. The authors do not present such a systematic comparison of conclusions, but in their summary they do state what are probably the most important similarities and differences. Similar to previous conclusions, the analysis of 20 years of data continues to show that ambient SO2 concentrations and deposition have decreased in approximately 1:1 proportion with SO2 emission decreases, whereas ambient sulfate concentration and deposition decreases are in less than 1:1 proportion. An important new finding is that ambient aerosol nitrate concentrations decreased between 1990-1994 (period 1) and 2005-2009 (period 4), in contrast to the earlier work that showed increases in ambient aerosol nitrate concentrations from 1990-1994 to 2000-2004. The authors' explanation of the different results for nitrate (e.g., last sentence of the abstract) borders on the speculative, however. It would be better to simply state that the longer monitoring period has revealed a different trend. Also, rather than introducing the nitrate changes as an illustrative example (last paragraph of abstract), possibly leading readers to wonder why this example has been chosen instead of other possible comparisons, it would be appropriate to explicitly state that the findings for aerosol nitrate represent the most important change between the conclusions of the present and earlier papers (if the authors concur that this is the case).

The statistical methodology is well documented in the authors' earlier papers. It is fundamentally a two-sample comparison carried out site-by-site and by region. In the present paper, the focus is on P1-to-P4 comparisons (1990-94 compared to 2005-09). It would seem that the analysis loses information by essentially ignoring the intervening years. Could the authors comment on this point?

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The authors present results for ozone in Section 3.7, but do not discuss them in Section 4. As noted in Section 3.7 and Figure 4, the largest ozone concentration decreases (6 - 17%) occur during summer. These decreases are substantially smaller on a percentage basis than the NOx emission decreases of 39 - 46% (Table 4). While ozone is not expected to decrease in 1:1 proportion with emission decreases, providing a comment on the result would be appropriate. The authors propose an explanation for the seasonal differences in ozone response in their summary (p 17978, aggressive ozone-season NOx emission controls), but do not provide seasonal emission data to support this statement. Are other explanations possible?

On page 17950, lines 13 - 24, the authors note that data from 34 locations can only be an approximate representation of regional air quality. At the authors' discretion, this would be an appropriate place to note that the CASTNET sites (13 of 14) cover only the northern half of the South. Other caveats may also be useful to note.

Figures 5 through 8 are difficult to read. Simple data tabulations might be preferable.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 17943, 2014.