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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

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Response to the comments of Referee #3. Thank you for taking the time to examine our manuscript and for providing useful comments.

Response to first comment. In the second paragraph of our abstract we explicitly listed over half of the approximately 50 metrics that we considered in our study. In the last paragraph we wanted to say what we did with those metrics (examined period-to-period changes) and provide at least one example of an important finding that could be articulated with minimal verbiage. I can't say that our choice is the most important finding

of our study, but it is an important one. Most of the important findings are given in the summary and conclusions (Section 5) of our document.

Response to second comment. Our study was designed to examine associations between legislated reductions of emissions and changes of monitored air quality and deposition in the eastern US by comparing 5-year period mean values of metrics derived from 20 years of monitoring data collected at the same sites. This study approach used the first period as the baseline for most comparisons, because it represented conditions prior to emissions reductions triggered by the Clean Air Act Amendments of 1990 and by other legislated emissions reduction programs. Using our statistical approach, six comparisons could be made (i.e., P1-to-P2, P1-to-P3, P1-to-P4, P2-to-P3, P2-to-P4, and P3-to-P4). Our earlier manuscripts have focused on P1-to-P2 and P1-to-P3 comparisons, emphasizing the latter. Our current manuscript focuses on P1-to-P4 and P3-to-P4, emphasizing the former by maintaining P1 as our baseline. Our approach may have resulted in losing information available from comparisons involving intervening years that received less attention (i.e., P1-to-P2, P2-to-P3, P2-to-P4, and P3-to-P4).

Response to third comment. Ozone (O3) is a secondary air pollutant that accumulates near the earth's surface from photochemical reactions involving sunlight and the precursors, volatile organic compounds (VOC) and nitrogen oxides (NOx). The O3-formation chemistry is highly non-linear, and as a result 1:1 relationships between O3 and its precursors are not normally expected. However, O3 may accumulate to levels that pose risks to health, especially in the summer, with long durations and high intensities of sunlight and high ambient temperatures. There is evidence that O3 accumulation in the rural areas of the eastern US is limited by NOx rather than VOC concentrations. This concept resulted in legislated O3-season (i.e., May to September) controls of NOx emissions from large combustion sources (primarily EGUs) starting in 1999 (see references in the text to these programs: OTC, NOx SIP Call, and BTP). This was an attempt to reduce O3 accumulation at the time of year (summer) when it

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was frequently a problem by reducing emissions of an important (and likely controlling) precursor, NOx. It is recognized that the accumulation of both primary and secondary pollutants vary from year to year with changing emissions and meteorological conditions. As a result, Camalier et al. (2007, Atmos. Environ.) developed a method for adjusting O3 data for year-to-year changes in important meteorological variables that are associated with O3 accumulation (i.e., daily maximum temperature, relative humidity, wind speed, and wind direction). The behavior of the resulting met-adjusted O3 might then be considered to depend mainly on the behavior of emissions (although absent the assumption of constant VOC emissions, it is difficult to narrow the cause only to NOx emissions). Other researchers have also developed methods to adjust O3 data for variable meteorology (e.g., Gego et al. (2007, JAM) used multiple regression; and Chan (2009, JGR) used rotated principal component analysis with generalized linear mixed models). These researchers examined the behavior of 1997-2004 and 1997-2006 met-adjusted O3-season O3 concentration in the eastern US. Butler et al. (2011, Atmos. Environ.), using Camalier's method, conducted a similar study for the period 1997-1999 to 2006-2008. Our comparisons using 5-year period means attempts to account roughly for year-to-year variability in meteorological conditions on O3 concentration (as well as other metrics). Summer P1-to-P4 O3 reductions reported in our current manuscript are numerically larger than those for P1-to-P3 (SS07b). In both cases we note that this behavior coincides with aggressive O3-season NOx emissions controls. In addition, both of these sets of findings are consistent with those in the previously noted citations by Butler, Gego, and Chan. Each of these studies reports significant reductions in ambient O3 concentration in the eastern US that coincide with time periods when legislated O3-season NOx emissions controls were in place. Each of these studies shows associations (not necessarily causality) between reductions in ambient O3 concentration in the eastern US and legislated O3-season NOx emissions. However, neglecting any unfavorable impacts of changes in VOC emissions during these periods reduces the likelihood of other explanations for this O3 behavior.

The first part of the previous paragraph has provided the rationale for aggressive O3-

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season NOx emissions controls (which apply primarily to EGUs). It should be remembered that on a mass basis the mobile contribution is substantially larger than that of EGUs (e.g., we note in our manuscript that nationwide in 1990 it was 57% versus 25%). However, impacts of NOx emissions from elevated stacks of EGUs may be strong regionally via transport; whereas, ground-based mobile sources may be strong initially on a local basis before thorough mixing and transport on a regional scale can occur. Emissions estimates are frequently provided in terms of annual values (Xing et al., 2013, ACP), as illustrated in Fig. 1 of our manuscript. Estimates of O3-season NOx emissions between 1997 and 2008 for a similar geographical area are illustrated by Butler et al. (2011, Atmos. Environ.) in his Fig. 2. In addition, several of my EPA colleagues who conduct air quality modeling require fine temporal resolution of emissions as input for model runs. Informal discussion with them suggests that summertime reductions in NOx emissions in the eastern US for the period between 1990-1992 and 2006-2008 (roughly corresponding to P1-to-P4) are approximately 6% greater than those for the aggregate of the remaining three seasons over the same time period.

Response to fourth comment. As noted in our manuscript, the 34-site CASTNET does provide sparse geographic, regional coverage of the study area. This is illustrated in our Fig. 2, where coverage is especially sparse in the southern part of the South and in the northeastern part of the Northeast, and this will be noted in the revision.

Response to last comment. Regarding Figs. 5-8, these figures can be enlarged on the computer screen by using the view/zoom feature. The message we hoped to convey via these figures is an overall general view of period-specific regional concentration and deposition values, their period-to-period relative changes, and a general appreciation of site-to-site regional variability for these metrics across regions and the east. We grouped three or four illustrations together in single figures to permit the reader to compare the period-to-period regional patterns of species concentration, deposition, and relative change simultaneously for several selected co-pollutants (i.e., concentration of S, OxN, N and CASTNET PM in Fig. 5 and total deposition of S, OxN and N, as well

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as wet deposition of H+ in Fig. 6). We focused on period-to-period seasonal concentration (aerosol NO3 and CASTNET PM) metrics in Figs. 7 and 8. We did not address or discuss site-specific behavior in our manuscript, except as a member of a collective contribution to regional behavior. Our intention was to display an arms-length view of patterns. Admittedly, a fine level of detail (more than one or two significant figures is missing); however, this is consistent with our intended purpose. Note that the top-to-bottom order of site IDs in Table 1 is given according region (i.e., MW, NE, and SO) and is in the same order as shown in Figs. 5-8. In addition, the site-specific period-to-period relative changes may be read not only at the bottom (right-hand abscissa) of each illustration, but they are also given (and in finer detail) on the right-hand ordinate of each illustration. Finally, note that: the P1-to-P4 changes in the concentration of S, OxN, and N for the E, MW, NE and SO regions (shown in Fig. 5) are also tabulated numerically in Fig. 4; the P1-to-P4 changes in the total deposition of S, OxN, and N, as well as wet deposition of H+ for the E, MW, NE and SO regions (shown in Fig. 6) are also tabulated numerically in Fig. 4; and both summer and winter P1-to-P4 changes in aerosol NO3 concentration for the E, MW, NE and SO regions (shown in Fig. 7) are also tabulated numerically in Fig. 4. In our opinion, the use of more extensive data tabulations required to display the information shown in Figs. 5-8 is not preferable because that approach does not permit simultaneous visual comparisons of period-to-period behavior (and especially patterns) across sites, regions, species, and sometimes seasons, that we consider critical to conveying an understanding of our results.

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