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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Chemical climatology of the southeastern United States, 1999–2013" *by* G. M. Hidy et al.

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Title: Chemical Climatology of the Southeastern United States, 1999-2013 Author(s): G. M. Hidy et al. MS No.: acp-2014-401

Author response to reviewer comments We thank the reviewers for their helpful comments on this manuscript. The following are our responses to the issues raised by reviewers 2 and 3. Both reviewers requested that we add data from 2013 to the tables and graphs where possible, given data limitations. We have done so. Data available through 2013 are extracted from the public SEARCH data set, unavailable for all sites until recently, so that our results can readily be reproduced by other investigators. The publically accessible data set has been carefully inspected for errors using a defined

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Interactive Discussion

Discussion Paper

quality control and assurance protocol adopted by Atmospheric Research and Analysis, Inc. We have extended Table 1 and the figures in the text and in the supplemental material to include 2013, where data are available. The updated figures include Figure 6, 7, 8, 9, 10, 11, and S2, S3, S4, S5, S6, S7, S8, S9, S10 and S18. In the review draft, Figure 2, 3, and 5 have already appeared in the discussion paper through 2013. Included in Figure S2 and other figures is an extension of annual emissions in the region from the National Emissions Inventory, EPA models and activity data based on methods we used in previous studies. The addition of 2012 and 2013 data to the table and figures yields overall results similar to those found in the discussion paper. Our analyses of the SEARCH data go well beyond our earlier studies that ended with 2011 data. We find that significant relations exist between observations and total (not just electrical generating units) emissions of SO2 and NOx, including new relationships between observations and total emissions of NOx, EC and CO. We also find statistically significant relationships between observations and mobile source EC, OC and CO emissions, which were not included in earlier analysis. In Table 1, the regression results strengthen the relations between observations and emissions adding 2012-2013 data with improved variance estimates and statistical significance. Adding CO regressions to the table for BHM and CTR with mobile source estimates of CO indicates that mobile source CO emissions tend to drive CO concentrations both in BHM as expected and CTR. For Table 2, we retained the two comparison periods (1999 – 2002 compared with 2009 – 2012), because two of the sites were not operational in 2013. We prefer to use the same comparison periods for all four sites. Reviewer #2. The comments refer mainly to ambiguities or typos in the text that we correct as follows. The questions about the figures are addressed, and mainly concern extension of the data to 2013, which we have done for the revised manuscript. 1. P. 17107 line 28 unclear sentence– revise sentence to read: “Also of interest were the observations showing that winds repeatedly arising at night in the boundary layer potentially result in transport of O3.” 2. P. 17112 line 28-29 unclear phrase– revised sentence to read: “But the continued OC and EC increase to summer (Q3) is inconsistent from year to year, sometimes

ACPD
14, C6657–C6663, 2014

occurring in fall." 3. P. 17145 Figure 1. The gray regions are counties identified with the Birmingham and Atlanta metropolitan areas. The divisions are county boundaries. 4. P. 17118 line 6 unclear sentenceâĂŤrevised sentence to read: "The morning rise in O3 concentrations at the ground also corroborates the potential for air mass transport with down-mixing of O3 from a residual (upper boundary) layer during break-up of the nocturnal inversion layer 2-3 h after sunrise (Baumann et al. 2000)." 5. P. 17123 line 16– period after 06:00 changed to comma, colon removed from 0600 (h). 6. P. 17125 line 18– 23 June is correct. 7. P. 17125 line 24 corrected–plume is added after power plant. 8. P.17127 line 20 unclear sentence revised to read: "The retained water in particles provides an opportunity for both gas and aqueous phase oxidation reactions of NMOC to occur in the CTR environment." Reviewer #3 1. Value added elements of the current work relative to previous analyses. Besides the above comments, we also added a number of comments in the discussion supported by data, including examples for extinction coefficient trends and retained water, inorganic vs. non-inorganic sulfate, multiyear diurnal profiles, diurnal power plant emissions, NMOC intercomparisons, and species trends beyond earlier studies and trends specific to the SOAS experiment period, including directional wind and pollution roses. We add a sentence in the paper abstract noting extended data trend analysis from earlier work, and note this extension in the revised introductory and discussion sections as well.

The differences or similarities from earlier studies (Blanchard et al., 2013, a, b and c) are noted in the revised text. The results in this review include not only annual trends, but trends in terms of emissions changes based on the extension of the NEI data through 2013, and ambiguities identified with regional open burning. The issues associated with the origins of particulate organic carbon (OC) are introduced, with emphasis on the significance of primary source contributions to OC. Our analysis will go into more detail in a forthcoming paper (Blanchard et al., 2014).

2. CTR site descriptionâĂŤA tabular summary of local conditions is included in the text with Figure S1 showing the local nature of the site (See attached [new] Table S2). A

ACPD
14, C6657–C6663, 2014

summary of regional climatic conditions and agricultural and mixed forest surroundings is provided in the current text, and the proximity of major metropolitan areas is noted. The data in Figure 13 show ground level wind conditions in the early summer. Prevailing winds in summer are generally from the northwest, with occasional southerly flow added, and winds are generally light in summer.

We added notes on the proximity of parts of the Talladega National Forest, and include the attached table to give more detail about regional population centers potentially affecting CTR, as well as notes about point sources or transportation routes that may affect CTR, as given in the new table added to the supplemental material (see attached).

3. The regional representativeness for atmospheric chemistry has been discussed extensively in previous papers on SEARCH data, referenced in the manuscript. Representativeness can be seen in a revised Fig. 13 (attached), which shows the SOAS period in June-July compared with conditions averaged over 2000-2012. The near symmetry of direction in the pollution roses, with the exception of SO2, indicates generally that CTR represents conditions in the SEARCH domain "integrating" sources and meteorology as expected. Representativeness also can be deduced from Table 2 and Tables S3 and S4, as well as Figure S19.

4. Inconsistency of language "2nd wettest and coolest" condition in the period of data collection vs. "marginally" wetter and cooler. We use marginal to suggest that 2013 was within the range of conditions observed over the period 2000-2013, but was wetter and cooler than the multiyear average or the range found since 2000.

5. Table 1 initially used data through 2011 consistent with the emissions inventory. The inventory has been extrapolated to 2013 using methods discussed in Blanchard et al 2013c so that Table 1 now covers 2000-2013 (see above).

6. Statistical criteria adopted for using SEARCH dataâĂŤThe data were accessed from the ARA public data base. Completeness is implicitly represented by standard

ACPD
14, C6657–C6663, 2014

error (std. deviation/sqr t(n)). For PM2.5 mass and species n ranges from 54 days (50% of 1 in 3 day sampling) to 355 days (nearly all of every day sampling). For gases, daily means were compiled requiring 12 hours per day, with annual means requiring 200 days data availability. Alternate completeness criteria (requiring 75% of all hours and days) were examined, but resulted in a loss of annual averages without yielding appreciably smaller standard errors. 7. Nitrogen oxides, NMOC and O3âĂŤlack of background relative to CO due to lifetime differences between more reactive species and CO. Probably true. It is interesting to note that trends in SO4 appear to be showing a background level of about 1 μg/m3. 8. Meteorological conditions cited for Fig. 12– Corrected box plots are Fig. S13.

9. Fig. 13 directional pollution roses. This figure has been modified to compare the SOAS period in 2013 with overall average of 2000-2012 for the same period, June 1-July 15 (see attached). The figure is easier to peruse with regard to representativeness of SOAS data period. The figure also includes wind direction frequency (WF) comparison between the average and the SOAS experiment period in 2013. This addresses the prevailing wind question raised by the reviewer.

10. Typos or other edits: Reviewer #3 list are taken care of in revision for the final version of the paper. Reference Blanchard, C., Hidy, G., Shaw, S., Baumann, K., and E. Edgerton. Constraining Contributions to Organic Aerosol in the Southeastern U.S. using SEARCH Network Measurements. In preparation, 2014.

Table S2 (added to revision). Pollution Sources/Source Areas Potentially Affecting Centreville, AL <150 km distant. Source Direction from CTR Approx. Distance from CTR (km) Pollutants Comments Counties Jefferson (659,000)a NNE nad all (CO, SO2, NOx, VOC, PMx, O3; [NH3]) Contains Birmingham/Hoover Shelby (204,000)a NNE nad all Part of BHM metro. area Montgomery (226,000)a SE na all Contains Montgomery Tuscaloosa (200,000)a WNW na all Contains Tuscaloosa Bibb (22,000)a central na all Contains CTR, Brent Cities/towns Birmingham/Hoover (300,000)a NNE 80 all BHM Metro area Heavy industry, commercial, residential Montgomery (201,000)a SE

ACPD
14, C6657–C6663, 2014

120 all Commercial, residential Tuscaloosa (95,000)a WNW 45 all " Centreville (2,700)a – nad all Agriculture, Residential Brent (4,900)a – na all Agriculture, Commercial, residential Demopolis (7,500)a WSW 80 all Commercial, residential Point Sources Plant Miller N 84 SO2, NOx, PM2.5 2600 MWc Plant Gorgas N 82 " 1200 MWc Plant Gaston ENE 75 " 1800 MWc Green County power plant ESE 72 " 560 MWc Bowater Ltd. Mill ENE 80 SO2, NOx, S 50 MW/ paperc Mercedes Int'l N 30 VOC, NOx, PM2.5? Auto manufact. Int'l Paper Mill SE 78 SO2?, NOx, S 65 MW (waste fuel)/paper Weyerhauser Pine Hill Mill SSW 110 SO2, NOx, S 72 MW/paperc Transportation Interstate 65 E 45b NOx, VOC, CO, PM2.5 Major hway N-S Interstate 20/59 N 30b " Major hway E-W US Hway 82 na na " SE hway to CTR US Hway 80 S 59b " E-W hway thru Montgomery Road 5/25 na na " Local state rd. apopulation bnearest distance to location ccoal fired d na– not applicable– locally present at or near CTR site

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 17101, 2014.

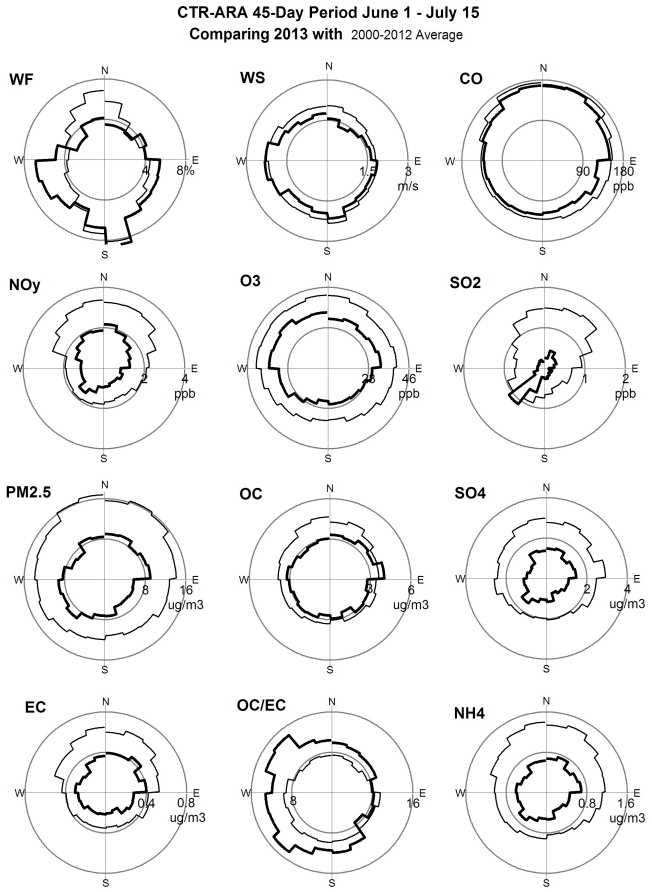

Fig. 1. Comparison of CTR average 2000-2012 winds, trace gas concentrations and PM2.5 mass and composition with the period 1 June-15July 2013

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Discussion Paper