

## ***Interactive comment on “Decadal change of summertime reactive nitrogen species and surface ozone over the Southeast United States” by Jingyi Li et al.***

### **Anonymous Referee #3**

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#### General comments:

This manuscript examines “decadal changes in summertime reactive nitrogen species and ozone over the Southeast U.S.”, and finds they “decline proportionally with decreasing NO<sub>x</sub> emissions in this region” and concludes that “this linear response is in part due to the nearly constant summertime supply of biogenic VOC emissions in this region”. There are several concerns with the overall quality of the current manuscript. (1) In the manuscript, some critical definition/terminology used are not accurate or ambiguous. For example, NO<sub>y</sub> refers to reactive oxidized nitrogen not reactive nitrogen, the latter includes NH<sub>3</sub>. It seems summertime is defined in the manuscript as

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July-August, but the three aircraft measuring campaigns, whose observations were extensively used to evaluate the modeling results and derive the changes in observed concentrations, were conducted at, respectively, July-August, 2004, June-July, 2013, and August-September, 2013. For regulatory purpose, surface ozone is studied for a period in a year defined as ozone season, which is usually defined as April-October in the Southeast of United States. (2) The decadal changes in both observation and simulations are not elucidated by using a well-designed comparison method. Reduction in NO<sub>x</sub> emissions are one of the major reasons that can cause the resulting reduced surface ozone and NO<sub>y</sub> concentrations, but it is not the only one. The method used in the manuscript is not convincing by removing other impacting factors such as meteorology and emissions reduction on other pollutants, which confounds the conclusions this manuscript makes. For example, the aircraft measurements were collected at different locations and different days/months, how exactly such measurements can reveal the real changes of NO<sub>y</sub> between the two years a decade apart. The model simulations were conducted for the same months for 2004 and 2013, a decade apart, but in what quantity are the impacts on species concentrations resulting from the differences in meteorology between the two years? (3) There is no quantitative evaluation results presented for the model simulation on surface ozone. But according to the description from the manuscript: “AM3 overestimates surface MDA8 ozone in both years by about 16ppb on average”, and “MDA8 ozone averaged ... is observed to decrease by 11 ppb (23% of observed mean MDA8 ozone in July-August of 2004)”, one can deduce that the overestimation of surface MDA8 ozone in July-August of 2004 and 2013 are roughly 33% and 43%, respectively. Note that the USEPA recommends a better than 30% of mean normalized error for surface ozone performance for regulatory modeling. With worse than the EPA recommended performance, the modeling results from this study are not that meaningful for surface ozone regulation purposes. (4) The organization and presentation of the manuscript cause a lot of confusions. The authors constantly blends trends found in observations and trends found in simulations next to each other without distinguishing them clearly. The purpose of the aircraft measurements and

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the surface observation, and the purpose of the simulations are not clearly presented. A lot of qualitative statements, only supported with citations of ambiguous supporting meanings.

Specific comments:

(1) Page 4, “high-resolution (50x50 km<sup>2</sup>)”. When conducting chemical transport modeling at regional scale, this is no way a high-resolution.

(2) Page 5 “. . . both inventories have a similar spatial distribution (Figure S1). Compare the two panels in Figure S1, apparently, the local maximum levels in the Southeast of RCP8.5 are somewhat 30% lower than the NEI2011 (no red spots are seen in the Southeast in the RCP8.5 panel). Also, why compared to NEI2011 version 1, why not the NEI2011 final version? More importantly, why don't just use NEI2011?

(3) Figure S3, why Florida surface ozone data were not included? This study is for the Southeast, which should include Florida.

(4) Page 9, lines 329-331, why aircraft measurements for biomass burning and urban plumes are excluded for the model evaluation?

(5) Page 9, lines 334-335, “. . . use model output sampled along the flight track with 1-min resolution”. How exactly this has been done? What are the time-steps of the model? What are the time intervals of the model outputs? Is this necessary since all the presented comparisons are for monthly averaged values anyway? Is there any statistical metrics calculated for the comparisons at the 1-min resolution?

(6) Table 1 and table 2, “Monthly averaged”, or two month (July-August) averaged? Table 1, Why not present the NO<sub>x</sub> emissions for the Southeastern US too, instead of for only North America totals? Are they still 40% reduction for the Southeast only? Also, how about those numbers of emissions amounts for the Southeast only used in the model compared to the NEI 2011 final version inventory? Also, what about anthropogenic emissions pollutants other than NO<sub>x</sub>, such as VOC, CO etc.?

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(7) Figure 7, there are bumps at around 30ppb in the 2013 simulations, but not seen from the 2004 simulation and any observations. Why those bumps?

(8) Page 10, lines 370-372, what is this “regionally-averaged NO<sub>y</sub>”? It seems jumped from the observations to simulations here?

(9) Page 10, line 369, “This is likely due to the different sampling regions (Figure S4) from the two campaigns”. If this is the case, then why you can trust the other derived reduction numbers from comparing the observations from the two campaigns? And why you can trust the changes derived from these observations to represent the real changes in the Southeastern US as a region?

(10) Page 18, lines 649-651. What are the quantitative differences in both simulated and observed RH and temperature between 2004 and 2013 in July-August? What about the differences in cloud cover, precipitation etc. that also impact on ozone formation? Lines 654-657, this statement, for changes between 2004 and 2013, is not supported by convincing evidence. How exactly the citation in lines 651-654 supports this statement? Since this is also the base for deriving the major finding: “reactive nitrogen species and ozone over the Southeast U.S.”, “decline proportionally with decreasing NO<sub>x</sub> emissions in this region”, solid demonstration of this statement is needed.

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