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 #1

Review of "Decadal change of summertime 1 reactive nitrogen species and surface ozone over the Southeast United States" by Li et al.

This manuscript investigates the ozone and reactive nitrogen changes over the southeastern US (SE) using a high-resolution global model (AM3), applied, apparently, to July and August of 2004 and 2013. They also look to see what a further 40% reduction in NOx emissions would do. (The reason I use "apparently" is that they sometimes use "summer" to say their application period, but do not specify exactly what summer entails: they should make this more clear.) As part of this study, they evaluate the model using both aircraft and ground-based, routine monitors. They find that reactive nitro-

gen and ozone have both decreased in the SE, and further decreases are expected in response to a 40% NOx reduction.

This study is both of interest to the community and, for the most part, well executed, though there are aspects that need to be corrected before it should be accepted for publication in ACP. The strength is the focus on the oxidized nitrogen species and associated chemistry. The weaknesses include an inadequate evaluation for the analysis conducted, a short application period (2 months), a potentially poor choice of years, lack of consideration of condensed phase species in their assessment and evaluation.

Evaluation of the model is particularly important in such applications where one is trying to explain the reasons behind the observed (both in the model as well as in the ambient) changes, and, further, when using the model to extrapolate to further changes. Currently, the article relies on presenting plots with no quantitative statistical analysis. This needs to be corrected for further consideration of the article. Such an evaluation should be summarized in the main article with details in the supplemental. Looking at Figure 7, one sees rather considerable differences. How does this relate to other studies? If one is to assess how well the model may be relied upon to provide details of why the model may be capturing observed changes, and to what degree one can rely on the model to simulate future air quality, a more rigorous evaluation is required. One can look at the recent work done at EPA (e.g., [Simon et al., 2012]), or as part of AQMEII (e.g., [Appel et al., 2012; Dennis et al., 2010] [Solazzo and Galmarini, 2015]) or Environ [Emery et al., 2017] to provide the types of metrics that should be considered. Along those lines, there are ways to adjust deposition results to account for differences in precipitation rates other than the way they have chosen, and those should be considered. They should use total deposition fields from their modeling with total deposition fields estimated by NADP (http://nadp.sws.uiuc.edu/committees/tdep/tdepmaps/).

I was a bit surprised that they focus on just two months (July and August) for their analysis (and that this was not more clearly stated, if that is, indeed, the case). This, along with focusing on just one historical and one semi-current year, makes the results

very sensitive to the choice of time period. Along those lines, the summer of 2013 was cold and wet in the Southeast, and the meteorological adjustment determined by EPA was relatively large (in the Southeast̆AĬdata available at https://www.epa.gov/air-trends/trends-ozone-adjusted-weather-conditions). This was also the case for 2004, but the concern here is the timing as the adjustments are for the season, while the modeling conducted is just two months. More analysis is needed to tell how much impact is just from the meteorology of these two years specific to the two months.

Such an analysis, particularly when considering reactive nitrogen species, should provide additional focus on aerosol nitrate, including in the regional model evaluation. When they use the term "reactive nitrogen" are they including ammonia and ammonium? If not, they should add "oxidized".

There is a logical mismatch in the current paper. They state that there is a linear relationship between ozone and NOx emissions (line 627). This indicates a constant OPE. However, they also state that there is a transition from low to high OPE (line 633), though, admittedly, they do not say that after transitioning to a high OPE, it does not become constant. However, the discussion of OPE suffers from their not actually calculating an OPE. I might suggest removing much, if not all, of the OPE discussion unless they can bolster it further. If they do not remove this section, line 639: stating that OPE has increased very little and had little impact on net ozone production needs more definitive evidence.

I might suggest they integrate some of their findings with those in Blanchard et al., "ACP (2016) "Effects of emission reductions on organic aerosol in the southeastern United States". While this article is focused on organic aerosol, it relates to NOx controls in the SE.

Line 66: EPA still targets VOC emissions. (Look at the reductions in mobile VOCs over the period of interest!). Over the 2004 to 2013 period, how much of the ozone reduction is due to NOx vs. VOC controls? Do mobile emission reductions have a big impact in

the rural areas under investigation here?

There should be more discussion about the potential reasons for model bias following the work by Travis et al., (2016), and how this paper fits into that discussion.

Abstract: The final sentence states that 'further reductions of NOx emissions will lead to...less frequent extreme ozone events', however, the paper does not address extreme ozone events, just averages. This should be removed. Some reorganization of the paper could help improve its interpretation. A few suggestions: 1. The operational evaluation of the model and discussion of trends over time overlap (e.g., lines 343-363 and 488-499 discuss changes over time). I recommend splitting the evaluation section into 'operational' and 'dynamic' subsections (see Dennis et al. 2010 for an example). The dynamic evaluation section can address observed/modeled changes as related to emissions reductions, but the bulk of the discussion on this point should be reserved for its own section (currently section 5). 2. Define metrics used for comparison. 'Bias' is used here in both absolute (e.g., line 352) and relative (e.g., line 401) In the paragraph from lines 488-499, for example, the authors combine discussion of operational and dynamic evaluation, observed changes in response to emissions, and comparisons with previous modeling efforts.

Lines 567-575: why does the response of NOy concentration change from linear (from 2004-2013) to nonlinear with further emissions reductions?

Change all mentions of 'future' 40% reduction in NOx emissions to 'hypothetical' reduction (e.g., line 661). This analysis was performed partly to investigate the hypothesis that NOx emissions are overestimated, and there's no proof that the future will bring continued reductions. Also, I believe this model run was performed with 2013 meteorology, but this should be made clear.

In the discussion or Data sections, add some mention of reliability/consistency of measurements as a basis for model evaluation across the decade

Line 715: Change upto to 'up to'

Appel, K. W., S. Roselle, G. Pouliot, B. Eder, T. Pierce, R. Mathur, K. Schere, S. Galmarini, and S. T. Rao (2012), Performance Summary of the 2006 Community Multiscale Air Quality (CMAQ) Simulation for the AQMEII Project: North American Application, in Air Pollution Modeling and Its Application XXI, edited by D. G. Steyn and S. T. Castelli, pp. 505-511, doi:10.1007/978-94-007-1359-8_84. Dennis, R., et al. (2010), A framework for evaluating regional-scale numerical photochemical modeling systems, Environ. Fluid Mech., 10(4), 471-489, doi:10.1007/s10652-009-9163-2. Emery, C., Z. Liu, A. G. Russell, M. T. Odman, G. Yarwood, and N. Kumar (2017), Recommendations on statistics and benchmarks to assess photochemical model performance, J. Air Waste Manage. Assoc., 67(5), 582-598, doi:10.1080/10962247.2016.1265027. Simon, H., K. R. Baker, and S. Phillips (2012), Compilation and interpretation of photochemical model performance statistics published between 2006 and 2012, Atmos. Environ., 61, 124-139, doi:10.1016/j.atmosenv.2012.07.012. Solazzo, E., and S. Galmarini (2015), Comparing apples with apples: Using spatially distributed time series of monitoring data for model evaluation, Atmos. Environ., 112, 234-245, doi:10.1016/j.atmosenv.2015.04.037.