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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Southeast Atmosphere Studies: learning from model-observation syntheses" *by* Jingqiu Mao et al.

Anonymous Referee #1

Received and published: 11 March 2017

This manuscript provides a thorough review of the current knowledge of the atmospheric chemistry in the Southeastern US and the recommendations for future modeling work. My suggestions are mainly associated to the gas-phase chemistry and the modeling approach for gas and particles, which need large improvements in terms of writing. Explanations are needed and results should be clearly presented with proper context (see the specific comments). Although modeling papers are cited, the results are only briefly mentioned. It is still unclear to me after several readings what the current models predict for SE US for gas-phase species and for aerosols, and how do the models compare to the observations. More importantly, what approaches have been adapted in the models and which parameter may lead the largest uncertainties which can be improved based on SAS findings. This should be improved in the revised version. There are also noticeable shifts in writing style between sections possibly be-

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cause of the different contributions from the coauthors, which could be improved in the revised version to avoid distracting. I recommend the manuscript be published after a major revision.

Specific comments:

(1) Line 99-100: Please clarify if the interference lead greater or lower concentrations of HOx and which instruments\what study may be affected by this interference.

(2) Line 113-114: Do “Different treatments of this reaction” mean different reaction rates, products, or mechanisms? And quantitatively, how different are the ozone budgets predicted by different models?

(3) Line 133: The conclusion that “isoprene+NO3 reaction is both a major pathway for isoprene removal and for NOx removal” may be right for nighttime. But during the day, photochemistry dominates. Please clarify.

(4) Line 145-146: “many instruments” basically mean GC and PTR-MS (the two commonly used methods for quantifying those products), right? Please be precise.

(5) Line 149-156: It is not obvious for readers about the relationship between the HOx regime and the NOx concentrations. What are the NOx concentrations in the past and now? Why were the past experiments dominated by NOx chemistry but not now? Overall, I think this paragraph fits better with the background section not the findings.

(6) Line 157-166: These two paragraphs seem just restating what was presented in the introduction and background, which do not provide much information about the findings.

(7) Line 168-170: If I understand correctly, SAS has many sites and the HOx data reported by Feiner et al. (2016) are from just one site. For the statement here, the authors need to prove such HOx measurements can represent the whole case of southeast US.

(8) Line 174: “There are many observations that are central to improved understanding

of the detailed mechanisms. . .” What are the results/key findings then? Please give a summary.

(9) Line 175-176: Please specify what “many of the instruments used in this experiment and in many prior ones” stands for. References are needed.

(10) Line 183-186: Please explain how the lifetime of organic nitrates could affect “the lifetime of NOx, the spatial pattern of transported Nox, and the oxidation rates by OH, O3, and NO3”.

(11) Line 189-190: I am curious about the satellite findings of the ratio of glyoxal to HCHO. Does it different or similar to 2% which was observed in SAS? Please clarify.

(12) Line 191-203: Paragraphs (5) and (7) are repeated statements in Section 3 – Organic aerosol and do not show much about gas-phase chemistry at least from what was written right now.

(13) Line 208-209: I think that the statements of “no evidence from these studies” (it was one study) and “at any NOx concentration sampled in the rural Southeast” are overstated. What about urban plumes and different seasons? Please clarify.

(14) Line 220-224: There is lack of explanation about “The largest NOx and BVOC emissions are not collocated.” Is this also a key finding from SAS? If so, please provide more information and references for it. Besides, I don’t think readers can understand the following sentences: “Resolution is especially important for the 15% or so at the tails of the NOx and HCHO distribution – less so for O3. . .resolve this last 15% which probably requires a horizontal resolution of order 12 km or less”. Please clarify.

(15) Line 229-230: The statement of “these errors are approximately linear” needs explanation.

(16) Line 237-238: Is this a common finding for Southeast US?

(17) Line 270-292: Models have difficulties to reproduce the mass loading of OA but the

problems mainly happen in urban area and aloft. It would be very helpful to summarize the model results regarding Southeast US here.

(18) Line 298: What are “a diversity of modeling approaches”?

(19) Line 313-315: This sentence is confusing. Does “Their structure” mean the structure indicated by the instrument? So “Their structure” is not the actual structures of the original molecules because of the thermal decomposition?

(20) Line 319-322: The authors need to tell readers what the relative humidity and degree of oxygenation of organic compounds in the Southeast US and the conditions leading a phase separation are? The phase behavior also depends on the OA type (precursor).

(21) Line 333-334: “directly equal” is not the right word. Although HOA has been widely used as a surrogate for POA, it has involved some degree of oxidation (if you look at the O:C ratio of HOA in various studies). AMS-PMF analysis also identified factors like CCOA (coal combustion OA), which is in model supposed to be part of POA but it involves some degree of oxidation. Both the semivolatile feature of “POA” and the aging process complicate the model tracers. I would suggest not to link the model tracers to PMF factors. Instead, we need to make cautions that the model OA tracers may not physically match their names for historical reasons. When making comparisons to AMS PMF factors, efforts are needed for understanding the attributions.

(22) Line 354-356: This also argues the statement in Line 133 (also see my comment #3).

(23) Line 373-374: What about the “old” isoprene SOA in the models? Does it overlap somewhat with the IEPOX-SOA? How should the models do?

(24) Line 383-385: The phase also regulates the particle-phase reactions that produce IEPOX-SOA (Kuwata et al., 2015). It is worth to add that.

(25) Line 422: What kind of measurements? Please clarify.

(26) Line 507-513: Since there are different versions of BEIS and MEGAN, there should be first a comparison among versions. So we know for example, in Line 510, which is compared to which.

(27) Line 811-812: This statement is ambiguous. From my reading of section 3.2.6, models cannot neglect SOA from urban emissions for SE US". The parameterization based on CO is an option. But it is unclear at least in section 3.2.6 whether the parameterization based on CO works well for SE US.

Technical remarks: Line 72-74: According to ACP's guidelines, works "submitted to", "in preparation", "in review" should be included in the reference list. Line 74: Warneke et al. is already published. Please cite the AMT version in the reference list. Line 86-87: Add "e.g." in the parentheses. Line 106: Remove the redundant "Peeters et al.,". Line 131: Remove "," after "2012)". Line 137: Since this is a review article and most of the presented results are published ones, I suggest to remove "preliminary" or to rewrite the subtitles. Line 147-148: The last sentence seems being misplaced. Line 154: I could guess that "These experiments" stand for SAS studies. But there is lack of context. Line 217: Missing a word between "there" and "be". Line 288: Remove the "," before "and glyoxal". Line 323-324: This is already stated in Line 291 and since it is about cloud processing which doesn't match directly with the partitioning and phase problems. I would suggest to remove this statement here. Line 322: Pye et al. is already published. Please cite the right one. Line 333-334: Add "the" before "model POA" and "the" before "AMS". Line 461: Should "highest" be "high"? Line 473: "." is missing before "Past". Line 491: 0.25×0.3125 or 0.25×0.25? Line 513, 522, 548: According to ACP's guidelines, works "submitted to", "in preparation", "in review" should be included in the reference list. Line 610: "Wm-2" should be "W m-2" Line 623: "a-1" should be "a-1"

Reference: Kuwata, M., Liu, Y., McKinney, K. A., and Martin, S. T.: Physical state and acidity of inorganic sulfate can regulate the production of secondary organic material from isoprene photooxidation products, Phys. Chem. Chem. Phys., 17, 5670-5678,

10.1039/C4CP04942J, 2015.

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