Author response to reviews

The authors appreciate ACP Editorial Office, Dr. Müller, and both referees' continued efforts. These elaborated comments helped us further improve this manuscript significantly. Following Dr. Müller's guidance, we paid particular attention to the feedback in Report #2 when revising the manuscript. Please see below our point-by-point response (in blue) to all comments by the ACP Editorial Office and both referees (in black). Quoted text from the revised manuscript is *in italic*. A "tracked-changed" version of the manuscript is submitted together with this document.

Response to ACP Editorial Office comments

I noticed that your table 3 contains coloured cells. Please note that this will not be possible in the final revised version of the paper due to HTML conversion of the paper. When revising the final version, you can use footnotes or italic/bold font.

In Table 3, degradations and improvements are now highlighted in italic and in bold, respectively. Regarding the paper format, the titles and footnotes of tables have been carefully edited, and the unit of temperature has been changed from "^oK" to "K" in the text, Table 2, as well as several figures in the manuscript and the SI.

Response to Report #1 by Referee #2

The authors have addressed the reviewer comments reasonably well, revising the text, adding a useful additional simulation and refining their explanations. The resulting manuscript is an improvement over the original, and a better account of their results. The perspective remains rather narrow, focusing more on documenting results than on furthering understanding. This seems like a missed opportunity, but the paper nevertheless presents some useful results that are worthy of publication.

Thank you for recognizing the improvements in this manuscript. As this referee pointed out in the earlier review report, the narrow perspective is partially due to the choice of the Noah land surface model. This will be better addressed by presenting results based on a different land surface model in a follow-up study. To improve our understanding of how SM DA affected ozone at process level based on the widely-used Noah model, especially at/near the surface, additional analysis has been conducted, the results sections in the manuscript and the SI have been reorganized, and the abstract has been rewritten. Please refer to Figure R1 in this document for a clear illustration of SM DA impacts on modeled ozone and its performance at process level which are discussed in the revised paper. Please see our responses to your next two comments for the changes made to the abstract and the reorganizations of results.

The abstract describes what was done, but still doesn't summarize the results of the study in a meaningful way beyond stating that SMDA impacts the model results. Not only is there no quantification, the statements are barely even qualitative. The only conclusive statement is the new comment that "SMDA improved the model treatment of convective transport" (although the "and/or" that follows suggests that even that is uncertain). The abstract should state clearly what the study found.

It would be helpful to have an overarching conclusion that the reader can take away (that SMDA doesn't make much difference given other biases?) The conclusions quickly get into technical detail and explanation, and the reader loses the big picture.

Please note that unlike aircraft CO measurements which were used to assess the model treatment of transport during both the ACT-America and SEAC⁴RS campaigns, aircraft NO_x measurements which helped evaluate WRF-Chem lightning NO_x at higher altitudes were available only during the SEAC⁴RS campaign. This is a reason for using "and/or" in that sentence.

The abstract has been rewritten, including quantitatively descriptions of key ozone-related findings and other highlights. Following this referee's suggestion, we use the following sentence to deliver an overarching conclusion of this study, and to emphasize the strong needs of continuing such studies with improved model parameterizations (e.g., deposition) which will be covered in a follow-up study: "In the cases that the DA improved the modeled SM, weather fields and some O₃related processes, its influences on the model's O₃ performance at various altitudes are not always as desirable, due to the uncertainty in the model's key chemical inputs (e.g., anthropogenic emissions), as well as the shortcomings in model parameterizations (e.g., chemical mechanism, natural emission, photolysis and deposition schemes) and the model representation of stratosphere-troposphere exchanges".

Many of the changes have involved adding additional material to the supplement. While this makes the record of the results more complete, it doesn't help the reader focus on the key aspects that matter.

Some of the previous SI contents (i.e., previous Figures S3-S4) have been moved into the main manuscript, and additional contents (i.e., current Section S2, Figures S4 and S6, two new panels in current Figure S5, and a new column in Table 2) have been added to support the extended discussions.

Line 99: The term "semicoupled" is still not adequately defined. Remove "here is similar to weakly-coupled as opposed to fully or strongly coupled, which" as this does not help the reader at all. The remainder of the sentence at least states what is coupled.

The cited text has been removed. This paragraph introduces the paper organization, whereas the following "methods" section 2.1 includes most of the technical details, e.g., regarding the coupling, please see the paragraph starting with "*All WRF-Chem cases, except case "minus001*…".

Response to Report #2 by Referee #1

I have two main issues: First, the revised MS does not address my comments substantively. The revised MS shows only a few significant changes. I do not consider that the structural problems in the MS raised in the review have been addressed - the focus is on model response but there is not enough time spent quantifying at the process level the impact of DA. Second, there's been no restructuring to tighten up the focus to the subject mentioned in the title, and so this remains a paper where the apparent subject of the paper is lost in the details of comparison between model and observations and the simple sensitivity studies that would give a much better process-level understanding of the impacts of DA of SM on model performance are not there.

We appreciate the elaborated comments and suggested changes by this referee. To improve our understanding of SM DA impacts on the modeled ozone at process level, especially at/near the surface, additional analysis has been conducted, results sections in the manuscript and the SI have been reorganized (please refer to our responses to your following comments for details), and the

abstract has been rewritten. Figure R1 below clearly illustrates the SM DA impacts on modeled ozone and its performance at process level which are discussed in the revised paper.



Figure R1. Illustration of how SM and its performance changes due to the DA affect the modeled surface weather fields (e.g., temperature), individual ozone-related processes (e.g., biogenic emissions and dry deposition), and ozone. Some of the areas in the western/southern Missouri and Ohio (highlighted in green) are examples where the model responses were strong, and in these areas, the SM DA improved SM and surface weather fields but not necessarily all individual ozone-related processes and the modeled ozone performance (reasons in red text). In this work, temperature response to the DA is the main reason for the changes in biogenic emissions, reaction rates, and deposition which are critical to ozone, and more elaborations are provided in brown text.

In lots of places the discussion in the manuscript remains rather qualitative. The revised MS spends too much time discussing factors not included in the study (e.g. L532-538, discussion of possible modifications/ missing processes in O3 deposition), and too little time is spent on discussion of the model itself (e.g. L529- L530 which is all that's said on the results regarding O3 deposition). I'd suggest removing as much as possible of this speculation about model inadequacies that does not add to the interpretation of model response, and refocusing the MS by adding a separate detailed section on what processes are susceptible to modification by SM DA and on quantifying the change in these at the process level (that is, for instance, at the emissions level rather than on the impact on the very small change in ozone levels).

An additional section (S2) has been added to the SI to introduce the Wesely dry deposition scheme, and dry deposition related results are presented in more detail (i.e., including individual terms of the surface resistance in Figure S6, and how they may be altered by environmental conditions). Section 3 has been reconstructed to logically explain in detail on how individual ozone-related processes (e.g., biogenic VOC and NO emissions, dry deposition velocities, chemical reactions) responded to the weather fields and affected ozone, with additional model fields (e.g., radiation

fluxes, peroxyacetyl nitrate concentrations) examined to support the extended discussions. Please also refer to Figure R1 for a clear illustration of this flow.

A MS that had a section on 'model responses to SM DA' moving figures S3 and S4 into the main text and then a section on 'comparision with observations' would allow the reader to assess better how the impact of SM DA propagates through to model skill in simulating ozone. Then add significant extra text to 'model responses to SM DA' section in the MS discussing these S3 and S4 figures and similar process level responses of model inputs or other parameterisations before considering the impact on O3.

As suggested, Figures S3 and S4 have been moved into the main text. The comparison of modeled and measured "surface" SM is now shown in current Figure 2c;f;g, which, together with the previous Figure S3 (current Figure 2b;e), helps us better understand how the modeled SM performance was changed by the DA. The previous Figure S4 (current Figure 4), as well as the added column in Table 2, supports the extended discussions on "model responses to SM DA".

As mentioned above, we now follow the new flow in Figure R1 to discuss how "the impact of SM DA propagates through to model skill in simulating ozone". In the results sections of the revised manuscript, we discuss the "model responses to SM DA", the driver(s) of some responses, as well as observation-model comparisons along with discussions on how errors propagated. For surface conditions, an emphasis is given to regions like Missouri and Ohio (highlighted in green in Figure R1) where the model responses are strong and statistically significant (e.g., for temperature).

Consider L220 of the revision where extra text has been added. The revised discussion is not based around how the biogenic emissions varied with the assimilated SM conditions in this study, or even quantifies the difference. In L515 finally the authors state that the MEGAN emissions have no dependency on SM, but again 'do not anticipate' that emissions were changed. This is not a sufficient response to the review. Please insert a quantitative discussion as to how SM DA affected, and why, the biogenic VOC and NO emissions in your study.

We extended the text near previous L220 and L515. It is necessary to clarify that the text near previous L220 is within the "methods" section and therefore it only introduces MEGAN and prior knowledge of it. It is not intended for showing results from this study. The text near previous L515 is within the "results and discussions" section where results from this study are presented and discussed. Please note that the regions where we "do not anticipate that biogenic VOC emissions would be changed significantly by the SMAP DA even if their dependency on SM was realistically included in MEGAN", are some "Atlantic states that were in the early-middle phases of drought in August 2016" and for this particular case SM and temperature only changed mildly in response to the SMAP DA. The strongest SMAP DA impacts over high biogenic emission regions were not experiencing droughts during the study period, and we concluded that the presented emission changes are largely due to temperature changes after examining the modeled radiation fields (see current Figure S4). These have been made clearer in the revision.

Consider, as a second, but not final example, dry deposition, L240/L535 again does not make clear how the deposition velocity is affected by SM DA - is it solely via modification to surface temperature? Again, it would help to use the Weseley scheme to estimate the response of the deposition velocity to the temperature. This would help to understand the S4 figure panel on deposition. At present, deposition has only been addressed with extra text stating qualitatively how

things might be (pg L221, L516, L535), but the calculations/diganostics are not there. Again, what drives the change? Again, please add a quantitative discussion as to how SM DA affected deposition. Similar comments pertain to my other major points where I asked for quantitative discussion. In a study such as this where you are trying to unpick how SM DA affects O3, these process-level attributions are essential to the success of your study.

An additional section (S2) has been added to the SI to introduce the Wesely dry deposition scheme, and dry deposition related results are presented in more detail (i.e., including individual terms of the surface resistance in Figure S6, and how they may be altered by environmental conditions). The modeled radiation fields have also been examined (see current Figure S4), to support the conclusion that radiation contributed less importantly to the changes in modeled O₃ dry deposition velocity. Also, note that cuticular resistance is treated differently for wet and dry surfaces (flagged based on the modeled humidity and precipitation). Therefore, the DA-induced humidity and precipitation changes can have impacts on the modeled cuticular resistance and deposition velocity was minor in this study. The paragraphs starting from previous L240 and L535 have both been rewritten to more clearly describe what drove the changes in WRF-Chem dry deposition and its individual terms.

Additional analysis and discussions have been added concerning chemical reaction rates. For example, please see added panels in current Figures 9 and S5 concerning peroxyacetyl nitrate as well as discussions on its relationships with temperature and ozone in Section 3.3.1.