Response to Comments by Reviewer 1

General Comments

Comment: This manuscript summarizes experiments that compare dry deposition schemes available in the CMAQ model. The conclusion that careful diagnostic grid-scale and land use specific dry deposition variables should be adopted especially for model intercomparison purposes is an important message of this work. In general, I think this manuscript presents a reasonable analysis with many valuable details that will be of interest to the relevant community.

Response: We would like to thank the reviewer for the overall positive assessment of our manuscript and the constructive suggestion to consider revising Section 3.1. Our responses to reviewer comments are shown below in italics while changes incorporated into the revised manuscript are shown in bold font.

Comment: My major comment is really related to what I felt was a lack of focus in the earlier parts of the manuscript: There is a lot of text in this paper (throughout the Methods and Section 3.1) that spends time explaining differences in their simulations with the output from model experiments in Appel et al. (2021). I was a bit confused by this. The main purpose of this manuscript is to compare the M3Dry and STAGE dry deposition schemes in an internal CMAQ implementation from 2016. To give this manuscript a more targeted focus, I would have suggested just to evaluate their model implementation directly without commenting so much on the indirect evaluation through the Appel et al. (2021) implementation. Likewise, I was a bit confused why there are so many tables and figures focused on evaluating 2010 model output, when none of this output is relevant to the dry deposition scheme discussion that uses only the 2016 model experiment (from Sections 3.2 onward). A great deal of unnecessary (and seemingly unrelated) text could be avoided if the focus was simply on the 2016 model experiments to begin with. This focus would also help remove many tables and figures (e.g., avoiding at least Table 2b, 3b, and many panels throughout Figure 1a-2b) to tighten the content.

To illustrate my point, one of the sentences early in the conclusion section mentions that lateral boundary conditions are a driver of the main differences between the present simulations and those from Appel et al. (2021). Why is this relevant to the internal comparison of CMAQ dry deposition schemes? Following this one (and only) mention in the conclusion, the remaining two and half pages of summary discuss, appropriately, only on the 2016 dry deposition scheme comparison. I highly recommend a substantial revision of the earlier sections of this manuscript so that it focuses on the relevant 2016 simulations only, and perhaps deemphasizes all the comparisons with Appel et al. (2021). In my opinion, all this extra material detracted from the motivation of the paper. Otherwise, perhaps I missed something important, and the authors could provide better motivation for why all this analysis was included.

I really don't have many other substantial comments. Sections 3.2 and 3.3 contain a great deal of valuable information about dry deposition modeling in CMAQ, with implications for other models as intercomparison projects, that I believe could be of interest to many readers of ACP.

Response: After considering the reviewer's comments, we agree that a restructuring and partial reduction of the material presented in Section 3.1 would help with the flow of the manuscript. We have therefore shortened Section 3.1 by removing any model performance results of the Appel et al. (2021) CMAQ simulations from the tables and figures in this section and also from any associated discussion.

Furthermore, we have also eliminated the RMSE time series previously shown in Figures 2a and 2b since they contained similar information to what was already shown in Figure 1 and the tables. These changes have hopefully made Section 3.1 more focused and easier to read.

While we agree that discussing the performance of the CMAQ simulations performed for this study in the context of the comprehensive Appel et al. (2021) CMAQv5.3.1 evaluation study is not directly related to the diagnostic comparison of the CMAQ M3Dry and STAGE AQMEII4 simulations, we believe that doing so is still desirable within the context of the overall AQMEII4 activity to document how different model configuration choices and input files affect the CMAQ results contributed to this activity. Forthcoming AQMEII4 analyses targeted for the same ACP special issue for which this manuscript is intended will perform model evaluation across all participating models and will benefit from having access to a more detailed analysis of the CMAQ M3Dry and STAGE model results than will be possible to perform in those multi-model studies. To balance this motivation for keeping these comparisons with the justified concern raised by the reviewer that doing so as part of Section 3.1 would result in a less focused manuscript, we decided to move all of the figures, tables, and discussions related to comparing the AQMEII4 CMAQ simulations to the Appel et al. (2021) CMAQ simulations, to the supplemental material. The following text has been added to the beginning of Section 3.1:

"Comparisons of modeled and observed MDA8 O_3, SO_2, NO_x, $PM_{2.5}$, SO_4^{2-}, NO_3^-, OC, and EC concentrations at AQS monitors, MDA8 O_3, SO_2, NO_x, and $PM_{2.5}$ at NAPS monitors, and precipitation and wet deposition of SO_4^{2-}, NO_3^-, and NH_4 at NADP NTN monitors are presented in Figure 1 and Tables 2 – 3. This section summarizes the performance of the M3DRY_2016, STAGE_2016, M3DRY_2010, and STAGE_2010 base case simulations. To provide context for these results, a comparison to the model performance of 2016 CMAQv5.3.1 simulations from a recent comprehensive evaluation study (Appel et al., 2021) and the differences in model configurations driving differences in model performance can be found in the supplemental material (Figures S1 – S7 and Tables S2 – S3)."

Regarding the reviewer's concern that model evaluation results in Section 3.1 are provided for both 2010 and 2016 while the diagnostic analysis in Sections 3.2 and 3.3 focus on 2016, we again note that having CMAQ performance documented for all simulations contributed to the AQMEII4 activity and analyzed in forthcoming multi-model analysis manuscripts will be beneficial for such manuscripts. We have therefore retained the 2010 model evaluation results in Section 3.1, but have also added the following text to the beginning of Sections 3.1 and 3.2, respectively, to provide more clarity on the different choice of years in both sections.

"Even though the diagnostic analyses presented in subsequent sections of this manuscript are focused on 2016, this section documents model performance results for both 2010 and 2016 because results from the 2010 simulations will be included in forthcoming AQMEII4 analyses."

"To avoid repetition, all analyses in these sections focus on the CMAQ AQMEII4 simulations performed for 2016 because the differences between the M3Dry and STAGE CMAQ simulations for 2010 were very similar to those for 2016 and because the sensitivity simulation quantifying the impacts of using a different LU classification scheme was performed for 2016."

<u>Minor comments:</u>

Comment: Line 171: First introduction of "VEGF" – please define.

Response: Thank you for catching this omission, the definition has been added in the revised manuscript.

Comment: Line 494-496: "…treats cells with more than 10% water as either all land or all water depending on whether the fractional water coverage is below or above 50%". I had trouble following this sentence. Could it be reworded or broken up to help with its clarity?

Response: The relevant section has been reworded as follows in the revised manuscript, hopefully providing greater clarity:

"For grid cells with water fractions between 10% and 50%, the water fraction is reset to zero and the non-water categories are renormalized to 100%. For grid cells with water fractions exceeding 50%, the water fraction is reset to 100% and the fractions for non-water categories are set to zero. No renormalization is performed for grid cells with water fraction coverage below 10%."

Response to Comments by Reviewer 2

General Comments

Comment: The manuscript is a very detailed investigation of dry deposition within the CMAQ model, which places the runs for the AQMEII4 exercise in context with other configurations and inputs that have been used recently or are more typically used for CMAQ. The sensitivity studies provide a great deal of new or updated information about the impact on air quality concentrations and fluxes of land use definitions and treatment, the underlying details of the resistance parametrization, and inputs such as meteorology and boundary conditions. The results will be primarily of interest to atmospheric modelers, but also are a significant contribution to the larger project that is of broad interest to the air quality and deposition impacts communities. Given the length and level of granularity, the paper needs to be carefully organized, and the authors have done a good job of this. I recommend publication with very minor revisions.

Response: We would like to thank the reviewer for the overall positive assessment of our manuscript and the careful review and helpful suggestions which have led to an improvement in some of the figures. Our responses to reviewer comments are shown below in italics while changes incorporated into the revised manuscript are shown in bold font.

Specific comments:

Comment: Section 2.2: the description of the set of model runs is missing the M3DRY_APPEL_EMIS_2016 one

Response: Thank you for catching this omission. The relevant sentence has been updated in the revised manuscript as follows:

"M3DRY_HCMAQ_2016 can be used to assess the impact of using chemical boundary conditions from CAMS compared to using boundary conditions from H-CMAQ as in Appel et al. (2021) **while M3DRY_APPEL_EMIS_2016 can be used to quantify the impacts of the different anthropogenic and wildland fire emissions used in this study vs. Appel et al. (2021).**"

Comment: Section 2.4: There's significant discussion in the paper about results in southern Canada; what was the reason for not using data from Canadian sites (or Mexican, if available)?

Response: Thank you for raising this point. In the revised manuscript, we have added observations of O_3, SO_2, NO_x, and $PM_{2.5}$ from the Canadian National Air Pollution Surveillance (NAPS) program to the tables in Section 3.1 and the model bias maps in the supplemental material. Evaluation results at NAPS monitors are generally comparable to those at the AQS monitors with the exception of SO_2 for which the simulations are biased low at AQS sites and high at NAPS sites. We do not have access to air quality observations at sites in Mexico.

Comment: Fig. 1: it's a bit difficult to see the observations distinctly. Can you make them dashed, or points, to bring that line out a bit better?

Response: Thank you for this suggestion. We have changed the line depicting observations from a solid to a dash pattern which indeed makes the observations easier to distinguish from the model results.

Furthermore, we have also removed the lines depicting the Appel et al. (2021) simulations from the figures shown in the main manuscript, and the new figures S2 and S3 in the supplement that include the Appel et al. (2021) simulations are for 2016 only. Both of these changes also helped to better distinguish observations from model simulations in these figures.

Comment: Fig. 6: maybe choose a more contrasting color for the WRF lines

Response: Thank you for this suggestion, we have changed the color of the WRF lines to orange in the revised manuscript.

Comment: Fig. 8: are these plots linear scale? I can't tell with only 2 values.

Response: Yes, the y-axis scale is linear. In the revised manuscript, we have updated this figure with additional y-axis tick labels to make this clear.

Comment: Fig. 9: I suggest you swap the M3DRY and STAGE order on the plots to better correspond with Fig. 7

Response: The M3Dry and STAGE order was actually specified consistently in the plotting code for both figures, but for horizontal bar plots such as the one in Figure 9, the plotting software arranges bars from bottom to top rather than using a top to bottom order that for many readers might be more consistent with the left-to-right ordering in Figure 7. For the revised manuscript, we have modified the plotting code to now use top to bottom ordering for Figure 9, and have also made the hatching angle and density consistent between both plots. In addition, we have also revised Figure 13 comparing the fractional land use coverage for M3DRY_2016 and M3DRY_NLCD40_2016 to use the same top-to-bottom ordering, now showing the M3DRY_2016 base case at the top of each pair of bars.

Comment: L. 695-6: I would include deciduous broadleaf in the list of LUCs where NLCD > MODIS flux, it's a significant difference (here and in the summary/abstract)

Response: Thank you for this good suggestion, deciduous broadleaf forest has been added to the list of landuse categories for which using NLCD40 results in a higher flux compared to using MODIS, both here and in the summary.

Comment: L. 732: pretty sure you mean NO2, not SO2, here

Response: No, we believe the statement is correct as written. Figure 7b shows that the M3Dry cuticular effective flux is larger than the STAGE cuticular effective for SO_2 while they are very similar for NO2. This statement in the summary is the same as on line 468 in Section 3.2.1 where Figure 7b is discussed.

Comment: Section 4: It would be helpful to provide some context as to how significant are these deposition velocity (or flux) differences between configurations compared to differences that are seen between CMAQ and other models with different schemes entirely. Presumably this will be discussed in the larger AQMEII4 exercise, but there are studies in the literature that could be cited to give the reader a sense of the relative magnitude.

Response: Thank you for this suggestion. We have added the following text passage to Section 4:

"Absolute differences in seasonal mean O3 Vd are on the order of 0.05 – 0.1 cm/s for many locations, While these differences tend to be smaller than the range of model differences reported in intercomparison studies performed at flux measurement sites (e.g. Wu et al., 2018 and Clifton et al., 2023) and for global models (Hardacre et al., 2015), their magnitude nevertheless represents a variation of about 10-30% of CMAQ simulated seasonal mean Vd with the highest relative differences generally occurring during winter."

"Initial analysis of results from all AQMEII4 grid model simulations show that the differences in simulated O3 Vd, deposition pathways, and deposition fluxes between the CMAQ M3Dry and STAGE simulations analyzed in this study tend to be smaller than the differences relative to other AQMEII4 grid models."

In addition, we also added the following discussion at the end of revised Section 3.1 to provide additional context:

"The results presented in the supplemental material show that the choice of the CMAQ dry deposition scheme (M3Dry vs. STAGE) has a smaller impact on aggregated model performance metrics than the sensitivity of CMAQ results to model input data sets and boundary conditions that represent the large-scale chemical environment. However, it is important to note that M3Dry and STAGE share many structural similarities (see Figures B2 and B3 in Galmarini et al., 2021) and that the similarity in model evaluation results therefore does not imply that uncertainties and potential errors in representing dry deposition are not a potentially important source of overall model error. An analysis of point model simulations at eight ozone flux measurement sites performed with all dry deposition schemes participating in AQMEII4 shows that differences in seasonal cycles of Vd and deposition pathways between M3Dry and STAGE are generally smaller than differences relative to other schemes. In addition, the following sections demonstrate that the choice of M3Dry vs. STAGE in CMAQ can have more pronounced impacts for specific seasons, regions, and deposition pathways than its impact on these domain-wide model performance results."

Comment: References: I expect you are aware that the Clifton "in prep" paper is now citable on ACPD

Response: Yes, this reference has been updated to refer to the ACPD manuscript.