General comments:

The manuscript "Evaluation of HNO3, SO2, and NH3 in the Surface Tiled Aerosol and Gaseous Exchange (STAGE) option in the Community Multiscale Air Quality Model version 5.3.2 against field-scale, in situ and satellite observations" has been submitted to the journal GMD. The research emphasizes the uncertainty in the deposition mechanism that developed based on the resistance framework for estimating NH3 flux, and the model improvement using additional micrometeorological fluxes. In general, the complex deposition component makes the approach vital for understanding model performance and, further, for improving it. However, the entire research contains two major issues. (1) The main objective seems to modify the commonly used resistance model based on the previous deposition approach. However, the reason for doing this is not clearly explained. In the Introduction, the author mentioned the emission and deposition are treated differently in the regional model. This is understandable because the sources of emissions, including anthropogenic and natural, differ. Moreover, the deposition process, which depends on land surface, particle size, and meteorological conditions, is highly variable and uncertain. The present manuscript primarily evaluates the NH3 flux by comparing micrometeorological fluxes with CMAQv5.3 tabular, which is not relevant to the literature review. Both emission flux and deposition flux need to be analyzed to address the uncertainty mentioned. Or else, simply emphasizing the limitation of CMAQ STAGE and the importance of using micrometeorological flux measurement for STAGE improvement. (2) The entire manuscript is not well written and requires proofreading. For instance, "…CMAQv5.3.2, table 2…Table 3…" in Line 304-306 is confusing.

Response: We would like to thank the reviewer for their detailed comments and they have been used as a basis for reorganizing the results and discussion section of the manuscript.

Response to issue (1):

As stated in the first introductory paragraph:

"The exchange of atmospheric aerosols and trace gases between the atmosphere and biosphere is an essential process in the source, transport and fate of atmospheric pollutants and represents an important vector of ecosystem and human health exposures (Eschelman and Sabo, 2016; Greaver et al., 2012; Burnett et al., 1998; Galloway et al., 2020)."

Yet there is still a considerable amount of model variability in modeling these processes (Galmarini et al., 2021) and there are inconsistencies in the description of dry deposition and bidirectional exchange processes over natural surfaces that should be governed by the same dynamics in prior versions of CMAQ, lines 63-69. The variability in the model deposition parameterizations are likely due to a paucity of observational data. Here we adapted a commonly used resistance model was for use at the field and regional scale to leverage the utility of the available observations. At the field scale, models aid in understanding dynamics of the air-surface exchange that cannot be directly measured, e.g. stomatal, soil, and cuticular contributions to the net flux. Here we apply the findings of a field scale evaluation to a regional scale simulation and utilize CrIS satellite observations to evaluate if the findings are generalizable.

Response to issue (2):

The manuscript has been revised to address many of the comments below. Specifically, the model simulations have been grouped into cases using default CMAQ v5.3.2 tabular and site specific observed input data. We feel that this has made the presentation of the results and discussion more clear. The following cases have been added to streamline the results, discussion, and Table 3.

- LM – STAGE model simulation of Lillington, NC fluxes using the default CMAQ v5.3.2 tabular inputs
- LO – STAGE model simulation of Lillington, NC fluxes using observed stomatal, dew, and soil NH3 emission potentials and minimum stomatal resistance.
- DFM – STAGE model simulations of the Duke Forest, NC fluxes using the default CMAQ v5.3.2 tabular inputs
- DFOS - STAGE model simulation of Lillington, NC fluxes using observed stomatal, dew, and soil NH3 emission potentials and minimum stomatal resistance.
- DFOL - STAGE model simulation of Lillington, NC fluxes using observed stomatal, dew, and leaf litter NH3 emission potentials and minimum stomatal resistance.

Specific comment:

Line 18: What is the importance of this methodology? Why is micrometeorological flux measurement significant for the model evaluation and improvement? What is the main limitation of STAGE? The research question and motivation remain unclear.

Response: The specification of micrometeorological flux measurements is important in this case as these techniques do not alter the environment being measured like chamber techniques and offer higher temporal resolution necessary to capture the dynamics of the processes than can be inferred from biological sampling or isotopic techniques.

Line 81-83: STAGE in CMAQv5.3 has already been publicly released. Is the present research proposing a new parameterization (e.g., new STAGE)?

Response: This research evaluates the STAGE model against fluxes measurements in an agricultural field for NH3 and a grassland site for NH3, HNO3, and SO2. This is the initial evaluation of the model at these sites and the initial evaluation of it at a grassland site for multiple pollutants. Furthermore, we found that updating the regional scale model NH3 emission potentials improved the evaluation of the regional scale model against satellite observations. We are not proposing a new STAGE model as the emission potentials are an input to both the field scale and regional scale versions of the model.

Line 237: What do you mean “would be returned”?

Response: This has been revised. The intent is that U/u^{*2} is numerically the same as Ra in Table 1 if U is estimated for the log linear wind profile in Pleim and Ran 2011. This sentence has been removed.

Line 291: Figure 1 or Figure 2?

Response: This is indeed referencing Figure 2. The text has been corrected.

Line 295: Higher error than which site? Please revise the sentence. How can low LAI and minimal stomatal resistance affect model performance?

Response: Section 3 was revised addressing the general comments. This specific area did need clarification and the text was revised as follows:

The evaluation of modeled H_2O and NH_3 fluxes at Duke Forest had higher error at than those at the Lillington site (Table 3). This is likely influenced by uncertainty introduced by the lower magnitude of the measured fluxes at the Duke Forest site and the lack of LAI and minimum stomatal resistance measurements which govern the cuticular and stomatal exchange processes respectively

Line 295-298: Please revise the whole sentence!

Response: We have revised this section to include model cases as discussed in the general comments.

Line 298: This seems to be a statistical error? Please use another statistical index that would exclude the effect of the outlier.

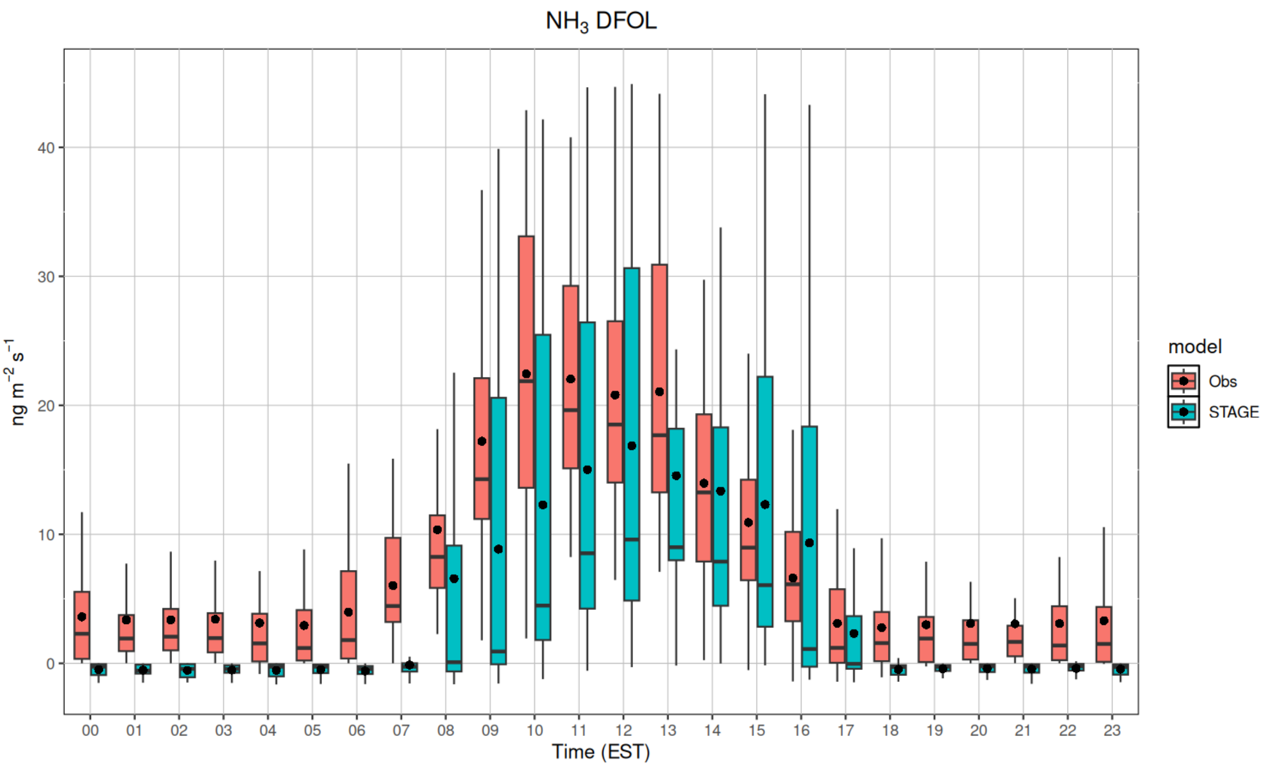
Response: This section has been rewritten.

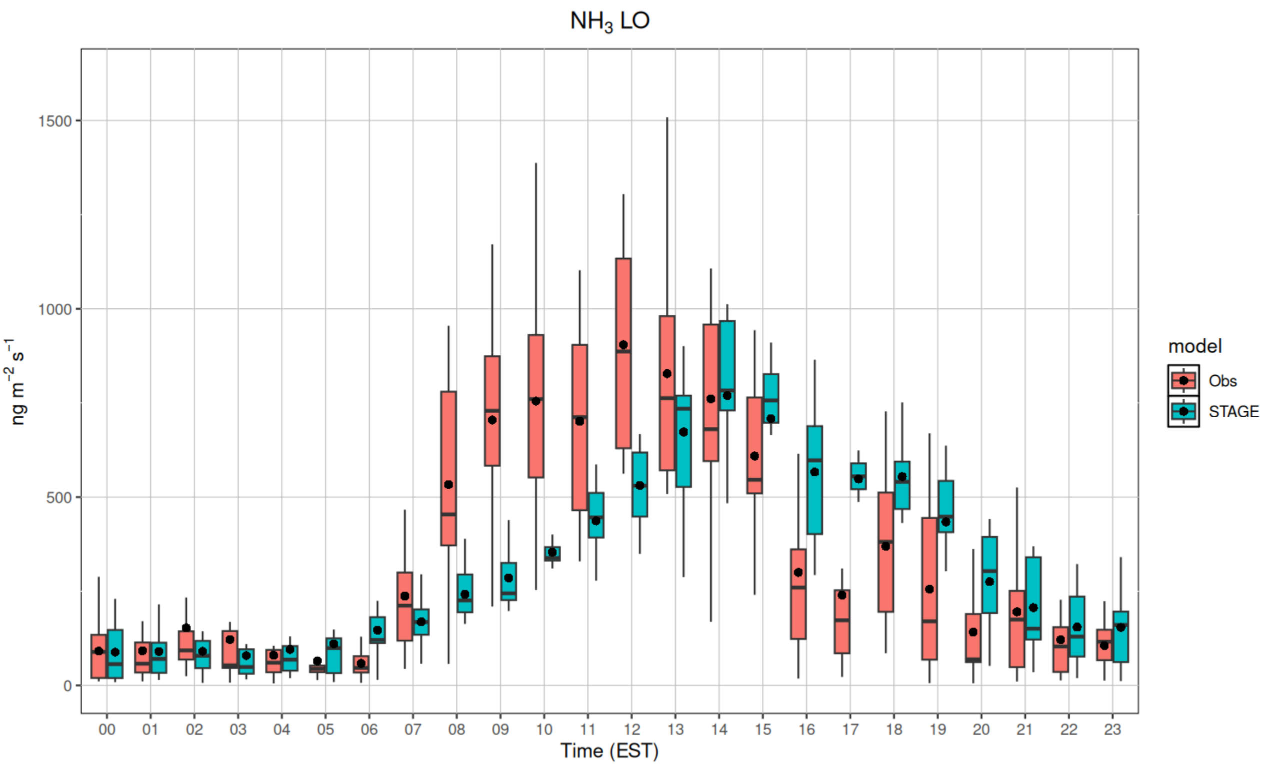
Line 307: How did you define "most sensitive"? Such a description is subjective.

Response: This is a fair criticism. This discussion has been rewritten to highlight that using the observed emission potentials at the Duke Forest site instead of the tabular values from CMAQ v5.3.2 resulted in capturing the direction of the mean flux and improving the model correlation.

Line 309: What do “NH_3 observed $\Gamma ll, \Gamma st \Gamma dew$” in Figure 3 stand for in general?

Response: This the model case where the STAGE simulation used the observed apoplast, leaf litter, and leaf dew observations. These scenarios are now organized as model cases in Table 3, and as addressed in the general comments. Figures 2b and 3 have been revised as follows:





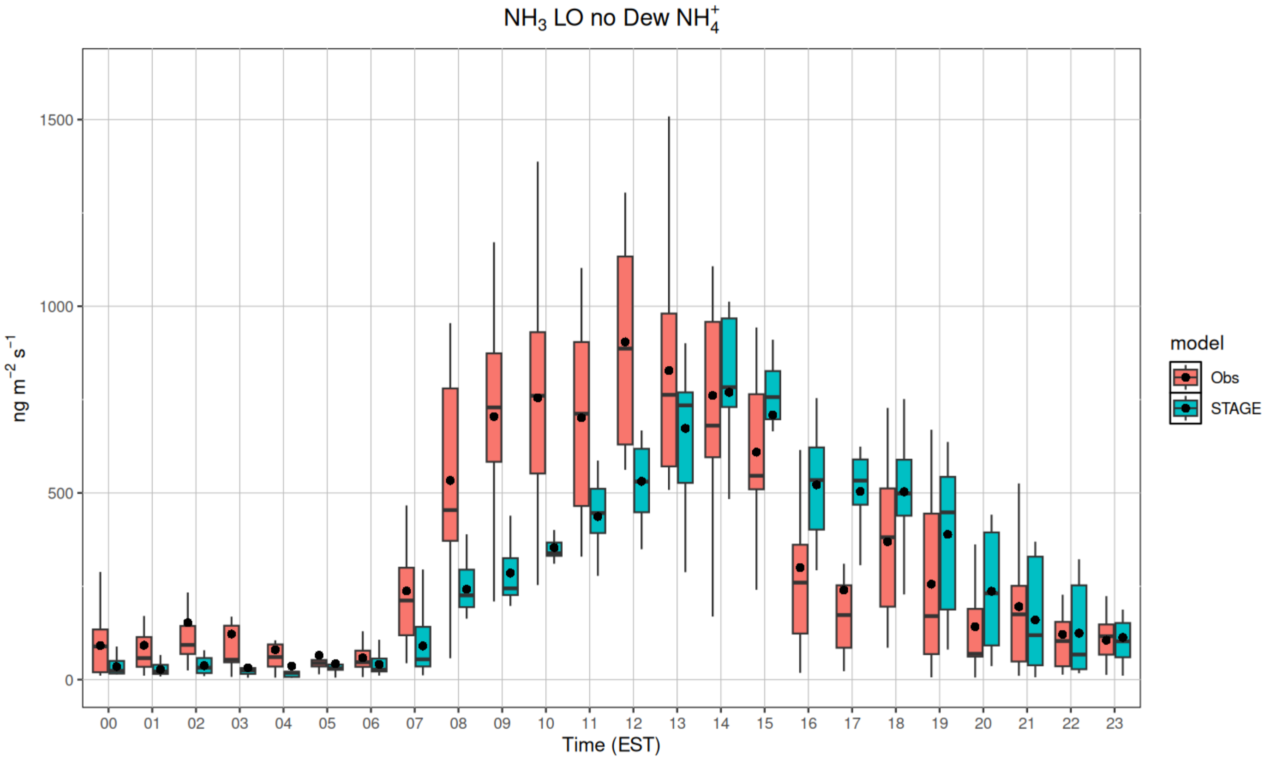
Line 325: In Figure 3, STAGE overestimates during 7-13h, and underestimates during 14-23h. This is an interesting contrast and is expected for detail explanation. Is there any

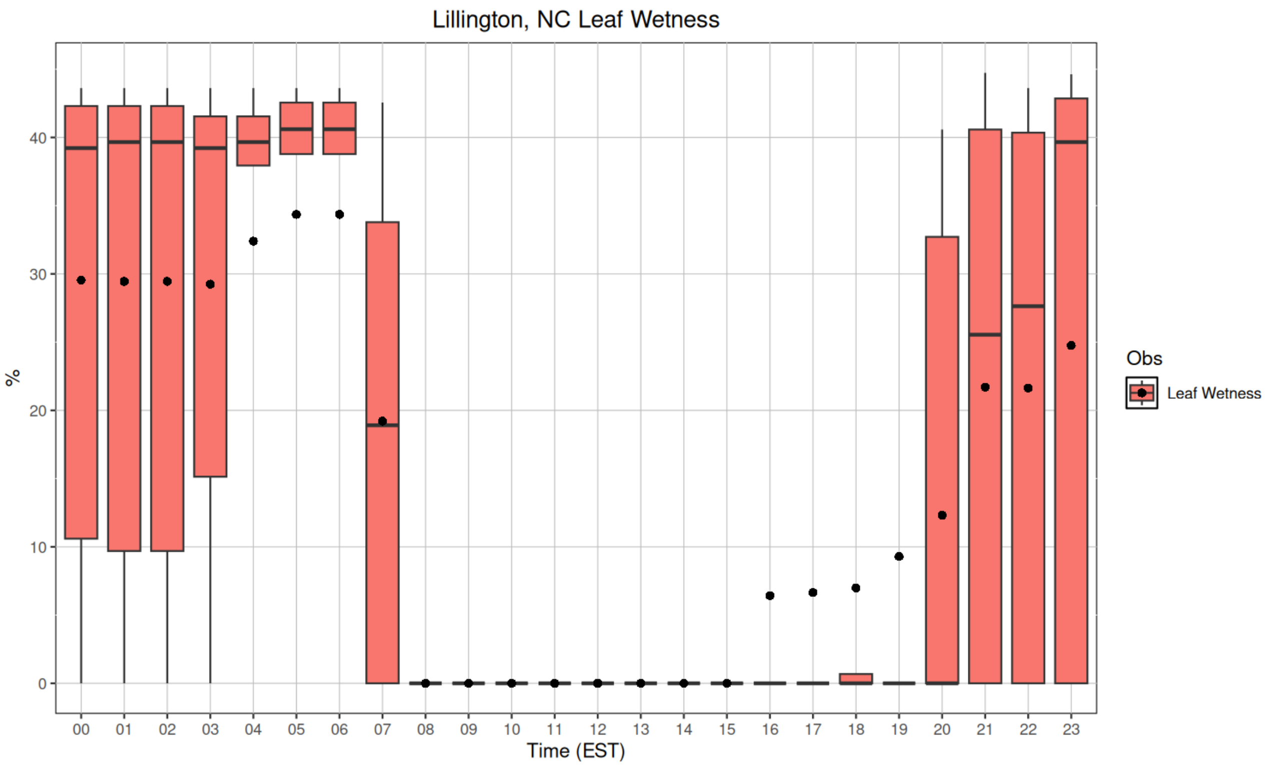
possibility that NH3 flux is related to daytime meteorological factors, such as intense solar radiation?

Response: We agree that this is interesting and have added a more detailed discussion and figures showing the impact NH3 evasion from dew at the Lillinton, NC site in the SI. This can also be seen in figure 2 but is likely more pronounced at the Lillington site due the higher ammonium concentrations in the soil, vegetation, and dew. In general, we have not been able to capture the morning peak in NH3 emissions. The emissions appear to be related to the drying of the canopy in the morning and is accompanied by a morning peak in emissions (Walker et al., 2013; Wentworth et al., 2016). We did measure emission emission potentials on the same order of magnitude as stomatal sources in the leaf dew water that we collected in the morning but the evasion form this source does not completely account for the observed emissions in agreement with the findings in Walker et al., 2013. The following was added to section 3.1

A consistent min morning emission peak was observed at the Lillington, NC site (Walker et al., 2013). The emissions appear to be related to the drying of the canopy in the morning and similar morning emissions have been observed for a grass canopy (Wentworth et al., 2016). A high emission potential was measured on the dew present on the leaves in the morning, Table 2. When the dew compensation point was included in the STAGE model, case LO, there was insufficient ammonium in the dew to explain the emission peak, Figure S1, in agreement with Walker et al., 2013. At the Lillington site, the canopy did not contain dew according to the leaf wetness measurements after 8:00 EST, Figure S2, and the modeled evasion from dew occurred before this period while the observed morning evasion occurred primarily between 8:00 and 11:00 EST when the canopy was dry, Figure 3. The soil between plants and rows at the Lillington site was exposed and had a much higher emission potential than the canopy. We speculate that the mid morning emission peak could be due to the wetting and drying of surface this NH4+ rich soil that was not captured by the soil moisture probes. These results are in contrast to the the dew drying experiments of Wentworth et al., 2016, where they estimated that the evasion of NH3 during morning dew evaporation could account for morning increases in NH3 at a high elevation grassland field site at Rock Mountain National Park, CO.

The following figures have been added to the SI:





Line 326: How well is the model performing in capturing SO2, HNO3, and NH3 after the measured soil and canopy parameters are used? Please include the statistics before and after the parameter changes.

Response: The use of observational data only impacted the compensation points and NH3 flux as we did not have an observed minimum stomatal resistance for the Duke

Forest site where we measured the SO2 and HNO3 fluxes. The changes in the NH3 statistics using the tabular CMAQ parameterization and the observed values are in Table 3.

Line 328-329: What do you mean by CMAQ tabular data? Are you referring to the default setup? How poor is the original STAGE? Pls explain with the bias or correlation index.

(add to discussion)

Response: Yes, this is referring to the default CMAQ v5.3.2 setup. In Table 3, the LB and DFB cases use the default CMAQ/STAGE parameters. The STAGE model evaluated well against Lillington observations but was initially uncorrelated with the flux observations at Duke Forest. However, the model performed well when observed emission potentials were used as inputs. With the exception of the morning emission peak, the STAGE model can capture the magnitude and dynamics of the observations well indicating that improving the characterization ammonium in surface vegetation and soil media have the potential to improve modeled NH3 and reduced nitrogen fluxes.

Line 409-410: Are you referring to Figure 4?

Response: Yes, this has been corrected.

Technical comment:

Line 81: Two fullstop.

Response: Thanks, this has been corrected.