Response to Reviewer #2:

We would like to thank the reviewer for their careful and thorough reading of this manuscript and for the thoughtful and constructive comments and suggestions, which will help improve the overall quality of this manuscript. Our responses are denoted in **red**.

General Comments:

This manuscript presents a good showcase of the use of the next-generation global model MUSICAv0 with regional refinements to study the summertime distribution of ozone and its precursors in the Southeast Michigan region (SEMI) evaluated against the observations from the MOOSE campaign in 2021 and ground-based measurements. As one of the first studies to evaluate MUSICAv0 simulations with an extended campaign, this manuscript would be a notable publication. The study discusses in particular the effect on model performance of using higher grid resolution and implementing a diurnal cycle of anthropogenic NO emissions. It is shown that higher grid resolution is more important for simulating the distribution of O3 precursors than O3 itself, while implementing a diurnal cycle of anthropogenic NO emissions can improve model performance for nighttime O3. This conclusion is in agreement with the other modelling studies.

While this study clearly shows the advantage of using a global model with regional refinements, such as MUSICA, over the conventional global model, the manuscript does not discuss how these new generation models might improve on regional models. Perhaps the authors can add a short discussion on this issue and how these new generation models can be applied to better study regional air quality problems.

I recommend that this manuscript be published with the following comments and suggestions:

We thank Reviewer #2 for their careful observations, and appreciate their feedback and recommendations for improving the manuscript. We have carefully gone through all of your comments, and have addressed them below and in the main text.

Section 1

Line 56: A brief description of the instruments involved in the MOOSE campaign could be included here to provide a more comprehensive introduction to the campaign.

Response: To show the scope of MOOSE, we have added the sentence: "The MOOSE observations included a mobile lab with detailed measurements of ozone and its precursors, ground-based remote sensors (i.e., Pandora), and an airborne remote sensor (i.e., GCAS)."

Line 100: Please explicitly mention CAMS-GLOB-ANTv5.1 here, as there are a number of CAMS emission datasets. The resolution of the emission data (0.1 degree, ~10 km) can also be mentioned here to illustrate that the emission resolution is comparable to the model grid resolution. Please add a reference to the CAMS emissions dataset used here.

Response: The details on the emission datasets used for the simulations are presented in Section 2.1.3. As this sentence is about the diurnal variation, mention of the specific anthropogenic emissions has been left out here.

Line 103: Please add "emissions" after the end of the sentence

Response: The correction has been made.

Section 2.1.2

- Can the authors explain why the ne30x8 configuration covers the entire CONUS instead of just over Michigan?

Response: The ne30x8 configuration over CONUS is the default resolution used in MUSICAv0 and is mentioned in lines 110-112 (https://wiki.ucar.edu/spaces/MUSICA/pages/418448638/MUSICA+Home). The authors decided to use this grid mesh rather than create a new grid mesh over Michigan at 1/8-degree because it is a ready-to-use configuration with many of the input datasets readily available and in an NCAR repository. It was also a way of gauging the efforts and computational cost associated with creating a new grid mesh versus using an already available mesh.

Line 135: Can you include a reference to the Community Mesh Generation Toolkit?

Response: A citation for this software has been added.

Section 2.1.3

- A more updated version of the CAMS-GLOB-ANT dataset should be considered in the future. For temporal profiles, the CAMS-GLOB-TEMPO datasets may be useful.

Response: At the start of this work, the latest available inventory was used. A note of this to be considered in the future has been added in the conclusions section as: "Future work should also take into consideration the use of a more updated version of the CAMS-GLOB-ANT emissions, as well as the diurnal variation profiles of CAMS-GLOB-TEMPO (Guevara et al., 2021; Soulie et al., 2024), or more regional emission inventories such as the National Emission Inventory (NEI) from the US EPA."

Section 2.1.4

- Apart from the diurnal cycle of NOx emissions, the evolution of the PBL probably plays a role in the daytime and nighttime O3 and NOx concentrations. Can the authors comment briefly on this?

Response: We acknowledge that apart from the diurnal cycle for anthropogenic NO emissions, the evolution of the planetary boundary layer (PBL) can play a significant role in O3 and NOx formation. We have added this statement in the conclusions acknowledging this uncertainty: "In addition, we acknowledge that apart from applying a diurnal cycle for anthropogenic NO emissions, the evolution of the PBL can also play a significant role in the formation of O_3 and NO_x. In the daytime, a rising PBL can mix surface NO_x and VOCs upwards, reducing O_3 concentrations near the surface, while in the nighttime, a shallower PBL can trap emissions near the surface leading to higher NO_x titration. Uncertainties associated with the PBL could lead to underpredictions of NO_2 in the model and misrepresentations of O_3 peaks."

Section 2.2.1

Table 2: First column, first row: "Selected" VOCs

Response: This has been corrected in Table 2.

Section 3.1

- The diurnal cycle of NO emissions probably plays little role in meteorology. Please consider focusing the discussion on the effect of model grid resolution and select specific time periods where the simulations of the two resolutions show significant discrepancy for discussion.

Response: We have expanded the discussion here and have elaborated a bit further on the meteorological consistencies and inconsistencies. We have also included two additional time series (see Figure 3) plots for wind speed and wind direction to further show the meteorological performance from simulation to simulation.

Expanded Section 3.1: "SEMI is a region that faces unique air quality challenges due to large industrial and automotive activity, dense population, and geographic factors. SEMI has a diverse terrain, ranging from highly urbanized areas, such as the city of Detroit, expansive agricultural lands in more remote areas, and forests surrounded by both inland and coastal lakes. The region consists of a relatively flat terrain, with a humid continental climate. Additionally, large air masses of humidity can be transported into the region from the Great Lakes (i.e., Lake Huron and Lake Erie) through the lake effect winds (Scott and Huff, 1996). A time series along the AML track of meteorological values – temperature, relative humidity, planetary boundary layer height, cloud total, wind speed, and wind direction – from the models (and observations for temperature and relative humidity) are shown in **Fig. 3**. During the campaign period in the summer of 2021, temperatures reached up to approximately 305 K and relative humidity to almost 100%. The planetary boundary layer reached more than 2500 m on most days, while cloud total was relatively varied. Modeled wind speeds follow the trend for the campaign period quite well, but are comparatively high compared to the observations, while wind directions perform generally well except on some specific days. The AML track covered a large part of the SEMI region, making its way through both very urban and rural areas. Meteorological parameters, such as temperature, are highly impacted by

urbanization through the reductions in vegetated land cover and increases in energy consumption (Wang et al., 2021). Urbanization can lead to higher temperatures, and thus increasing O_3 production. In the simulations presented here, meteorological parameters (i.e., temperature and horizontal winds) are nudged towards reanalysis data to obtain a more realistic depiction of reality in the coarser resolution regions, leaving the regional refinement area to freely run, as the resolution of the refined area is finer than the resolution of the reanalysis dataset that is being used. Regional refinement grids, with high horizontal resolution, are capable of resolving areas with large geographical differences (Jo et al., 2023). Meteorological fields in these simulations are generally consistent indicating that meteorology is performing similarly, even with the changes in horizontal resolution. Although temperatures, relative humidity, and planetary boundary layer height remain consistent among all the simulations, cloud total varies between the simulations, which can significantly impact photochemical production."

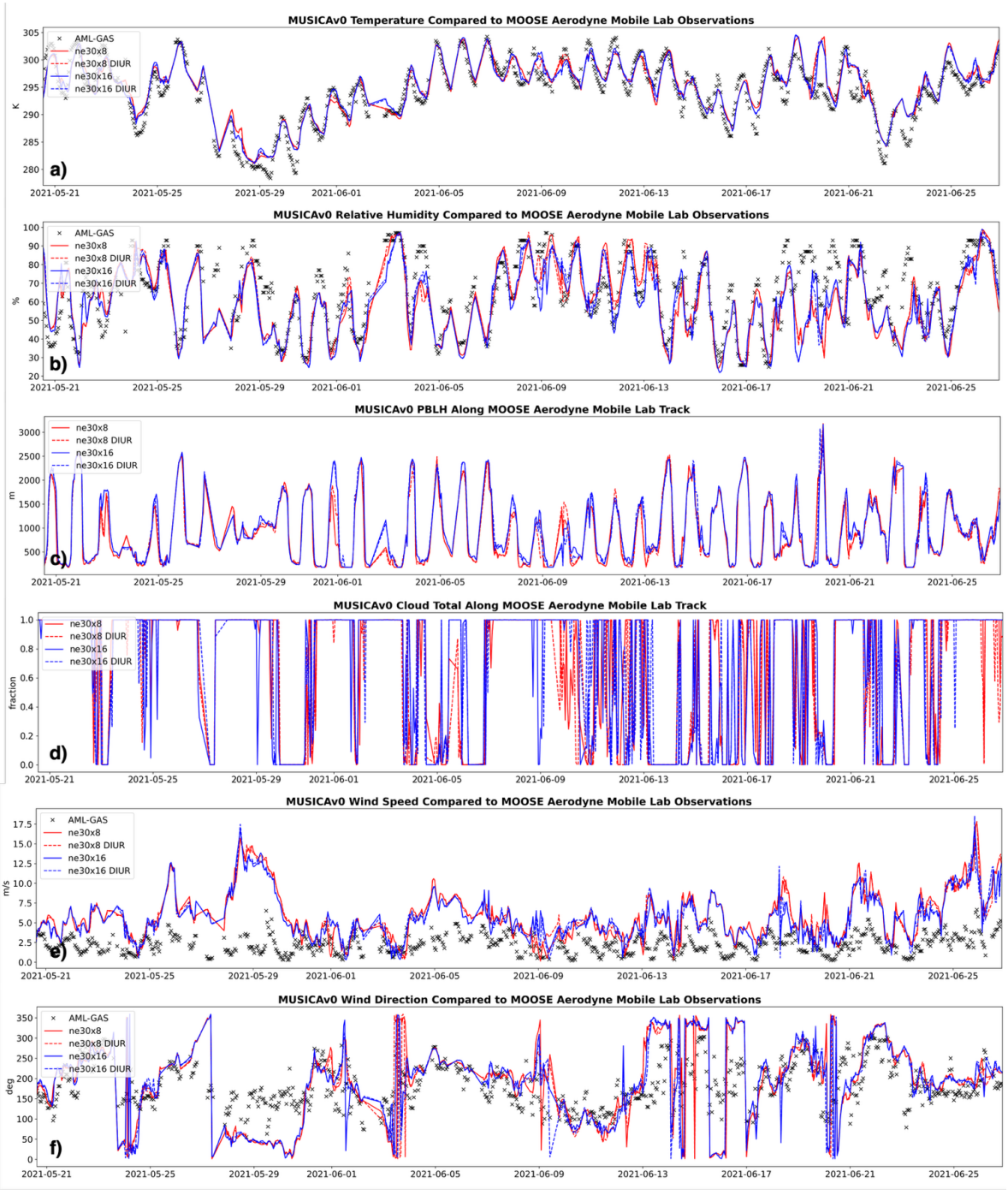

Figure 3: Time series of (a) temperature, (b) relative humidity, (c) planetary boundary layer height, (d) cloud total, (e) wind speed, and (f) wind direction along the Aerodyne Mobile Laboratory (AML) track. Measurements of temperature and relative humidity were available and displayed as black x's in Fig. 3a and 3b. The model results are shown in red (ne30x8) and blue (ne30x16) corresponding to horizontal resolutions. The dashed lines represent model simulation results when adding the diurnal cycle for nitric oxide anthropogenic emissions, color-coded to their respective horizontal resolution.

Section 3.2

Line 326: Please state in the text that Fig. 4 is a time series of hourly averaged diurnal profiles of ozone concentrations over a specific time period.

Response: As the reviewer suggests, text further describing Fig. 4 has been added. Here is the updated figure description: "The evaluation of the four model simulations with stationary measurements for O_3 at seven locations in SEMI – Allen Park (Suburban Downwind), Detroit-E 7 Mile (Suburban), New Haven (Rural), Oak Park (Suburban, Near Highway), Port Huron (Urban Port), Warren (Suburban), and Ypsilanti (Suburban, Near Highway) – are shown in **Fig. 4** as a time series of their hourly averaged diurnal profiles during the MOOSE campaign."

Figures 4 and 5: Please try to show time series of O3 and NO2 concentrations from the same stations in the same order for better comparison

Response: O3 and NO2 are not available at all sites, so Figures 4 and 5 necessarily show different sites. Out of the shown sites, only one – Detroit – E 7 Mile – has data available for both O3 and NO2. We have included all of the sites in SEMI with data available to show comparison with a wide range of sites in different locations throughout the area. Additionally, in the discussion section (Sec. 4), we include diurnally averaged plots (Figs. 15 and 18) that compare multiple species at several locations to have a more representative perspective on O3 production and loss in the area of study.

Line 358: Can the authors explain in more detail how O3 concentrations are affected by the aforementioned effect on NO2 concentrations?

Response: We have included a more detailed explanation of how O3, NOx, and VOCs are intertwined in the beginning of Section 3, to tie together the different evaluations being done, as so: "O_3 concentrations are highly associated with NO_2, where NO_x, in general, plays a critical role in the photochemical production and destruction of O_3 in the presence of sunlight. O_3 production in the troposphere is largely dependent on the availability of NO_x and VOCs, and can give great insight on O_3 control. This dependency is classified into NO_x- and VOC-limited regimes. In a NO_x-limited regime,

the rate of O_3 production relies on the abundance of NO_x and increases with NO_x concentrations, but is not dependent on the concentrations of VOCs (Wang et al., 2019) . In action, decreasing NO_x concentrations would lead to reductions on O_3 (Jacob, 1999). On the other hand, in a VOC-limited regime (or NO_x-saturated regime) the rate of O_3 production increases with VOC concentrations and is not dependent of NO_x (Wang et al., 2019), therefore reducing the amount of VOCs would lead to reductions in O_3 (Jacob, 1999). The chemical relationship between O_3-NO_x-VOCs is critically important for defining mitigation strategies set to improve O_3 from region to region."

Section 3.3

- Can the authors also discuss whether there are significant differences between daytime and nighttime concentrations between the four simulations?

Response: Overall differences between daytime and nighttime concentrations between the simulations are detailed in Sec. 3.2 of the main text, where the simulations are compared to stationary measurements of O3 and NO2. In Sec. 3.3, we omit a discussion on daytime and nighttime concentrations because we are summarizing the data from AML and the models with a Taylor diagram. We do this because for data along the AML track, the mobile lab was moving on different days in different locations across SEMI (some urban areas, some more remote areas, etc.), and stationary at night. Comparing hourly averaged data for this section, may have been misleading if trying to compare the entirety of the campaign. If we were to include specific case study days, a discussion of the daytime and nighttime performance would have been critical. The goal of using the Taylor diagram here was to summarize the overall model performance compared to these observations.

Section 3.4

- The authors should better illustrate how the Pandora measurements can be related to the stations and AML measurements and how these comparisons can lead to the different performance of simulated O3 concentrations.

Response: This is a great suggestion, as we realize we do not quite explain why we use different datasets for the model evaluation. We have elaborated further at the beginning of the results section (Sec. 3) to emphasize why model evaluations are needed – "We evaluate the models using diverse datasets from MOOSE for a comprehensive analysis, as no single dataset has the ability to capture all aspects of atmospheric composition (e.g., emissions, chemistry, transport, meteorology). These different datasets can also help capture different aspects of a model such as near-surface chemistry (i.e., in-situ measurements) and column burdens (i.e., aircraft-based remote sensing), to determine model skill, characterize model errors, improve model representation, and measure our confidence in the model results for reproducing reality" – and what needs to be considered, as well as adding a comment at the beginning of Section 3.4: "Both Pandora monitoring sites (SWDetroitMI and DearbornMI) were located in an industrial and high-traffic setting, providing continuous observations in urban conditions and complementing the other observations".

Section 3.5

- Instead of narrative in the text, the authors could include wind vectors or maps of meteorological variables to illustrate how the different models capture the NO2 plumes at the different times shown, as the readers may not be familiar with the geographical locations shown.

Response: We have added maps for each of the days to include wind vectors and temperature to the supplemental information (Figures S16-S31), and have referenced them in the main text (line 534) as so "The direction of the pollution plume are supported by plots of temperature and wind vectors in Figs. S16-S31 in the SI for each of the flight days".

Section 4

Figures 13 and 14: The authors should explain why they show the conservatively regridded model outputs in panel (c).

Response: We thank the reviewer's attention to these figures. We have added an introduction to this discussion to motivate the comparison of the regridded high resolution output to the coarser resolution:

"To quantitatively assess the impact of the finer resolution on the simulation of ozone and its precursors, the ne30x16 (7 km) results have been conservatively regridded to the ne30x8 grid. These regridded results illustrate the impact model resolution can have on atmospheric chemistry."

Line 512: The authors should explain the consequences of the regridding method not being able to reproduce the higher resolution simulation results

Response: We've incorporated a couple of sentences in/around line 589 to address these consequences: "When the model is run at 1/16˚ horizontal resolution, localized features (e.g., pollution plumes, sharp emission gradients) are better resolved and land use is better represented."

Figure 15: Please consider also including the observed concentrations of O3, NO and NO2 in the time series to better illustrate which model configuration is closer to the observed values.

Response: Thank you for your suggestion. The available observed values for O3, NO, and NO2 at these stationary monitoring locations are presented in Section 3.2 in Figures 4 and 5.

Section 5

Line 618: In addition to the South Korean simulation, can the authors compare their work with other studies using MUSICA over other regions of the USA?

Response: We have included a sentence comparing this work with Schwantes et al. (2022) which used MUSICAv0 to study the Southeastern US: "Schwantes et al. (2022) found that O_3 was better simulated over urban regions across the Southeastern US, especially when using a ~14 km regional refinement grid and updated chemistry in MUSICAv0. This work took into consideration a finer grid resolution mesh (~7 km) and compared to ~14 km to show that regional refinement improves O_3 representativeness in the model."

- The authors should consider briefly discussing how MUSICA or similar next-generation global models with regional refinement capability can be used to formulate regional/local air pollution monitoring and mitigation strategies.

Response: We have expanded Sec. 4 to include a better link with Sec. 3. We have also established a better discussion on different regimes governing SEMI and what potential mitigation strategies could be applied to the area to improve air quality. We have also discussed future work in helping define potential mitigation effects.

These linkages are reflected in Section 4 as so:

- "This difference results in an improvement for the ne30x16 simulations based on the findings from in Fig. 4, where peak O_3 performed best in the finer resolution simulations when compared to the surface sites."
- "These findings are directly supported by **Fig. 3**, where although temperatures between the simulations are not significantly different, there are changes in cloud totals and winds that could impact solar radiation and thus the isoprene emissions. The differences in temperature between the resolutions are also illustrated in the maps in **Fig. S16-S31** in the SI."

An extended discussion on the different regimes governing SEMI and potential mitigation is added to the end of Section 4: "The findings of this study show that O_3 production in SEMI is strongly governed by the spatial distribution of emissions and different chemical regimes. The urban location analysis showed that Detroit, which is a major industrial hub in the region, is consistent with a VOC-limited regime, where in the daytime, O_3 concentrations are suppressed by high NO_x titration, but can become sensitive to changes in VOCs during peak O_3 times. The suburban and remote location analysis (i.e., Allen Park and New Haven, respectively) showed that they are in a more NO_x-limited regime, where higher BVOCs and lower NO_x titration can lead to more efficient O_3 production. The spatial distribution is seen more clearly as we move towards finer resolutions indicating more realistic emissions.
In VOC-limited regimes, targeting reductions in VOCs, such as those from the industrial sectors, is crucial compared to reductions in NO_x, as it could lead to temporary increases in O_3 production. In NO_x-limited regimes, where NO_2 drives O_3 production, reductions in transportation emissions and long-range transport would decrease O_3. The improvement in model representation of NO_2 and in turn, O_3, during rush hour times (**Fig. 4-5**) shows how emissions can be misrepresented in the models. It is

necessary that future work considers incorporating higher resolution temporal profile and regional emissions to better distinguish different O_3 processes. Future work should also explore the impacts of targeting the contribution of different emission scenarios in SEMI to demonstrate the impact of different regulatory decision-making. ”