Response to Reviewer #1:

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:

The manuscript analyzes the distribution of ozone and some of its precursors over the Southeast Michigan (SEMI) region during summer 2021 based on model simulations with MUSICAv0 and observations from the MOOSE filed campaign. The authors discuss the impact of grid resolution and diurnal cycle of anthropogenic NO emissions and show that night-time ozone is mostly improved by applying diurnal cycles for NO emissions, while grid resolution is found to have more impact on ozone precursors. The study also shows that using a good conceptualization of grid resolution within MUSICAv0, with finer resolution could lead to more efficient computational costs, which could be beneficial for other local-scale studies including in other regions.

The paper shows the interesting potential of using global models with zooming capabilities like MUSICAv0 to investigate air pollution characteristics even at specific small regions like SEMI. Overall, the paper is well structured and easy to read. However, the analysis and discussion sections are in some cases rather short and could be further improved in order to better identify the processes controlling summertime ozone in different parts of the SEMI region.

I recommend the manuscript to be accepted for publication after addressing the following comments and suggestions:

Response: We thank Reviewer #1 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 2.1.1: Initial conditions are considered from a restart file based on MOZART-TS1. Which initial conditions are considered for the additional species in TS2 not included in TS1?

Response: We thank the reviewer for their observation. We have added a sentence addressing the usage of the initial condition file with TS1 for TS2 simulations in Section 2.1.1: "Although the initial condition file was based on MOZART-TS1 chemistry and the additional species in MOZART-TS2 were initiated from zero, the majority of these species are short-lived and equilibrate quickly within the one-month spin-up period."

Section 2.1.3:

- Anthropogenic emissions are considered from CAMS_GLOB_ANTv5.1. A recent study from Soulie et al. (2024, ESSD) shows significant differences in the estimated emissions between CAMS_GLOB_ANTv5.1 and the EPA inventory in USA. In particular, EPA exhibits higher NMVOCs but lower NOx and SO2 emissions. Can the authors comment on the potential impact of such uncertainties in emissions on the model results?

Response: Yes, we acknowledge the discrepancies between CAMS_GLOB_ANTv5.1 and the US EPA's National Emission Inventory (NEI) described in Soulie et al. (2024). The differences in the emissions, especially the high NMVOCs and low NOx and SO2, in NEI compared to CAMS definitely has the potential to introduce uncertainties in the model results because emissions directly influence atmospheric chemistry and pollutant concentrations. Increased availability of NMVOCs could lead to increased O3 production, especially in VOC-limited regimes (i.e., more urban areas; O3 decreases with increase in NOx and increases with increase in VOC) and potentially alter the oxidizing capacity of the atmosphere. On the other hand, in NOx-limited regimes (i.e., more rural areas; O3 increases with increases in NOx and changes very little with changes in VOC), the lower availability of NOx could reduce O3 concentrations in the model. While these uncertainties reflect broader challenges pertaining to emission inventories such as spatial distribution and sectoral estimates, our study uses CAMS for consistency with other global studies, and explores the impact of relative changes such as adding diurnal variation of emissions. Future work should include the use of regional inventories, such as NEI, or inventories derived from inverse modeling. In addition, the lower emissions for NOx and SO2 could also alter secondary

aerosol formation (e.g., lower NOx could decrease nitrate aerosol formation). Although this is a non-negligible issue, it is out of the scope of this study and should be addressed in future work.

We included a sentence commenting on future considerations in Section 5 as so: "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."

- It is not clear how soil NOx emissions are considered for the simulations.

Response: Global soil NOx emissions are based on the natural emissions of NO as described in Emmons et al. (2020). This is now mentioned in Section 2.1.3 as so: "Other emissions, from soil, lightning, volcanoes and oceans, are described in Emmons et al. (2020)."

- The authors include calculated NO emissions from agriculture waste burning (AWB) in Table S1, but it is not clear if emissions from this sector are considered or not. This could lead to double counting of emissions with QFED, although the contribution of NO AWB emissions seems to be minor compared to other sectors.

Response: CAMS anthropogenic emissions from all available NO sectors are considered when applying the diurnal cycle, where we've used a sector-based and country-specific temporal profile. There is a possibility that agricultural waste burning (AWB) emissions could be double counted in the emissions, as QFED uses satellite observations of the fire radiative power (i.e., rate of radiative energy emitted by an active fire) to estimate global gridded fire emissions, but for our region of study, this impact is minimal. Regardless, we have added a footnote in Table S1 of the Supplemental Information to note this uncertainty (see below).

Table S1: Total anthropogenic nitric oxide (NO) emissions from the CAMSv5.1 in Michigan and Southeast Michigan, and their ratio from various sectors*.

	MICH [kt]	SEMI [kt]	SEMI/MICH
AGS	0.46	0.04	9.0%
AWB[1]	0.10	0.01	10.8%
ENE	9.44	2.86	30.3%
RES	1.03	0.48	47.1%
TNR	1.99	0.36	18.1%
TRO	15.13	3.50	23.2%

*AGS = Agriculture Soils; AWB = Agriculture Waste Burning; ENE = Power Generation; RES = Residential; TNR = Off-Road Transportation; TRO = Road Transportation

[1]It is possible that AWB emissions could be double counted via biomass burning emissions from QFED (QuickFire Emissions Dataset), where the fire radiative power obtained from the satellite is used to estimate the global gridded fire emissions (Darmenov and da Silva, 2015). Although it has been found that AWB can increase fire emissions over regions, in Southeast Michigan this contribution is minimal.

Section 2.1.4:

- Can the authors comment why only NO diurnal distribution is considered, while diurnal distribution of other species like VOCs or SO2 could also impact the model results?

Response: We acknowledge that the diurnal distribution of VOCs and SO2 could impact model results, especially for O3 chemistry. In our study, we focus on applying the diurnal cycle for anthropogenic NO emissions due to its dominant role in controlling tropospheric O3 and titration processes, which are highly sensitive in areas dominated by industrial and transportation-based activities, like SEMI. We recognize that applying diurnal variation for VOCs and SO2 could also affect O3 production as they can be temperature-driven (e.g., biogenic VOCs) and based on industrial activity. Future work will incorporate temporal profiles for all available anthropogenic emissions, as they would help refine model results and further assess the impact on other critical air pollutants.

We have added text to Section 2.1.4: "While emissions of other anthropogenic compounds, such as VOCs, do have diurnal variations, we have only implemented the diurnal variation for NO emissions in this work, due to its dominant role in controlling tropospheric O3 and titration processes."

- Including a figure showing the diurnal distribution of NO emissions from different sectors, as used in the simulation is a useful information.

Response: We have included a figure of the diurnal profiles for each sector in Fig. S2 in the supplemental information of the manuscript and reference it in Section 2.1.4.

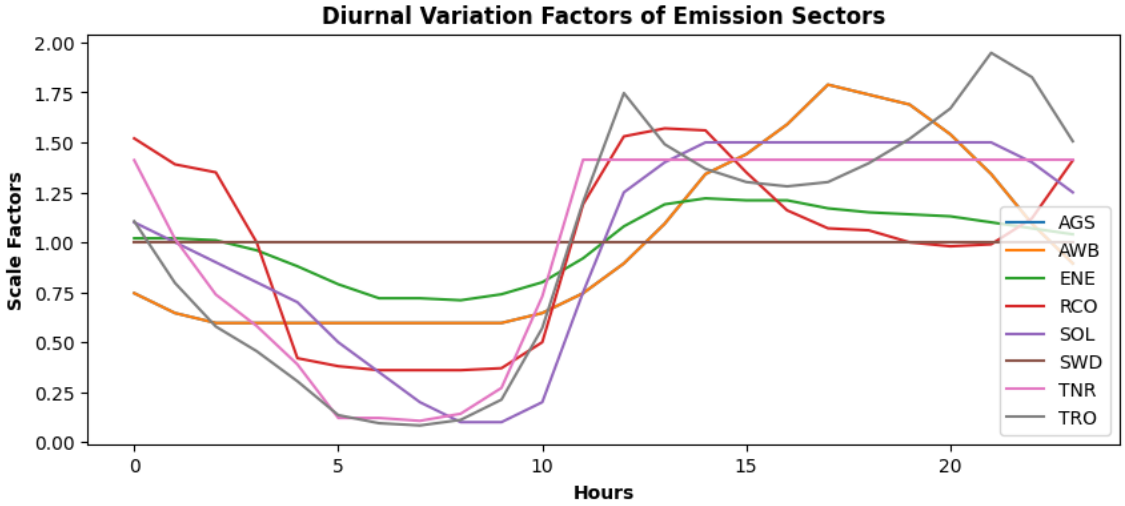

Figure S2: The diurnal variation scale factors applied to NO emissions for each anthropogenic emission sector used in the simulations.

Section 3.1:

- This section is rather short and doesn't fully cover the model's ability to capture meteorological features in the considered region. In addition to the model evaluation, this section is also expected to contain a description of the meteorological situation that characterized the SEMI/MI region during the campaign period. This section can also be significantly improved by considering other meteorological variables (e.g. wind speed/direction), other networks or datasets (e.g. reanalysis).

Response: We have expanded this section in the main text to elaborate further on the campaign period and the presented figure. We have also included a time series comparison of the AML wind speeds and wind directions to better support this section.

Expanded Text: "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."

Updated Figure:

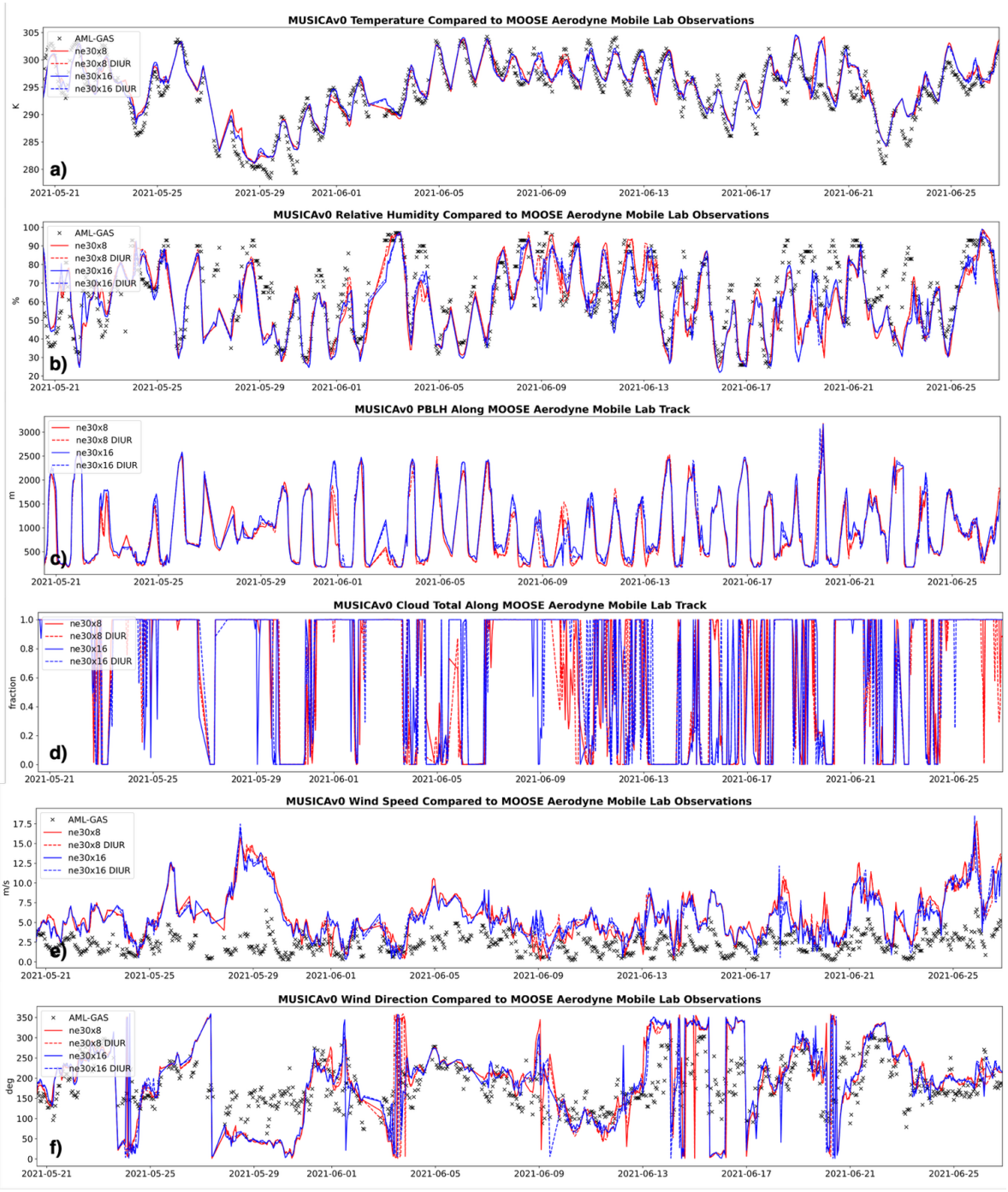

Figure 3: Time series of (a) temperature, (b) relative humidity, (c) planetary boundary layer height, (d) cloud total, (e) wind speed, and (f) wind direction along the Aerodyne Mobile Laboratory (AML) track. Measurements of temperature and relative humidity were available and displayed as black x's in 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:

- The authors could elaborate a bit more the discussion on the reasons behind the diurnal changes in the model bias and link with results in Sect. 4. For this, a map showing location of the stations could be very useful.

Response: A map that includes the stationary site locations for this evaluation can be found in Fig. 2 of the manuscript. A stronger link has been made between Sec. 3 and 4 to better enhance the discussion.

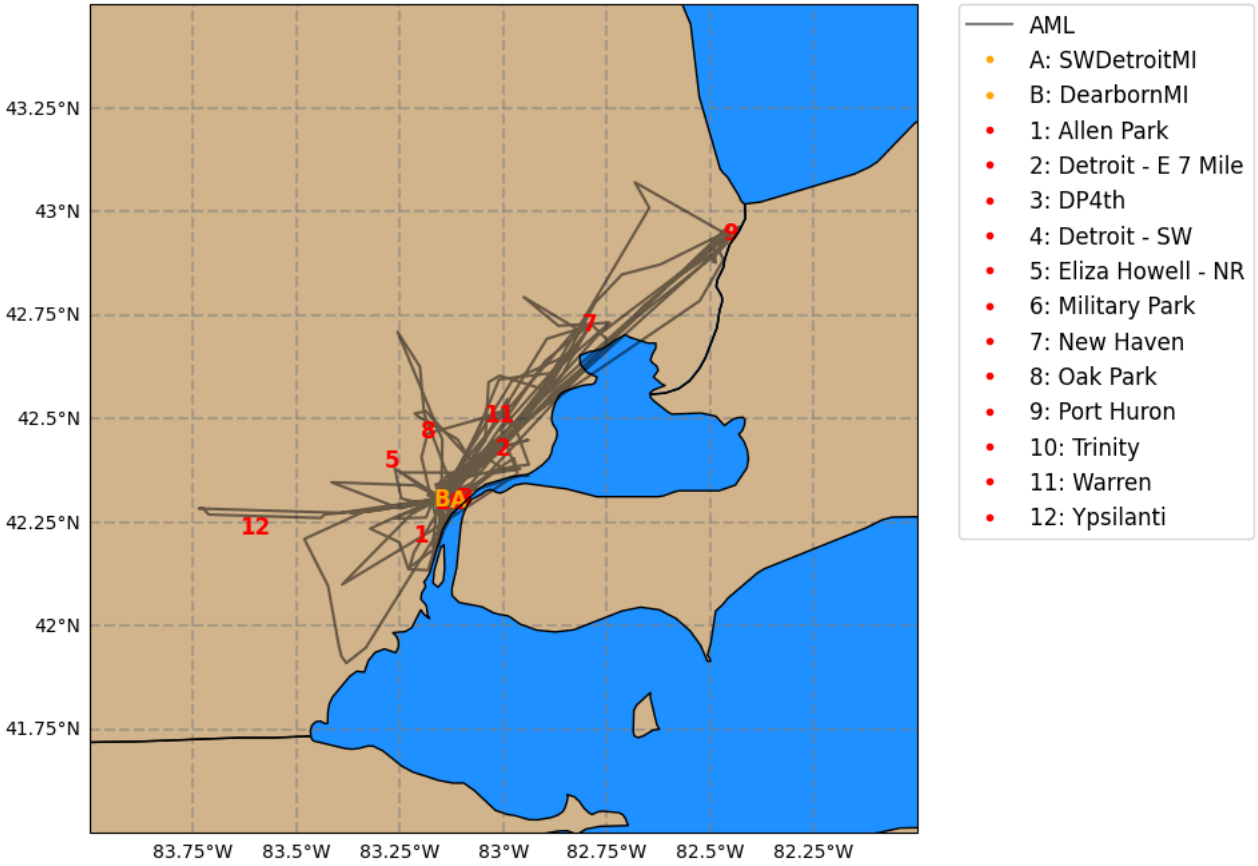

Figure 2: Location of observations from Phase I (24 May to 30 June 2021) of the Michigan-Ontario Ozone Source Experiment (MOOSE) used in this study. The gray line shows the track of the Aerodyne Mobile Laboratory across Southeast Michigan. Stationary sites from the Michigan Department of Environment, Great Lakes, and Energy (MI EGLE) are shown as the red numbers (1-12), and the Pandora monitoring sites are shown as the yellow letters (A-B).

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."

- The night-time NO2, in particular between 00 and 05 AM, although improved, remain high and the morning peak is less visible when NO diurnal cycle is applied. The authors should discuss the impact of potential uncertainties in the considered diurnal cycle, including the fact that this was applied only for NO.

Response: We thank the reviewer for their insightful observation regarding the nighttime NO2 concentrations and the less prominent morning peak when the diurnal cycle for anthropogenic NO emissions is applied in the model. We acknowledge the potential uncertainties in the presented diurnal cycle and have expanded our discussion to take this into account.

- Possible causes for the high nighttime NO2 concentrations include an overestimation of nighttime NO emissions. While the temporal profile applied to NO scales the anthropogenic emissions, some emission sectors (i.e., ENE, AWB; see temporal profile figure) have flatter temporal cycles that could lead to the sustained concentrations of NO2 via the reaction of NO and O3. Nighttime NO2 can also accumulate due to reduced O3 titration, as well as due to a shallow nighttime boundary layer that can trap NOx concentrations.
- The less prominent peak could be due to the temporal profile application to anthropogenic NO emissions, as the morning rush may be too gradual, so the NO2 peak appears more delayed than the observed values. In addition, the morning VOC emissions from transportation-related activities could further enhance the NO-to-NO2 conversion, but since VOC diurnal cycles are not included, this feature could be underrepresented.

We have added a few sentences stating this uncertainty in Section 5 of the main text: "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 3.3:

- The authors relate the differences in simulated isoprene (and hence HCHO?) to potential changes in meteorological field leading to changes in calculated BVOCs. Although this could be true, no results (i.e. changes in meteorology) are provided to assess this especially in the discussion in Sect. 3.1.

Response: Thank you for pointing out this discrepancy between the sections 3.1 and 3.3. As mentioned in an earlier part of this review, some necessary corrections and additions to section 3.1 have been made. Based on this, we have added some further explanation on what else could be impacting simulated isoprene. We have added discussion of how the biogenic emissions could be impacted by changes in cloud totals between the simulations.

This new addition to section 3.3 is: "Although temperatures are not greatly affected by grid resolution, as was seen in **Fig. 3**, cloud totals are different in the two resolutions, which can impact the amount of solar radiation reaching the surface. Clouds in the model can be impacted by several changes, such as changes in aerosols, which is out of the scope of this study, or related to changes in meteorology (e.g., winds). Yan et al. (2023) demonstrated that aerosols are able to impact precursor accumulation and photolysis (e.g., isoprene), where tropospheric chemical loss is enhanced due to photolysis and NO_x accumulation. Cheng et al. (2022) also found that changing clouds in chemical transport models can impact photochemical reaction rates and BVOCs. Future work on evaluating model grid resolution and diurnal cycle impacts on O_3 formation should look more closely into aerosol-cloud interactions and how they impact photochemical production in SEMI."

- Similarly, the discrepancies in other species (hydrocarbons and aromatics) is explained by misrepresentation of their anthropogenic sources in the CAMS inventory. The authors can assess such uncertainties in the considered emissions by comparisons with EPA emissions in SEMI.

Response: We have made a quick comparison of CAMSv5.1 emissions with HTAP_v3.1 mosaic, which includes emissions from the US National Emission Inventory (NEI) (Crippa et al., 2023). Because HTAP_v3.1 is only available up until 2020, we have compared with CAMSv5.1 for 2020 to illustrate the misrepresentation of emissions based on the emission inventory without year-to-year discrepancies. We have also included emissions for 2021 from CAMSv5.1 to show the differences between years. The emission totals shown in this table are representative of summertime emissions (June, July, August) for a domain over the state of Michigan (longitude: 273˚W to 278˚W; latitude: 41.5˚N to 46˚N). The comparison of CAMSv5.1 to HTAP_v3.1 shows large differences, especially when comparing the energy, fugitives, solvents, road transport, and residential sectors. NEI is more regionally representative of the United States, so taking these differences into account would provide different results in the model simulations.

Naming Convention		**Emission (in kt)**		
CAMS	***HTAP***	**CAMSv5.1 (2021)**	**CAMSv5.1 (2020)**	**HTAP_v3.1 (2020)**
ene	Energy	5.223	5.254	0.327
ind	Industry	7.683	7.715	4.484
fef	Fugitives	9.557	9.651	7.241
slv	Solvents	15.548	15.529	29.011
tro	Road Transport	22.294	23.23	9.197
tnr	Other Ground Transport	0.365	0.39	1.042
res	Residential	1.856	1.887	12.002
swd	Waste	0.004	0.004	1.672
awb	Agricultural Waste Burning	1.007	1.005	0.102
agl	Agriculture Livestock	1.898	1.889	2.062
-	Agriculture Crops	-	-	3.274
shp	International Shipping	0.025	0.025	0
	Domestic Shipping			0.023
SUM		65.46	66.579	70.437

Crippa, M., Guizzardi, D., Butler, T., Keating, T., Wu, R., Kaminski, J., Kuenen, J., Kurokawa, J., Chatani, S., Morikawa, T., Pouliot, G., Racine, J., Moran, M. D., Klimont, Z., Manseau, P. M., Mashayekhi, R., Henderson, B. H., Smith, S. J., Suchyta, H., Muntean, M., Solazzo, E., Banja, M., Schaaf, E., Pagani, F., Woo, J.-H., Kim, J., Monforti-Ferrario, F., Pisoni, E., Zhang, J., Niemi, D., Sassi, M., Ansari, T., and Foley, K.: The HTAP_v3.1 emission mosaic: merging regional and global monthly emissions (2000–2018) to support air quality modelling and policies, Earth Syst. Sci. Data, 15, 2667–2694, doi:10.5194/essd-15-2667-2023, 2023.

Section 3.4:

- The discussion on evaluation of modeled HCHO columns contradicts the conclusion in Section 3.3: the authors say there is a combined effect of grid resolution and application NO diurnal cycle on HCHO (Line 429), whereas Sect. 3.3 states no obvious impact of NO on HCHO in the model (Line 383).

Response: This has been corrected in the main text as "The differences in grid resolution are seen more strongly than the inclusion of diurnal NO emissions for HCHO concentrations in **Fig. 6b**" in Section 3.3 and the removal of "Because HCHO does not have an obvious diurnal cycle and is different from NO_2, performance for HCHO columns was much more dependent on the combined effect of grid resolution and the application of the diurnal cycle for anthropogenic NO" in Section 3.4.

- The section could be improved by discussing the link between the location of the stations/sites and the changes in HCHO (e.g. induced impact from isoprene emissions under different Nox-regimes).

Response: The locations are relatively close to each other, in urban/near-urban areas surrounded by industry. Additional text has been added in Section 3.4 as so:

- "We compare NO_2 and HCHO tropospheric columns from two Pandora spectrometers to the four MUSICAv0 simulations. Both Pandora monitoring sites (SWDetroitMI and DearbornMI) were

273 located in an industrial and high-traffic setting, providing continuous observations in urban
274 conditions and complementing the other observations"
275 • "The locations of the Pandora spectrometers are in highly industrialized, urban areas. The large
276 model bias in HCHO columns could be an indication of missing emission sources in the area."
277

278 - Like for other Sect. 3 subsections, it would be useful to include the location of the monitoring sites
279 and link the results with those discussed in Sect. 4.

280 Response: A map that includes the Pandora sites shown for this evaluation can be found in Fig. 2 of the
281 manuscript (can also be seen in the response for Section 3.2 above). A stronger link between Sec. 3 and
282 4 has been included in the main text. We've added a transition paragraph at the end of Section 3, which
283 is shown below.
284 "Section 3 has evaluated the model simulations against four different types of observations obtained
285 during MOOSE 2021. Taken together, the model evaluation shows (i) that refining the horizontal grid
286 resolution in the model is the dominant factor leading to reductions in bias for peak O_3 concentrations,
287 enhances NO_2 source region plumes, and better separates contrast between urban and suburban locations,
288 such as Allen Park and Trinity St. Marks; (ii) that the diurnal cycle for anthropogenic NO emissions
289 corrects the early morning biases in NO_2 and slightly impacts O_3, while having small impacts on peak O_3
290 values; and (iii) the high biases in VOCs points to deficiencies in the emission inventory rather than grid
291 resolution and temporal allocation. These findings motivate the more in-depth analysis described in Sec.
292 4, where we discuss resolution- and diurnal emission-driven changes governing O_3 production and loss
293 across SEMI."
294

295 **Section 3.5:**

296 - Surface maps for winds, temperature and other meteorological parameters could be added to the
297 Supplement to better understand the conditions during the analyzed days and times.

We included a table with detailed information of each GCAS raster on the flight days during MOOSE 2021 in the supplemental information as Table S8. We have also included maps during the GCAS flights of temperature and winds in the supplemental information as Figures S16-S31.

Section 4:

- The significant changes in isoprene emissions from MEGANv2.1 depending on the grid resolution is linked to the induced changes in meteorological parameters. This needs to be supported by maps of meteorological fields showing these changes.

Response: We thank the reviewer for this suggestion. We have incorporated a couple of sentences referencing Figures 3 and S16-S31, where different meteorological parameters are shown. We have noted some of the differences in relation to how they can impact isoprene. This is noted in Section 4 as: "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."

- The link between Sect 4. and Sect 3. should be strengthened in either or both sections to better understand what drives the changes in the different sites, locations, etc.

Response: We have strengthened the link between sections 3 and 4 to better understand the changes driving O3 chemistry in the region. We have added a transition paragraph at the end of Sec. 3, and an updated introduction for Sec. 4. We have also linked some of the findings in Sec 4. with what was found in Sec. 3. Line 612 is an example of this: "This difference results in an improvement for the ne30x16 simulations based on the findings in Fig. 4, where peak O_3 performed best in the finer resolution simulations when compared to the surface sites."

- The discussion section is rather short and could/should be improved by strong arguments on e.g. what controls O3 in different parts of the SEMI region and what mitigation strategies could be adopted to reduce the pollution.

Response: We have elaborated further on what is controlling O3 in diverse locations across SEMI and incorporated some potential mitigation strategies, as well as future work using this modeling framework at the end of Sec. 4. The added contents in the updated manuscript are shown as follows.

"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, was 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 were 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, it is necessary to reduce emissions of VOCs as well as NOx to avoid increasing O_3 concentrations. Thus, targeting reductions in VOCs, such as those from the industrial sectors, is crucial. 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 profiles 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."