Response to Reviewer #1

Thank you for your careful and thorough reading of this manuscript and your thoughtful comments and suggestions. Our responses follow your comments (in *Italics*).

This manuscript reports the extensive comparison of the REAM chemical transport model (CTM) simulations with the NOx and NO_y observations acquired during the DISCOVER-AQ 2011 over the Baltimore and Washington-DC area. The observations include the data from surface monitors, PANDORA, P3 aircraft, ACAM, and satellites OMI and GOME-2. The model results with two spatial resolutions, 36 km and 4 km are compared in order to elucidate the impact of the resolutions on the model NOx and NOy simulations. Differences between the model and observations are discussed in details and causes for the discrepancies are suggested.

The manuscript reflects the extensive works dealing with almost all available data sets to evaluate NO₂ measurements and CTM results over the Baltimore and Washington-DC area for July 2011. I appreciate the efforts the authors made for this study. The manuscript will be more valuable if quality of presentation and interpretation of the results are enhanced.

Reply:

Thank you for your suggestions and comments. We have made several major revisions to the manuscript based on the suggestions by you and another reviewer.

- We have updated our WRF and REAM simulations using WSM6 (WRF Single-Moment 6class scheme) instead of WSM3, as listed in Table S2 (Line 34) in the revised supplemental table file. We also have downloaded the updated DISCOVER-AQ 2011 P-3B observations. All relevant results, including figures and tables, have been updated accordingly. The new results are almost the same as before except for some minor differences. The changes have no impact on our conclusions. The WSM3 results are now used as a sensitivity test (Lines 626 – 632) to confirm the reliability of our results and conclusions.
- 2) We have deleted the discussion on the reliability of 36-km NO_x emissions and total NO_x emission amount in the DISCOVER-AQ region but added an analysis of the 36-km NO_x emission distribution issue. So the current manuscript just focuses on the distribution issue but does not include any judgment on the total NO_x emission amount. Please see Lines 43 47, 125 126, 666, 718 730, 732 733, 753 789, 794, 797 806, 850 857, and 1385 1390 (Figure 15) in the revised main manuscript and Lines 281 288 (Figure S27) in the revised supplemental figure file.
- 3) We have added individual site comparisons in the supplemental figure file (Figures S19 S23, Lines 242 260) to demonstrate the NO_x emission distribution issue. Please see Lines 37, 40 43, 668 678, 814, 829 831, and 837 848 in the revised main manuscript.
- We have added some more detailed explanations for the Pandora issue in the late afternoon and early morning. Please see Lines 536 – 544 in the revised main manuscript and Figure S13 (Lines 185 – 190) in the revised supplemental figure file.
- 5) We have moved the evaluation of WRF meteorological fields to a new section 3.1 and added the evaluation of vertical profiles for several meteorological variables in the new Figure S6. Please see Lines 158 183 and 339 366 in the revised main manuscript and Figure S4 S8 (Lines 108 146) in the revised supplemental figure file.
- 6) We have used stricter and more consistent criteria to filter out invalid satellite NO₂ TVCDs (Lines 205 206 and 224 225). The GOME-2A morning high bias is gone, as shown in

Figure 10 in the revised main manuscript (Lines 1344 - 1353). Relevant changes in the text are in Lines 564 - 567, 818, and 823 - 826.

Detailed responses are as follows.

The main focus of the paper seems to be the comparison of the model simulations with the 36 km and 4 km resolution and advocate the use of 36 km in the end. I think the authors should focus more on the analysis of 4 km resolution results and causes for the similarities and discrepancies with various observations. The emissions at 36 km resolution are simply accumulations of the emissions at 4 km. It is not important to compare the emissions at the two resolutions and judge which one is better. The authors have the best spatial resolution of emission inventory data and the model simulations at the comparable scale (4 km). If the model overestimates the NOx, NOy observations at one height or vertically column integrated, that simply means the model emissions are overestimated. For the pollution hot spots in the domain, the model values are higher than the observations (judging from the ACAM data). This may be about the spatial location error in the NEI as the authors jumped to the conclusions, but it is more probable that the uncertainties in the emission factors (or activities) over populated urban or roads as represented as MOVES caused the problem. Section 3.7 should be deleted or rewritten. This section is confusing and misleading.

Reply:

Thank you for your suggestions and comments. As we stated in the original manuscript, the model results between 4 and 36 km resolutions are different in comparison to the observations. Modeling with a higher spatial resolution does not necessarily improve model simulations. In any research that finds superior modeling with a higher resolution, the change of resolution is usually a minor reason; the better representation of physical and dynamical processes at a higher resolution is usually more important in atmospheric models. Therefore, it is scientifically important to compare model simulations in two or more resolutions if possible, as we did in this study. We did not "advocate" the use of a lower resolution model. What we suggested is that the 4-km emission distribution of NO_x emissions causes model errors in our evaluation using DISCOVER-AQ measurements, and it needs to be improved. We believe that we are in agreement with the reviewer on the importance of improving high-resolution emission inventories for NO_x and other pollutants. For users of the emission inventories, errors in MOVES are part of the distribution errors in the NEI. Very few ACP readers understand the details of MOVES. To identify the issues and uncertainties in MOVES, one will have to write another paper. The reviewer appeared to misunderstand our intentions.

This paper is not meant to promote lower-resolution air quality modeling. We want to understand the reasons why the high-resolution model does not reproduce the observations. To illustrate the potential NO_x emission distribution issue between 36-km and 4-km resolutions, we took a step by step approach. We first discussed the reliability and possible limitations of the 4km REAM (sections 3.1 - 3.5) and then identified NO_x emissions and gradients as a major factor causing the discrepancies among the 36-km REAM, the 4-km REAM, and observations through comprehensive evaluations and diagnostics of NO_x related chemistry and physics (sections 3.6). Next, through individual site comparisons, we found that the performances of the 36- and 4-km REAM simulations depend upon the observation locations. A uniform underestimation or overestimation of NO_x emissions cannot explain all the model biases, and there may be some distribution biases for the NEI NO_x emissions. Finally, we verified the potential distribution biases of NEI NO_x emissions at both 36- and 4-km resolutions by comparing NO₂ VCD distributions from OMI, GOME-2A, and ACAM with those from 36- and 4-km REAM simulations. This structure aims to make the manuscript reasonable and understandable.

In the old manuscript, we only discussed the distribution bias of NEI2011 NO_x emission distributions at high resolution, but it doesn't mean that we advocated using 36 km. What we really wanted to say was that the total NO_x emissions might be reasonable in the DISCOVER-AQ 2011 region since we defined the DISCOVER-AQ 2011 region as the six selected 36-km grid cells. And we also emphasized that our conclusion on the total NO_x emissions was only valid in the DISCOVER-AQ 2011 region, considering the significant spatial heterogeneity of NO_x emissions. In the new manuscript, we have deleted our judgments and discussions on the total NO_x emissions due to the reviewer's strong objection and the relatively small size of the DISCOVER-AQ region. We have added some discussion on the NO_x emission distribution issue at 36-km resolution to make the paper more balanced, which we think is what the reviewer asked for. The distribution issue is the most critical point we want to emphasize in this study.

The 36-km NEI emissions are indeed the sum of the 4-km NEI emissions. But the results from the 4-km REAM can differ significantly from the 36-km run because of nonlinear processes such as oxidation chemistry. Without comparisons between 36-km and 4-km REAM simulations, we would not know whether an issue only exists in 4-km REAM or also in coarse resolutions. Moreover, through the comparison between 36- and 4-km REAM simulations, we can derive the effects of NO_x emissions and gradient on reproducing NO₂ and NO_y observations. It would be hard to do by only using the 4-km REAM simulation results, as NO_x gradient-associated transport is hard to exclude in one simulation.

If the observations are limited, a distribution issue may be thought to be a simpler problem of overestimating or underestimating the sum of NO_x emissions. It is a reason why we used as many observations as possible in the DISCOVER-AQ experiment. The intensive field measurements make it possible to analyze the issue more in depth. Please also note some previous studies only focus on polluted urban areas, while the areas of study are much broader in this work. We have used more comprehensive observations than previous studies to show the distribution bias through our diagnostics and analyses. Even in the high-emission pixels (NO_x emissions > domain mean) in ACAM, as shown in Figure R1b, overestimation and underestimation of NO₂ VCDs coexist. The new content we have added in the revised manuscript, such as individual site comparisons and distribution uncertainties at 36-km resolution, can further show the potential distribution bias of the NEI NO_x emissions.

We have had working experience with MOVES. The setup of MOVES is very complex and involved a lot of observations and variables. It is not just a story of emission factors or Vehicle Miles Traveled (VMT or activity). Vehicle population, ages, and types (e.g, passenger cars, passenger trucks, short-haul trucks, long-haul trucks, etc.), road types, speed distributions, etc., are all crucial variables estimating running-exhaust emissions. Not all counties provided these data to the NEI setup, where MOVES may use national defaults as inputs and, therefore, missing local characteristics. Even for those counties providing local data, it is almost impossible to make a reasonable estimate of speed distributions (and some other parameters) for different vehicle types. Not to mention large uncertainties in further allocations to 4-km grids by SMOKE (MOVES can only resolve county-scale emissions). Recent GPS-based data have shown that speed distribution in the MOVES database is not accurate and cannot represent local conditions (DenBleyker et al., 2017). We understand some tunnel and roadside experiments found the overestimation of NO_x emissions by MOVES for some vehicle types in some regions. But the

underestimation of NO_x emissions for some vehicle types by MOVES was also found in other areas (https://www.epa.gov/sites/production/files/2017-11/documents/light_duty_nox.pdf). It is noteworthy that MOVES default emission factors are also based on the observations. Instead of talking about the overestimation or underestimation of MOVES NO_x emission factors, we would like to think of the problem from the perspective of representativeness. Is current MOVES input data able to represent all local conditions over the US? Are the allocations of VMT, speed distribution, etc., accurate enough for different counties or high-resolution grids? Here we have to say that spatial distribution is a much more complex problem than the simplification of an overestimation or underestimation of total NO_x emissions. Accurate spatial distribution requires a reasonable estimate of NO_x emissions for each grid cell. Having said all these, we acknowledge the tremendous effort that went into the development of MOVES and similar programs and that they have been and continue being indispensable for air quality research.

Some readers may think negatively about the NEI NO_x inventory because of the issues we find. However, most scientists we trust understand the important contributions of the NEI NO_x emissions and our intention that identifying problems is the first step to improve our understanding and modeling capability.



Figure R1. (a – b) Distribution of relative differences $(\frac{ACAM}{REAM} - 1)$ of NO₂ VCDs between ACAM and the 4-km REAM. (a) shows the relative differences for all available pixels, while (b) only shows the relative differences for high-NO_x emission pixels. Here, we define high-NO_x emission pixels as those pixels with NO_x emissions larger than the domain average. (c – d) Distributions of scaled NO_x emissions. Here, we scale NO_x emissions by the domain average. Similar to (b), (d) only shows high-NO_x emission pixels.

Except for Figure 10, the model results and observations were not analyzed at the measurement sites. The plots are all averages for large domains and many sites. As the ACAM data demonstrate, there are heterogenous distributions of NO₂ at fine scales. This is important. The model results should be compared at each site of PANDORA and P3 spiral locations.

Reply:

Thank you for your suggestion. We have added individual site comparisons in the supplemental figure file (Figures S19 - S23, Lines 242 - 260) to demonstrate the NO_x emission distribution

issue. Relevant text changes can be found in Lines 37, 40 - 43, 668 - 678, 814, 829 - 831, and 837 - 848.

Figure 2 to 5 (and Figure 9) are about the surface monitor data and interpretation. There are many other interesting, important data sets from the DISCOVER-AQ campaign, which is discussed in short compared to the surface routine monitors.

Reply:

Figures 4 – 6 (the old Figures 2 – 4) are used for different purposes from Figures 7 and 12 (the old figures 5 and 9). Figure 4 is about the scaling of measure NO_2 data due to instrument biases (from the conversion of other reactive nitrogen to NO), which is necessary for model evaluation. Figures 5 and 6 are mainly about the underestimated nighttime vertical mixing, which needs to be corrected before we use the model to understand the diurnal variation of NO_x . Only Figures 7 and 12 show the comparisons between the 36- and 4-km REAM simulations, one for NO_2 , and the other for NO_y . The descriptions related to these two figures are shorter than those for vertical profiles and NO_2 TVCDs.

The interpretation of nighttime PBL height (or PBLH as in the model output name) may be right, or may be wrong. As authors mentioned in the manuscript, this may be due to overestimation of nighttime emissions, which can not be ruled out. People do not know much about nighttime PBL height and nighttime emissions. PBLH in the model output is not simply PBL height during nighttime. The nighttime PBL height from YSU scheme is sometimes recalculated based on many other nocturnal PBL height definition. Thus Figure 4 may need to be carefully revised or explain limitation of this analysis.

Reply:

Thank you for your suggestion. What we use from the YSU scheme is the vertical diffusion coefficient, not the diagnosed PBLH. So the mixed layer height in this paper is a true mixing layer in the REAM model. It is what ELF measured as well and can therefore be compared to the ELF measurement data. We do not use the "PBLH" data in the WRF output file, which seem to be what the reviewer meant. The vertical mixing is computed using Equation R1.

$$\frac{dc}{dt} = -K_{zz} \frac{d^2c}{dz^2}$$

(R1)

Now the question is how to determine mixing depth or mixing height based on k_{zz} , so that we can evaluate it with the ELF observations.

WRF-YSU k_{zz} has some background or default values associated with the depth of each level following Equation R2 (Hong et al., 2006).

$$K_{zz \, background} = 0.001 \times \Delta h$$

(R2)

Here, it is noteworthy that k_{zz} is on the edge of each level, so Δh is the depth (m) between the middle point of the below-layer and the central point of the above-layer. We investigated the WRF-simulated k_{zz} values carefully, and they indeed satisfy this equation. We determined the mixing depth by comparing k_{zz} with k_{zz} -background, which is the k_{zz} -determined PBLH in the manuscript. Figure R2 shows the diurnal cycles of WRF-YSU outputted "PBLH", k_{zz} -determined PBLH, and the ELF mixing depth at the UMBC site in July 2011. The YSU

outputted "PBLH" is generally consistent with k_{zz} -determined PBLH, confirming the reliability of our method to determine mixing depth by comparing k_{zz} and its corresponding background values. That's why we used mixing height as the y-axis of Figure 6 in the revised manuscript. Here, we want to emphasize that the background k_{zz} values are very small, and our method to determine mixing depth by using k_{zz} should give an upper bound of the mixing depth estimates. However, Figure R2 and Figure 6 in the revised main manuscript still show a significant underestimation of k_{zz} -determined PBLHs compared to the ELF observations. We have added some brief explanations about k_{zz} -determined PBLH in Lines 387 – 388 in the revised manuscript.

We agree with you that there may be an overestimation or underestimation for nighttime NO_x emissions, which we cannot determine quantitatively based on our available observations. Most NO_2/NO_y observations in our study are in the daytime, and nighttime surface NO_2/NO_y concentrations are sensitive to vertical mixing. Since we have confirmed the underestimation of vertical diffusion in the late afternoon and nighttime compared to the ELF observations and increasing nighttime k_{zz} improved REAM simulation results, it is unreasonable to attribute the original REAM model bias to the speculation of overestimation of nighttime NO_x emissions. In addition, we did not state that underestimated nighttime vertical mixing is the only reason and can solve the nighttime model bias completely in the manuscript.



Figure R2. ELF observed and model simulated diurnal variations of PBLH at the UMBC site during the Discover-AQ campaign. "ELF" denotes ELF derived PBLHs by using the covariance wavelet transform method. "YSU_PBLH" denotes the 36-km WRF-YSU outputted "PBLH", and " k_{zz} -derived" denotes k_{zz} -determined PBLH by comparing k_{zz} with its background values. Vertical bars denote standard deviations.

Figure 7 is too busy. It is difficult see the details. More expansion of analysis in Figure 7 or a summary in Table would be helpful. A plot comparing satellite NO₂ spatial distributions and more presentation and discussions on the GMI, TM4, REAM as a priori for the retrieval would be useful.

Reply:

Thank you for your suggestion. We have added Table S3 in the revised supplemental table file (Lines 36 - 40) to summarize NO₂ TVCDs at 9:30 and 13:00 LT for all datasets and simulations. We also have added the comparisons of NO₂ TVCD distributions among different retrievals in Figure 15 (Lines 1385 - 1390) in the revised main manuscript and Figure S27 (Lines 281 - 288)

in the revised supplemental figure file. We have added descriptions about the comparison of NO_2 TVCD distributions among different datasets in Lines 718 – 730 in the revised main manuscript.

Authors frequently use the figures in Supplementary Material. It is difficult to read the manuscript with many supplementary figures. Because there are important plots in the supplementary, I suggest to move some of the plots in the supplementary to the main manuscript. For example, Figure S1, S2, S17, S21, S22, S23 and discussions about them would be useful. Differences between the model and ACAM NO2, differences between two ACAM NO2 retrievals, and differences between weekdays and weekends are interesting and can have important implications for emissions and model assessments. Figure S12, S13, and S19 or one of them can be also shown in the main text. For S19, one-to-one comparison of the model simulations at 36 km and 4km resolution would be more useful. It is difficult to understand the purpose of Figure 8.

Reply:

Thank you for your suggestion. We have moved the old Figures S1, S2, and S17 to the revised main manuscript (Figures 1, 2, and 13). The old Figures S21, S22, and S23 are similar to Figure 14 (old Figure 10) in the revised main manuscript and used for the same purpose. We don't think it is necessary to move them to the main manuscript. The Harvard team (Drs. Liu and Nowlan) will submit a separate paper on their ACAM dataset. That's why we put it in the supplement. The ACAM datasets are just used to show the model distribution bias. The weekday and weekend difference can be easily identified by comparing Figure 14 in the revised main manuscript and Figure S24 in the revised supplemental figure file as well as their domain averages. It is unnecessary to mention it again. Furthermore, we show the relative difference between weekday and weekend ACAM NO₂ VCDs below (Figure R3).

We have moved the old Figure S13 to the revised main manuscript as Figure 9. We don't understand the purpose of making a one-to-one comparison for Figure S17 (the old Figure S19) in Lines 632 - 644 in the revised main manuscript. Figure S17 is used to verify the transport effect, so we need sites with comparable NO_x emissions between 36-km and 4-km REAM simulations to exclude the impact of chemistry and NO_x emissions. Anyway, we show the one-to-one comparisons below (Figures R4 – R9) if the reviewer is interested in them. The purpose of Figure 11 (the old Figure 8) is to confirm further that the REAM simulations well capture the daytime variations of vertical mixing, chemistry, and transport since the VCDs at different height bins are mainly affected by these three factors besides emissions.



Figure R3. Distributions of relative differences $\left(\frac{weekend}{weekday} - 1\right)$ between weekday and weekend ACAM NO₂ VCDs.



Figure R4. The same as Figure S17 in the revised supplemental figure file but for Padonia.



Figure R5. The same as Figure R4 but for Fairhill.



Figure R6. The same as Figure R4 but for Essex.



Figure R7. The same as Figure R4 but for Edgewood.



Figure R8. The same as Figure R4 but for Beltsville.



Figure R9. The same as Figure R4 but for Aldino.

Regarding WRF model options, I am wondering why Single Moment 3 Class microphysics scheme is used. This scheme is for warm clouds.

Reply:

Just to be clear, the WSM3 scheme predicts three categories of hydrometeors: vapor, cloud water/ice, and rain/snow, considering ice processes below 0 °C. The WSM3 scheme is not always worse than WSM5 or WSM6 in predicting precipitation. Anyway, we have changed our study from WSM3 to WSM6. All relevant figures, tables, and results are adjusted accordingly. The current results based on WSM6 are very similar to those found on WSM3. The change has no impact on our conclusions. And the WSM3 results are used as a sensitivity test to verify our current results with WSM6.

There are Grell ensemble or other Grell cumulus parameterization options that were widely tested in CTM groups. Because the REAM model is an offline model, the performance of model at 4 km resolution may not be caused by original WRF physics, but by the integrator between the WRF and REAM. The performance of model at 12 km was not discussed, but on/off of cumulus parameterization option at this resolution may be another factor to be tested.

Reply:

The KF scheme is also widely tested and evaluated. There is not a general preference for Grell schemes over the KF schemes. The limitation of the KF (new eta) scheme (Table S2 in the revised supplemental table file) is that it is not designed for high-resolution simulations. We understand that some Grell schemes can be used for high-resolution simulations. The new updated Multi-scale Kain-Fritsch scheme can also be applied to high-resolution simulations. However, the Grell schemes and the Multi-scale KF scheme are not compatible with REAM now. The convection module in REAM needs intermediate variables from the WRF cumulus scheme. We have made a test with the KF (new eta) scheme turned on in the nested 4-km domain. We indeed found an increase in total precipitation in the DISCOVER-AQ region but not so much. And it reduces the afternoon vertical mixing very slightly, as shown below (Figure R10), with minimal larger NO₂ concentrations in lower levels and marginally lower NO₂ concentrations in higher levels. However, it doesn't mean that the inclusion of convection parameterization in the nested 4-km. Further studies by using more appropriate Grell schemes or the Multi-scale KF scheme are necessary to derive reliable conclusions.

There are only three processes transporting surface NO₂ to higher layers in REAM — vertical diffusion, vertical advection, and convection (related to cumulus parameterization). Convection and vertical advection are generally minimal. Moreover, in the 4-km REAM, convection is inactive as cumulus parametrization is turned off in the 4-km domain for the nested 4-km WRF simulation. Our sensitivity test with vertical diffusion turned off still shows fully mixed NO₂ vertical profiles in the boundary layer. The only reason is the overestimation of vertical advection (*w*), consistent with the larger *w* in nested 4-km WRF simulation than the 36-km WRF simulation, as shown in Figure S9 in the revised supplemental figure file. We replicate the convective transport of the KF scheme in REAM (which is the reason that updating the convective transport using a different scheme is complicated in REAM); it is not an "integrator". The advection module is based on the widely used Walcek scheme, not involving any complex equations. If it has any problems, it cannot be only problematic in the 4-km but not 36-km REAM simulation.

As we mentioned above, the goal of our study is to emphasize the potential distribution bias of NO_x emissions. Current comparisons between the 36- and 4-km REAM simulations can provide enough evidence to support what we want to show. Including the evaluation of 12-km simulation results does not add to the content of this work since the 12-km results should be within the range between the 36-km and 4-km results.



Figure R10. Comparison of afternoon (15:00 - 17:00 LT) NO₂ vertical profiles among different 4-km REAM simulations and P-3B observations on weekdays and weekends for the DISCOVER-AQ 2011 campaign. "cu" (blue lines) denotes the 4-km REAM simulation constrained by the nested 4-km WRF simulation with the KF scheme turned on in the 4-km domain. In contrast, "no cu" denotes the 4-km REAM simulation constrained by the nested 4-km WRF scheme turned off in the 4-km domain. "obs" refers to P-3B aircraft observations.

For the analysis like Figure 6, the model temperature (potential temperature), moisture (specific humidity), U, and V also need to be analyzed with observations, particularly for afternoon.

Reply:

As discussed above, vertical advection (*w*) is the only factor causing the overestimated vertical mixing in the afternoon in 4-km REAM. We don't understand the purpose of comparing temperature, moisture, U-wind, and V-wind. We have added the comparison of the diurnal

variations of vertical profiles for temperature, potential temperature, relative humidity, U, and V among the 36-km WRF, the nested 4-km WRF, and P-3B observations in Figure S6 in the revised supplemental figure file. Relevant text updates can be found in Lines 347 - 353 and 362 - 363 in the revised main manuscript.

One minor point is frequent use of red/green combinations in the plots, which is not ideal.

Reply:

We have changed some green lines to blue. Please see the new Figures 5, 6, 7, 11, and 12 (the old figures 3, 4, 5, 8, and 9) in the revised main manuscript.

If the manuscript is revised following the comments above, I think publication can be reconsidered.

Reply:

Thank you for being open minded.

References

DenBleyker, A., Koupal, J., DeFries, T., and Palacios, C.: Improvement of Default Inputs for MOVES and SMOKE-MOVES: CRC Project A-100, available at <u>https://crcao.org/reports/recentstudies2017/A-</u> <u>100/ERG_FinalReport_CRCA100_28Feb2017.pdf</u>, Eastern Research Group, Inc., Austin, TX, 86, 2017.

Hong, S.-Y., Noh, Y., and Dudhia, J.: A new vertical diffusion package with an explicit treatment of entrainment processes, Monthly weather review, 134, 2318-2341, <u>https://doi.org/10.1175/MWR3199.1</u>, 2006.