RC1

All typo and grammatical errors were adjusted according to the comments, the suggested citations were added. The responses to other comments are included below.

1. "… the authors themselves point out on page 4 (lines 11-15) the "unsatisfactory" nature of the 2020 paper's approach so why is it justified to separately publish that?", "It is of critical importance to address the significant overlap between this work and the cited companion paper of Fang & Michalski (2020) …"

We decided to combine the companion paper with this manuscript in order to deal with the overlapping and completeness issue.

2. "… While I value the need to focus on the detailing the model specifics in a journal such as GMD, the manuscript here lacks any real interpretation, quantification of the sensitivity of the output to the model parameters and consideration of the implications of the predicted values compared to previous studies in the literature of interpreting the isotopic composition of NOx and nitrate …"

The more in detail interpretation and quantification of the output have been included in the revised manuscript (In section 3.2, 3.3, 3.5, and 3.7)

3. "Second, the model is compared with one set of observations of "d15N-NOx" in Indiana (within the domain of the model runs) …"

The full simulation domain covers the whole Midwest while the sampling sites only locate in IN, IL, OH, and KY. As a result, the sampling site would have sufficient distance between the domain boundary to eliminate the bias near the domain boundary.

4. "Additionally, the measurements that are compared with are specifically d15N-NO2 and not d15N-NOx …"

Walters et al (2018) did include the d15N(NOx) values based on the measurements in Table 1 and 3.

"The first comparison is to the only direct measurements within the domain, which occurred in West Lafayette, IN. The δ15N(NOx) values were inferred from the measured δ15N(NO2) and the calculated δ15N(NO2) shift (Walters, Fang, & Michalski, 2018)."

5. "Third, the sensitivity to the starting emissions values should be evaluated …"

The range of d15N values for any source is generally a function of equilibrium, kinetics, or reaction progress happening in that source process. For example, automobiles show a wide range of both NOx amount and d15N values going from cold start to normal driving, but once the catalytic converter is warm the values are relatively constant because the NOx reduction by the CC becomes constant. We are using the average to account for these effects and for simplicity.

In future work hope to explicating model the sources variation in SMOKE or land surface models, but that is well beyond the scope of this work. Fig. S17 shows the uncertainties of d15N values within the research area. For most of the grids, the uncertainties are less than 5 ‰, which is well below the difference in d15N values between any two of the emission sources. For those regions dominated by biogenic source, the uncertainties in d15N values are less than 10 ‰, which is also significantly below the difference in d15N values between the emission from biogenic source and all the other sources.

6. “Fourth, one of the key conclusions of this work is that changes in the polluted boundary layer (PBL) are critical to transport and dispersion of NOx such that the pattern of d15N- NOx is importantly changed based on the PBL height. I’m not convinced the results shown support this conclusion …”

The more in detail interpretation and quantification of the relationship between d15N and PBL height has been included in section 3.3 of the revised manuscript

7. “Fifth, “the role of deposition” section and comparison of d15N-NOx with d15N-NO3- seems out of place in this work …”

CMAQ simulated the d15NOx effect by NOx removal using enhanced deposition. These “emission + mixing + enhanced deposition” simulations were not imposing an isotope effect related to dry/wet deposition, rather they are an attempt to show how “lifetime chemistry” alters NOx d15N values by removing NOx before it can be transported significant distances.

8. “Finally, it needs to be addressed why in this work there are only 8 NADP sites being compared with, while it appears that 82 measurement sites are included in FM2020?”

We decided to use the exact measurement from our lab at the 8 NADP site to validate the simulation, instead of using the values of literature review and compared with the simulation values at the grids contains the NADP site within the simulation domain.

9. “Title: Is it necessary to have the CMAQ, SMOKE and WRF versions as part of the title?”

The editor requested us to include the model’s name and the version number in the title

10. “I would argue that atmospheric “processes” are not really being tested here, it’s really transport or meteorology …”

Transport, mixing, dispersion, and deposition are all examples of atmospheric processes

RC2

1. “I agree with the first reviewer about the scope of this paper being too narrow, and I think that reviewer’s suggestion of combining this work with Fang and Michalski (2020) would be appropriate ...", "I have serious concerns about the overlap in content with Fang and Michalski (2020). "
We decided to combine the companion paper with this manuscript in order to deal with the overlapping and completeness issue.

2. "I am not sure what we are learning from the many figures showing the seasonal variation in δ15N concentrations for transport only, transport with different emissions, transport with different meteorology, and transport with deposition on, etc ..."
The more organized interpretation and quantification of the output have been included in the revised manuscript in section 3

3. "The conclusion that the PBL is the “key driver for the mixture of anthropogenic and natural NOx emission” seems odd ..."
The more in detail interpretation and quantification of the output have been included in the revised manuscript in section 3.3

4. "Please add quantitative metrics to the abstract to more precisely communicate the impact that adding CMAQ’s process-level understanding has on the evaluations in Indiana."
Confirmed

5. "Page 2, Line 31: Better add the reference to the FIVE mobile emission model (McDonald et al., 2018: https://doi.org/10.1021/acs.est.8b00778) to these references."
Confirmed

6. Page 3, Line 18: Consider changing “NOx/NOy” to “NOx” or “NOx and NOy”
Confirmed

7. "Section 2.1: Why run with an extracted domain? Was this just to make the model go faster?"
To eliminate the bias near the domain boundary

8. "Much of section 2.3 can be moved to SI. The main manuscript can just state what models and version numbers were used for each part."
Confirmed

9. "Page 10, Lines 3-18: It is hard to understand exactly what was accomplished in the deposition velocity tuning approach and what its limitations are. This would all be solved if the authors were able to include chemistry in the study and turn chemistry off for a transport+deposition only case."
Including chemistry in the study and turn chemistry off is what exactly we did. The more in detail explanation of deposition have been included in the revised manuscript in section 3.7.

10. "Section 2.6: We need some idea of how the emission datasets performed against coincident observations from routine networks for conventional pollutants like NO2, EC, O3 and particulate Sulfate to check that they were processed with reasonable assumptions."
We decide to give up this section, and explore the sensitivity of emission dataset in future work

11. "Please consider removing Tables S1 and S2 from the supplemental information. Just refer readers to the MCIP user guide."
Table S1 and S2 were summarized from user's guide, it is easier to understand for the reader and more relevant to the corresponding section in the manuscript.

12. "Section 2.7: I'm not sure what is meant by the 'research area' and 'emission-free zone'. Is it just U.S. versus Canada? The term 'nested' usually refers to an area of higher resolution. Although this doesn't strictly have to be the case, I urge the authors to consider renaming their 'nested' grid..."
We do not have emission dataset in Canada, thus when the air mass transports out of the Midwest, the atmospheric NOx is diluted. In addition, we set atmospheric NOx δ15N = 0‰ for initial condition and boundary condition, the air mass from Canada, or boundary of the domain could flattened the atmospheric δ15N(NOx). Details in section 2.2.2-2.2.4. The term 'nested' was changed to 'extracted'

13. "Page 14: Rather than using Fig. 3 to show the expected dispersion of NOx in the model domain, why not include a figure as a barplot that quantifies the differences in weighted average δ15N values for the different categories discussed like agricultural areas, big cities, highways and EGUs?"
Fig. S6 in the revised manuscript

14. "Page 14, line 28: Consider adding a figure with distance from power plant on the x and δ15N on the y to show the decay along a couple of trajectories from an important facility."
Fig. 6 in the revised manuscript

15. "Page 18: Is there any data to evaluate the PBL heights?...", "Page 18, line 12: Why not have a figure showing the positive correlation between PBL height and δ15N ..."
Fig. 9 and Fig. 10 in the revised manuscript

16. Fig 7 can go to the SI.
Confirmed

17. "Fig. 10 is not terribly informative other than to show that the southern boundary should probably also be restricted for the nested area."
The purpose of this analysis is showing how the simulation over the extracted domain improve the performance (by 1‰ near the southern boundary)

18. I recommend moving Section 3.7 to the first section of the Results.
We decide to start with the interpretation and quantification of the simulation first, then comparing with the measurements.

19. "Strongly recommend putting Figs. 11 and 12 together so that measurements and models are on the same figure. Why not pair the model and obs in time for the figure ..."
Fig. 11 and Fig. 12 shows the characteristic of measurements and simulations, the comparison between them are in the next figure and the associated paragraphs in the manuscript

20. "Fig. 14: Consider normalizing both panels to remove influence of chemistry bias. It seems like there is a signal here that the CMAQ modeling is able to capture, but it is hard to tell."
We do agree the normalization could make the figure looks more beautiful. However, the similar monthly variation and seasonal trend with the gap between the simulated d15N(NOx) and measured d15N(NO3-) is exactly what we are expecting, since there are isotope effects associated with the photochemical transformation of NOx into NOy. After including the 15N into the chemical mechanism in our future work, this gap will be resolved.