Fang and Michalski have put forth a lot of hard work paving the way for $\delta^{15}N$ values to be modeled correctly for regional-scale evaluation of NO_x emissions, chemical mechanisms, atmospheric transport, and deposition rates. Based on the importance of the topic of using stable isotopes to probe our understanding of atmospheric processes and the general soundness of their approach, I am in favor of this study eventually being published in GMD. However, I think major changes are necessary before it should be accepted as detailed below.

General Comments:

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. An alternative solution would be to keep these papers separate but extend this paper to consider chemistry and aerosol processes in addition to transport and deposition. This alternative solution would allow the authors to keep the focus of each paper narrowed to the model application that is being used (i.e. SMOKE and CMAQ respectively) rather than mixing the two. There are a lot of details and useful figures presented in Fang and Michalski (2020) that warrant its own paper in my view. I suspect that the chemistry mechanism is still under development and/or characterization or it would have been included here. Unfortunately, it is hard for me to view this current paper as a complete study without that critical piece. If it can be added, I think the results shown here for the transport-only cases are still somewhat useful to present, especially in a journal like GMD. As presented though, I think the paper slices the development steps too thin to be as impactful as the authors would hope.

2. I have serious concerns about the overlap in content with Fang and Michalski (2020). Fig. 1 must be removed and the introduction before Page 3, line 14 should be rewritten to focus on issues of transport and deposition (and chemistry) if that is to be the focus of the methods and results. The reader should be referred to Fang and Michalski (2020) for a discussion of sources. Feel free to reprint the data in Fig. 1 as a table in the SI if you want it to travel with the paper, but it's inappropriate to use it again in the main text.

3. I am not sure what we are learning from the many figures showing the seasonal variation in $\delta^{15}N$ concentrations for transport only, transport with different emissions, transport with different meteorology, and transport with deposition on, etc. For example, Fig. 9 could probably be one map since there isn't much variability among seasons. The conclusion that the PBL is the "key driver for the mixture of anthropogenic and natural NO_x emission" seems odd. Are the authors highlighting PBL height to distinguish vertical dispersion from horizontal advection and deposition? If so, I would think a more systematic approach would be to show box-whisker plots for all the $\delta^{15}N$ values like in Fig. 13 but with only vertical mixing and emissions on, then with horizontal advection and dispersion (and vertical advection) added, and finally with deposition on and show which is having the largest overall impact. The authors could also add the cases with varied emissions and meteorology, but most importantly, all of these sensitivities need to be summarized visually somehow for the message to come through.

4. It seems like the measurements could be leveraged to evaluate the model more directly (see specific comments below). For example, is the model getting the day-night trend correct at West Lafayette?

5. I second the first reviewer about the grammar issues throughout the text. I am happy to provide specific comments on a future draft after the major issues are resolved or justified.

Specific Comments:

1. 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.

2. 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. It would also be good to add a more recent reference for the MOVES model from US EPA.

3. Page 3, Line 18: Consider changing "NO_x/NO_y" to "NO_x" or "NO_x and NO_y"

4. Section 2.1: Why run with an extracted domain? Was this just to make the model go faster?

5. Consider moving most of section 2.2 to SI since it is covered in the companion paper.

6. Section 2.3: Please specify model version numbers for WRF, SMOKE, and MCIP in this section. 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. The details of how the data were processed can be included in the SI, especially because I understand they probably took an enormous amount of the authors' time during this project. But as presented, I think they dilute the narrative of isotopic NO_x that the authors want to stick to.

7. 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.

8. 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.

9. Please consider removing Tables S1 and S2 from the supplemental information. Just refer readers to the MCIP user guide.

10. 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 to the 'research area' or something similar, to identify that this is the area they are using for their analysis as it is far from the interfering model boundaries.

11. Page 14: Rather than using Fig. 3 to show the expected dispersion of NO_x in the model domain, why not include a figure as a barplot that quantifies the differences in weighted average $\delta^{15}N$ values for the different categories discussed like agricultural areas, big cities, highways and EGUs?

12. Page 14, line 28: Consider adding a figure with distance from power plant on the x and $\delta^{15}N$ on the y to show the decay along a couple of trajectories from an important facility.

13. Page 18: Is there any data to evaluate the PBL heights? This is a critical part of the study the way the authors have framed it. Also which PBL model scheme in WRF and CMAQ did the authors use? Maybe they should try a different one?

14. Page 18, line 12: Why not have a figure showing the positive correlation between PBL height and $\delta^{15}N$ or whatever would correspond to the Chinese studies to demonstrate the consistency check.

15. Fig 7 can go to the SI.

16. Fig. 10 is not terribly informative other than to show that the southern boundary should probably also be restricted for the nested area. The authors should consider redefining their nested area with this in mind for all future simulations on this domain.

17. I recommend moving Section 3.7 to the first section of the Results. I find it helpful to start with the model-obs comparison and then dive into model predictions that are unconstrained by observations.

18. 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? Page 28 text should be moved to methods section. Figure 12 and associated text does not really belong in this section as it is not a model-obs comparison.

19. 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.