

Report 1

In addition to the minor comments below, I have one main reservation remaining. The language is often awkwardly phrased or unnecessarily verbose throughout the text and needs some more polishing. It doesn't interfere with communicating the meaning of the science but streamlining the details and focusing on the main points would crystallize the main messages better. I estimate 25-30% of the text could be cut down or moved to the SI. That said, I understand the authors want to move along to the full-science study with chemistry.

Adjusted accordingly

1. P2L5: "What is meant by "gasoline and diesel in vehicle emissions?" I think some words are missing from this sentence.

Changed to emissions from gasoline- and diesel-powered engine

2. P2L20-24: It's not clear from the abstract how SMOKE only can yield a prediction of concentration of isotope. Consider deleting and focusing instead on...

Rephrased

3. P4L2: Consider revising " $N_2 = 0.0036$ " to " $(^{15}N_2/^{14}N_2)_{air} = 0.0036$ ". Is that correct?

Yes

4. P5L3: Recommend rewriting: "simulations by the CMAQ (Community Multiscale Air Quality) modeling system."

Confirmed

5. P5L5: Fang et al., 2021 is not in the citations list.

Deleted, this paper hasn't been published yet.

6. P5L11: Please rewrite to "The EPA trace pollutant emissions mode..."

Confirmed

7. Fig. 2 caption: there is no light purple in t Fig. 2.

Fixed, renamed the color to "light grayish purple"

8. Equation 4: Is 't' the vehicle travel time? Please specify in the text. Does the onroad diesel use the same equation for its isotope contribution or are there different parameters? Does t vary throughout the simulation or is -2.7 assumed constant throughout? I didn't quite understand from the text.

Rephrased

9. P9L29-30: replace 'blur' with 'disperse' and 'blurring' with 'dispersion'.

Confirmed

10. P9L36-P10L6: This paragraph is a bit awkward. I recommend starting out mentioning that chemistry will be explored later. Then define what you mean by "enhanced deposition". Then mention that you assume no difference in the deposition of the isotope and the stable NO_x. The

explanation of the impact of deposition on hypothetical powerplant emissions is fine to remove, in my opinion.

Adjusted accordingly

11. Sections 2.2.1-2.2.4: These sections are organized and titled in alignment with CMAQ inputs and processes but often sensitivity runs are mentioned within these sections without being explicitly introduced. I recommend creating a section 2.2.5 that explicitly lists and describes every simulation. Also, the names of these simulations vary.

A summary table is included

Sometimes the first one is called "emission only" and sometimes it's called "no transport".

Fixed, stick to "emission only"

12. P10L32: A clean atmosphere is not necessarily ideal. Was the model provided time for 'spin-up' or initialization so that evaluation against observations would be more appropriate?

The initial concentration is based on the default setting of CMAQ, we only incorporated 15NO_x to the input files, using [15NO_x] = 0.0036[14NO_x]

13. Section 2.2.3: Please include a sentence to explain why chemistry needed to be emulated with enhance deposition. The paragraph transitions from deposition to chemistry abruptly and needs some more detail there.

Confirmed

14. P11L6: "2.2.4"? → 2.2.2

15. P11L8: This is not a complete sentence. Typo fixed

16. Fig. 3: Is this tons/day or tons N/day? The text on page 12 uses both units. I suggest sticking with one if possible. tons N/day

17. Fig. 8: I don't think the inset for Indiana is discussed. Why is it so dramatically different from the results in the same location for the larger domain?

Because the interval of the d15N values for zoom-in view is narrower. The purpose for using narrower interval is to show more details in d15N values in Indiana. We are not going to discuss the d15N values over Indiana, just to give the reader more convenience for reading this figure.

18. Fig. 16: I'm glad this comparison is here. Please make sure the x and y axes are equivalent for at least the last figure, but preferably for all of these panels. It will be interesting to see what happens with realistic deposition and chemistry.

Adjusted

19. Fig. S19: Consider promoting to the manuscript. I can't tell what symbols are the NADP values I should be looking at.

Moved the whole paragraph to supplementary

20. PBL height analysis: I'm glad this was added to the study. The discussion is rather vague

though – the authors just ascribe large isotope effects to the stronger effect of atmospheric processes. Can you please expand on this? Are these point source emissions that are being mixed back down to the surface? Or some other explanation?

Detail explanation included

21. P31L18-19: How are the authors concluding that uncertainties in the simulation are less than 5 part per mil? Do they mean variability instead of uncertainty? Please explain how uncertainty is estimated in the text.

Rephrased

Report 2

There are number of errors and a need for some better organization for the manuscript to be publishable, as these do change the scientific meaning of the results in several places. Of key importance is that authors do a better job of distinguishing NO_x versus NO versus NO₂. This needs to be made very clear upfront in the manuscript and if the authors are going to choose to use NO_x as a shorthand when referring to NO or NO₂ then that needs to be spelled out specifically. NO is the primary emission from all sources in this work, except potentially diesel emissions. This is never mentioned nor discussed and has an important context here since there are no direct observations of d15NO alone and what the model is actually simulating is the release of NO, not the release of NO_x. This should be rectified if the authors want the atmospheric chemistry community to be interested in this work.

Adjusted accordingly

Abstract -

It should be made clear in the abstract the difference between SMOKE and CMAQ (i.e. that the latter includes mixing and transport). This will help setup the reader for why these two models are used and what differences can be expected.

Fixed

Spell out NADP since this is the first use and also make clear that these are rainwater nitrate measurements.

Confirmed

Introduction-

Page 3, Line 3: The very first word of the introduction should not be a symbol – define nitrogen oxides first, then use NO_x subsequently.

Confirmed

Line 16: It seems strange to give a single percent (15%) and then give a range of values for soil NO_x emissions. It would be better to introduce the range in total NO_x emissions from global models and then point out the uncertainty range associated with individual sources such biogenic soil emissions. Also, it should be explained what soil emissions represent (ie. Nitrification and denitrification).

Fixed

Line 17-18: Satellite observations should be mentioned as reference cited in this sentence include these.

Confirmed

Line 24: change “while acting” to “versus acting” or similar, otherwise the sentence is incomplete

Confirmed

Line 40-41: Natural versus anthropogenic sources is not correct here as anthropogenic sources themselves has distinct isotopic range. I suggest you remove “natural and anthropogenic” from the sentence

Confirmed

Page 4, line 11: change “implement” to “implementation”

Confirmed

Line 20: add “species” after d15Noy

Confirmed

Figure 1 caption: CFPP needs to be defined in the figure caption.

It has been defined, coal-fired power plant (CFPP)

Page 5, line 4-7: might make sense to have this sentence be past tense since that paper is already published.

Confirmed

Throughout section 2 and its subsections there needs to be consistent use of n values, consistent use of ranges versus average values, consistent discussion of methodologies and there needs to appear a discussion of NO_x versus NO₂ versus NO. Many of the measurements are not able to be compared to one another as some measure NO_x and some measure NO₂.

Fixed

Page 7, line 1-4: Miller et al and Yu & Elliott both use active sampling techniques for collecting soil emissions. Not clear what “using a similar methodology at the end of Line 1 means, since the previous study mentioned used a passive sampling technique. Further, the passive measurements should report an n value and not a range since I believe these two numbers represent two discrete measurements that represent month long averages.

Fixed

Section 2.1.2 – it needs to be justified why Miller et al (2017) is not included. This was brought up in the previous review both reviewers. This study is an actual on-road study that captures the mix of tailpipe emissions. The Walter’s study is more representative of tailpipe type measurements, not the signal that is found on-road when vehicles are moving under real traffic conditions with a mix of vehicle types. The Miller study seems more representative in this case. If it is not, it should be reasoned why other values are being used.

Fixed

Line 22: remove “s” in this sentence

Confirmed

Page 8, section 2.1.3: this section does not make mention of the methods used, nor does it distinguish what was measured in terms of NO_x vs NO₂.

The sum of NO and NO₂

Line 19-20: how can a range of 9 to 26 lead to an average of 4.51? Please double check.

It has been written as “standard deviation”

Page 9, line 36: change the terminology of “d15NOx effect” as this is not an accurate statement. It’s really the effect on d15NOx

Confirmed

Line 37: unbold “not”

Confirmed

Page 10, line 6: change “the” to “a”

Confirmed

Page 11, line 3: change “to” to “in”

Confirmed

Page 14, line 2-3: please also define the anthropogenic fraction since that is used several times in the text below

Confirmed

Line 21: change “the clear” to “a clear”

Confirmed

Line 20-24: justify to the reader why your are selecting April to June to show here (I assume b/c this is the season when soil NOx emissions should be maximum?)

Clarified, highest variation during April to June

Page 15, Figure 5 caption: explain that the white box is defined in the Figure 6 caption.

Confirmed

Page 18, line 8: change here since April to October is not a “month”

Confirmed

Page 20, line 20: change to “a seasonal”

Confirmed

Line 22: change to “NOx was emitted”

Confirmed

Line 31: this line does not make sense, please rephrase.

Page 20, Figure 8: draw a line to make clear that the insert is Indiana (ie draw a line to the state to connect it) or label the smaller box just above as Indiana to make clear to the reader.

Confirmed

Page 21, line 18: This should start a new section as the emphasis here is no longer on seasonality and rather on the impact of different meteorological fields.

Confirmed

I would argue that Figure 9 and 10 could be combined with a different coloring or dashed lines to represent the different years of met fields.

Moved Fig. 10 to supplementary

Page 22, line 4: add “the” before simulation

Confirmed

Figure 11 is unnecessary as the text described this very well and the changes are smaller than even the analytical reproducibility of measurements so there is no way to even test where this distribution is realistic (and the boundary lines are clearly not realistic/an artifact of the model domain).

Moved to supplementary

Page 24, Section 3.5: the phrasing here would be better as NO_x loss rather than NO_x deposition. The increase in NO_x deposition is purely artificial – i.e. this is not, for instance, testing a range in the literature to look at sensitivity. It is a change in the amount of NO_x loss to simulate what happens to the isotopic composition given NO_x loss from the atmosphere.

Confirmed

Page 25, line 7: remove “of atmospheric”

Confirmed

Line 9: there is no Figure 12a. Figure 12 is not really utilized in this section and it is never explained why a particular time point in the model is chosen for this comparison. Either robustly explain the purpose of the figure and the conclusion being drawn from it or remove it.

Fixed

Line 10: change “as a similar fashion” to “in a similar fashion”

Confirmed

Line 17: change to “lead to lower”

Confirmed

Line 19-20: change to “atmospheric NO_x on a seasonal basis”

Confirmed

Line 20: the range here says a maximum of 5.34 per mil but the figure range only goes to 4 per mil?

Fixed

Line 25-27: the sentence beginning with “The enhanced deposition simulation somehow presents the isotope effects...” This sentence is not accurate. This cannot represent the fractionations associated with chemistry. What it does show should be the focus here. And it should be explained here that this is used as a proxy for NO_x loss from the atmosphere as would be the case in conversion to NO_y species. But this is not at all representative of how and why chemical processes would lead to fractionation of NO_y species.

Fixed

Page 26, line 4: change “comparing” to “compared”

Confirmed

Page 27-28, Section 3.6: the two paragraphs of this section are repetitive. This section should be re-organized. I would suggest making the section on comparison to observations its own as Section 4, etc. And the beginning of the section should explain what observations will be used for comparison purposes with the model. It should also be explained what the observations represent and how NO_x was computed from d15NO₂ measurements. It should also be explained by isotopic measurements of nitrate in rainwater from NADP sites is used as a comparison here as well. Then move onto the actual comparisons.

Fixed

Page 28, line 20: change to “based on 2002”

Confirmed

Page 29, line 1: “collect” should be “collected”

Confirmed

Line 3: change to “show similar monthly variations”

Confirmed

Line 10: change to “NO₃⁻ are about”

Confirmed

Page 30: The paragraph below Figure 16 belongs before the previous paragraph and these two paragraphs are repetitive and should be edited further.

Fix

Figure 16: It's not clear what is being gained from the Figure 16 comparison. The text focuses on discussion of how the seasonal variability is captured. Clearly the ranges are not always captured and while in some cases the variability is decently captured (i.e. $R^2 > 0.4$) we are not really learning anything by comparing the data in this way. And it is not discussed as to why some comparisons are good and others are not at all. How can we expect the model to ever get d15NO₃⁻ values exactly right when chemistry is not even included in the model? It would make a lot more sense to compare seasonal averages or time series from the model versus time series of the observations and focus on the relative changes from season to season rather than leading the reader to expect that there should be a 1:1 relationship for the model-observation comparison. Further the last line of this section (page 31, line 10) is not appropriate speculation. There's nothing to justify this statement and again the model is not simulating nitrate fields.

Fix

Page 31, Conclusion:

The discussion of “uncertainty” needs to be clarified. This is not addressed earlier in the text, so

it is not clear how an uncertainty of less than 5 per mil (line 18) was determined. The word uncertainty does not make sense in the context here. And discussion of uncertainty is also unwarranted since the model does not consider/test the full ranges of the observed NO_x source signatures for different emission sources. In fact the word uncertainty is used again to describe the uncertainty in NO_x emission, which is documented in the literature. Really what is being discussed is the narrow range predicted by the model – or so it appears to me.

Rephrased, the uncertainties here refers to the d15N values in emission input dataset, rather than the uncertainties of the prediction of the model.

Line 28: this sentence makes no sense and it is never discussed in the text that there is such a large uncertainty in NO_x power plant emissions.

Reference added

Line 35 and 37: change the phrase “are possible to be”

Confirmed

Line 37: change “estimated” to “predicted” and add “to be” following NO_x

Confirmed

Line 44: what constitutes a “better” simulation? Ie. Based on what is it deemed better?

Rephrased