nature portfolio

Peer Review File

Changing patterns of global nitrogen deposition driven by socio-economic development

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The authors present a large dataset of N deposition measurements derived from co-ordinated sampling programmes and ad hoc observations published in the scientific literature. This is certainly an interesting and important dataset that deserves publication. I have some reservations about the methods used for modelling global distributions of N deposition, and the claim that the proposed 'Normal Distribution Curve Model' is innovative. I would be happy to read a revised version of this paper if the authors can address the following questions:

1. Do you intend to publish your compiled N deposition data along with this manuscript?

2. N deposition is both spatially and temporally autocorrelated. Spatial interpolation, as attempted in this paper, is normally done using geostatistical methods (Kriging) on the residuals of a fitted model. Kriging also provides uncertainties for predictions. The methods described in this manuscript do not adequately indicate how spatial interpolation was conducted, nor how spatial autocorrelation was dealt with. The SEM used make sense, but I'm not clear as to why random forest models were employed.

3. The observational data (Fig. S1) are extremely biased. Therefore, I am highly skeptical that anything can be said about the status and trends in N deposition in regions like Africa, Central Asia, Latin America and Australia.

4. The NDCM is described as innovative but is nothing more than an Environmental Kuznets Curve (which must be what the authors mean by EKC). This is not a Normal (Gaussian) process even though it may approximate one. It is simply a function of economic development and a shift from agriculture and extractive industries to service industries, along with the introduction of environmental protection laws. The manuscript could be beneficially shortened by greatly reducing sections 4.1–4.3 and simply reporting the interesting empirical findings of the relationship between N deposition and per capita GDP.

5. If Fig. 5 is kept, please remove the background colours and use a sans serif font to make the figure more readable and pleasing to the eye.

6. In many figures, linear or quadratic trendlines are included which do not relate to any hypotheses. For example, in Fig. 1 b and c, there is no expectation that deposition follow a quadratic function of time. Just show the data and omit the lines.

7. In Fig. 3c, plot one line for Developed and one line for Developing regions against a single temporal axis (as in panel b). The arrows and coloured bars are superfluous.

8. I would like to have seen more explicit analysis and discussion of the relative contributions of agriculture (if possible, mineral vs. organic fertilizer) and industry to the changes.

9. Agriculture should be mentioned in the Abstract.

10. The Introduction section is too long and repetitive. Avoid statements like “quantifying new and changing patterns of N deposition and their effects on the global environment is urgent”. On the contrary. N deposition is declining, and in any case, the effects of N deposition have been well studied for decades and there is no urgency due to lack of knowledge.

11. The authors state several times that atmospheric transport models are highly uncertain, without stating a source. The onus is on the present paper to demonstrate that the highly biased observational data yield a more accurate and precise estimate of N deposition than these models.

Reviewer #2 (Remarks to the Author):

This study introduces a comprehensive global nitrogen (N) deposition database spanning from 1977 to 2021, compiled from diverse observation networks and published literature. Employing Machine Learning techniques, specifically Random Forest models, the database underwent reconstruction. Utilizing this resource, the authors conducted an analysis of current global N deposition patterns across both spatial and temporal dimensions while identifying key driving factors. Their findings indicate a shift in global N deposition hotspots from developed nations to developing ones, with gross domestic product per capita emerging as the primary influencer of these patterns. However, these conclusions lack novelty as they echo previous reports. The potential significance lies in the accuracy of a new estimate of global N deposition, if indeed accurate. According to their estimations, the global annual input of N through deposition to land in 2020 was approximately 116.8 Tg N, significantly surpassing previous estimates ranging from 63.9 to 90.4 Tg N. While the new estimate is of interest, several methodological weaknesses undermine confidence in the conclusions.

Firstly, the geographical bias in monitoring site distribution, heavily concentrated in North America, Europe, and East Asia, raises concerns regarding the accuracy of global estimations, particularly for underrepresented regions like South America and Africa. This bias may lead to an overestimation of N deposition in regions with more extensive coverage, skewing global estimates.

Secondly, the manuscript lacks a thorough analysis of uncertainty associated with input datasets and model hyperparameters, essential for assessing the robustness and reliability of results. Given the inherent uncertainties in input data and the sensitivity of random forest models to hyperparameters, providing uncertainty ranges for estimations is imperative.

Moreover, discrepancies between the estimated global N deposition and other model-based estimates raise questions about the database's accuracy and reliability. Additionally, the use of machine learning models, known for their "black box" nature, introduces further ambiguity, necessitating thorough evaluation and validation.

In conclusion, while the study sheds light on global N deposition dynamics, addressing the outlined methodological concerns is crucial for bolstering the credibility and generalizability of the findings. I recommend that the authors undertake rigorous efforts in data accuracy, model validation, and uncertainty analysis before considering publication.

(Italics are comments from reviewers)

Reviewer #1 (Remarks to the Author):

The authors present a large dataset of N deposition measurements derived from co-ordinated sampling programmes and ad hoc observations published in the scientific literature. This is certainly an interesting and important dataset that deserves publication. I have some reservations about the methods used for modelling global distributions of N deposition, and the claim that the proposed 'Normal Distribution Curve Model' is innovative. I would be happy to read a revised version of this paper if the authors can address the following questions:

Response: Thank you very much for your positive feedback. Despite the significant time and effort, we have devoted to compiling and collecting global N deposition observational data, it is regrettable, as you and Reviewer 2 have noted that the observational site still suffers from severe geographical bias. Regions such as Africa, Central Asia, Latin America, and Australia have sparse or missing observation sites, which is an unavoidable truth at present. This underscores the importance of our study in establishing a data-driven global N deposition prediction model to assess areas lacking N deposition observation. Additionally, we call for enhanced site observations in these regions in the future to reduce prediction uncertainties, particularly in areas like Africa where socioeconomic statistical data is also incomplete.

In our analysis of regional dynamics, we primarily utilized arithmetic or weighted averages of observation sites within each region to obtain annual values. This region-specific analysis ensures the accuracy of depicting dynamic changes in N deposition. Moreover, structural equation modeling, correlation, and regression analyses were conducted using site or regional data, which guarantees the robustness of the relationships between driving factors and N deposition.

The random forest method was employed in this study to upscale site data to global products (2008-2020). Although the predicted results may have uncertainties, we believe this approach is necessary since current global N deposition products are only derived from atmospheric chemical transport models. In this new revised version, we developed a new framework for predicting global N deposition grid product to reduce prediction uncertainties. We also incorporated a grid search method for hyperparameter selection of the random forest prediction model and used recursive feature elimination (RFE) for variables selection, both of which have improved our prediction accuracy.

1. *Do you intend to publish your compiled N deposition data along with this manuscript?*

Response: The global N deposition grid dataset will be archived in a public repository after acceptance.

2. N deposition is both spatially and temporally autocorrelated. Spatial interpolation, as attempted in this paper, is normally done using geostatistical methods (Kriging) on the residuals of a fitted model. Kriging also provides uncertainties for predictions. The methods described in this manuscript do not adequately indicate how spatial interpolation was conducted, nor how spatial *autocorrelation was dealt with. The SEM used make sense, but I'm not clear as to why random forest models were employed.*

Response: Thank you very much for pointing out this issue. In the methods section, we mentioned that for some regions and years with missing values, we used interpolation methods to obtain them. This refers to temporal interpolation rather than spatial interpolation. For instance, if N deposition data for a region in Southeast Asia was missing for 1997, we fitted an optimal equation based on the long-term dynamic data series for that region and used this equation to interpolate the N deposition value for 1997. Therefore, the interpolation method used does not involve spatial autocorrelation issues. We have added a clarification in the methods section to better explain this issue.

The random forest model was used to upscale site data to global products (2008-2020). Although the estimated global N deposition through this method may have uncertainties, we believe the spatial patterns and temporal dynamics are consistent with prior knowledge. In the future, observations in regions such as Africa, Central Asia, Latin America, and Australia will be needed to constrain the uncertainties introduced by site biases in the upscaling process.

3. The observational data (Fig. S1) are extremely biased. Therefore, I am highly skeptical that anything can be said about the status and trends in N deposition in regions like Africa, Central Asia, Latin America and Australia.

Response: We fully agree with your viewpoint. Unfortunately, the sparse or missing distribution of observation sites in these regions is a current reality we face. In this revision, we developed a new framework for predicting global N deposition grid product to reduce prediction uncertainties. Meanwhile, we primarily utilized arithmetic or weighted averages of observation sites within each region to obtain annual values and report the regional dynamics (Fig. 2). This region-specific analysis ensures the accuracy of depicting dynamic changes in regional N deposition.

Although limited, there are some observation sites in the African region, and we have analyzed its dynamic changes using site data (Supplementary Figure 4). Additionally, remote sensing column concentration data of NH_3 and NO_2 provide support for the N deposition changes in the African region.

For other regions like Australia, where there are no observation sites, we can only understand their fluxes and spatiotemporal changes through the upscaled results of the random forest model. This indeed presents significant uncertainties, which we have discussed in the uncertainty analysis section of the paper.

4. The NDCM is described as innovative but is nothing more than an Environmental Kuznets Curve (which must be what the authors mean by EKC). This is not a Normal (Gaussian) process even though it may approximate one. It is simply a function of economic development and a shift from agriculture and extractive industries to service industries, along with the introduction of environmental protection laws. The manuscript could be beneficially shortened by greatly reducing sections 4.1–4.3 and simply reporting the interesting empirical findings of the relationship between

N deposition and per capita GDP.

Response: Thank you very much for your suggestion. In this revision, we have greatly simplified the text, making the article more concise.

5. If Fig. 5 is kept, please remove the background colours and use a sans serif font to make the figure more readable and pleasing to the eye.

Response: Thank you very much for your advice. We have deleted Fig.5 in this version.

6. In many figures, linear or quadratic trendlines are included which do not relate to any hypotheses. For example, in Fig. 1 b and c, there is no expectation that deposition follow a quadratic function of time. Just show the data and omit the lines.

Response: Thank you for your advice. The fitting curve we added in the figure is to better explain the changing trend of N deposition, such as continuous increase, first increase and then decrease, so we choose to retain it.

7. In Fig. 3c, plot one line for Developed and one line for Developing regions against a single temporal axis (as in panel b). The arrows and coloured bars are superfluous.

Response: Thanks for your suggestion. Change made.

8. I would like to have seen more explicit analysis and discussion of the relative contributions of agriculture (if possible, mineral vs. organic fertilizer) and industry to the changes.

Response: Thank you very much for your constructive suggestion. While previous studies have simulated long-term global N deposition dynamics using atmospheric chemical transport models and explored the contribution of agricultural ammonia deposition to total deposition (Liu et al., 2022), they did not investigate the relative contributions of N emissions from different sources (industrial and agricultural) to global N deposition changes.

Inspired by your suggestion, in the new version, we fitted curves for global total N deposition, ammonium deposition, and nitrate deposition from 1980 to 2020. Using the first derivatives, we calculated their respective deposition change rates. Then, by comparing the change rates of ammonium deposition (mainly from agricultural sources) and nitrate deposition (mainly from industrial sources), we determined their relative contributions to the changes in total deposition. This analysis yielded interesting results. We found that during the period from 1980 to 2020, as global total deposition transitioned from rapid increase to gradual stabilization, the relative contribution of agricultural ammonia deposition decreased, while the relative contribution of industrial nitrate deposition increased. We included a supplementary figure Supplementary Figure 12) to illustrate their relative contributions and added related discussions in the text.

Additionally, nitrogen emissions from agricultural sources primarily come from N fertilizer application and livestock farming. Currently, it is challenging to distinguish the relative contributions of N fertilizer (especially differentiating between inorganic and organic fertilizers) and livestock farming to N deposition changes. This represents a promising direction for future research.

9. Agriculture should be mentioned in the Abstract.

Response: Thank you for your advice. Change made.

10. The Introduction section is too long and repetitive. Avoid statements like "quantifying new and changing patterns of N deposition and their effects on the global environment is urgent". On the contrary. N deposition is declining, and in any case, the effects of N deposition have been well studied for decades and there is no urgency due to lack of knowledge.

Response: Thank you for your suggestion. Change made.

11. The authors state several times that atmospheric transport models are highly uncertain, without stating a source. The onus is on the present paper to demonstrate that the highly biased observational data yield a more accurate and precise estimate of N deposition than these models.

Response: We fully acknowledge your suggestion. Here, we provide a set of global N deposition data obtained directly through upscaling of observational data. This is the only dataset independent of atmospheric transport model simulation results. Of course, due to the uneven distribution of observational data, we must acknowledge that there may be uncertainties in the results, but they still fall within a reasonable range. We have recalculating global N deposition based on the new framework, and also cited relevant literature that discusses the uncertainties introduced by atmospheric transport models. In the future, we can improve the accuracy of global N deposition using methods like data-model fusion, which combines observational data and atmospheric transport models.

Reviewer #2 (Remarks to the Author):

This study introduces a comprehensive global nitrogen (N) deposition database spanning from 1977 to 2021, compiled from diverse observation networks and published literature. Employing Machine Learning techniques, specifically Random Forest models, the database underwent reconstruction. Utilizing this resource, the authors conducted an analysis of current global N deposition patterns across both spatial and temporal dimensions while identifying key driving factors. Their findings indicate a shift in global N deposition hotspots from developed nations to developing ones, with gross domestic product per capita emerging as the primary influencer of these patterns. However, these conclusions lack novelty as they echo previous reports. The potential significance lies in the accuracy of a new estimate of global N deposition, if indeed accurate. According to their estimations, *the global annual input of N through deposition to land in 2020 was approximately 116.8 Tg N, significantly surpassing previous estimates ranging from 63.9 to 90.4 Tg N. While the new estimate is of interest, several methodological weaknesses undermine confidence in the conclusions.*

Response: Thank you very much for your constructive comments. Despite the significant time and effort, we have devoted to compiling and collecting global N deposition observational data. It is regrettable that the observational site still suffers from severe geographical bias. This is an unavoidable reality at present.

Due to the time series limitations of NH_3 column concentration data, we used the random forest method to upscale site data to global products only for the period 2008-2020. For the period 1980-2007, we obtained global and regional means by calculating the weighted averages of regional site observational data. Therefore, we believe the temporal dynamic trends of global (and regional) N deposition are robust. Additionally, the analysis of the driving mechanisms of global N deposition was conducted using site or national-scale observational data, making these results reliable.

To minimize the influence of the uneven distribution observation site on predicting global N deposition, we developed a new framework to generate the global grid N deposition dataset in this revision. The new results are much lower than out previous estimations. Of course, due to the uneven distribution of observational data, there may be uncertainties. However, compared to the total emissions of global reactive N, the results fall within a reasonable range. To our knowledge, this is the only dataset independent of atmospheric transport model simulation results.

1. *Firstly, the geographical bias in monitoring site distribution, heavily concentrated in North America, Europe, and East Asia, raises concerns regarding the accuracy of global estimations, particularly for underrepresented regions like South America and Africa. This bias may lead to an overestimation of N deposition in regions with more extensive coverage, skewing global estimates.*

Response: We agree with you very much. Unfortunately, the uneven distribution of observed data on global nitrogen deposition is an unavoidable fact at present, which brings uncertainty to the assessment results.

In the revised version of our paper, we developed a new framework for predicting global N deposition grid data. Firstly, we divided land into two categories: wilderness and human modified area. And then we predicted their N deposition flux, respectively. We believed this framework can greatly reduce prediction uncertainties due to the reduction of forecast area, such as The Sahara Desert in Africa, high northern latitudes. Our previous results were also high in these regions. We also compared our prediction results in North America, Europe, and East Asia with previous studies. Their values are reasonable and close to previous researches.

2. *Secondly, the manuscript lacks a thorough analysis of uncertainty associated with input datasets and model hyperparameters, essential for assessing the robustness and reliability of results. Given the inherent uncertainties in input data and the sensitivity of random forest models to hyperparameters, providing uncertainty ranges for estimations is imperative.*

Response: Thank you very much for your suggestion. In this revision, we also incorporated a grid search method for hyperparameter selection of the random forest prediction model and used recursive feature elimination (RFE) for variables selection, both of which have improved our prediction accuracy.

3. *Moreover, discrepancies between the estimated global N deposition and other model-based estimates raise questions about the database's accuracy and reliability. Additionally, the use of machine learning models, known for their "black box" nature, introduces further ambiguity, necessitating thorough evaluation and validation.*

Response: Thank you very much for your suggestion. Our understanding of global atmospheric N deposition has primarily come from the simulation results of atmospheric transport models. The driving data for these models (such as reactive N emissions data) and parameter settings (such as the residence time of reactive nitrogen) can introduce significant uncertainties into the simulations. To our knowledge, our results represent the only global nitrogen deposition dataset obtained by upscaling observational data. Although there are uncertainties in the results, they still provide valuable insights. In the future, techniques that use observational data to constrain model simulations and data-model fusion should be further applied.

4. *In conclusion, while the study sheds light on global N deposition dynamics, addressing the outlined methodological concerns is crucial for bolstering the credibility and generalizability of the findings. I recommend that the authors undertake rigorous efforts in data accuracy, model validation, and uncertainty analysis before considering publication.*

Response: We fully agree with your viewpoint and suggestions. In the new version, we have simplified the main content of the paper, developed a new framework, and strengthened the discussion of the random forest model prediction results and uncertainty analysis. We hope this meets your expectations.

REVIEWER COMMENTS

Reviewer #1 (Remarks to the Author):

The authors have improved the manuscript and removed a lot of unnecessary material.

The results for areas with large amounts of observational data (North America, Europe, China), i.e. the human-modified areas, are likely to be robust.

The estimation of economic development stage at which N emissions begin to decline is valuable and interesting. The switch from agricultural to industrial emissions is well known but this is a nice illustration.

The results for areas with no observational data (Africa, Latin America, large parts of Southeast Asia, Australia) are based only on “normalized” satellite-derived column concentrations of NH3 and NO2 (Equation 1). What this normalization entails is not described. This model does not appear to take depositional pathways (wet or dry) or meteorological effects into account. This seems to be a weakness. Also, what does the 0.01 multiplier do?

I would like to see a map of uncertainty estimation (e.g. 95% confidence intervals) for the gridded estimates.

How does this manuscript compare to the results presented by (Rubin et al., 2023; Vishwakarma et al., 2023)? Does this manuscript provide any substantial advance over these results?

The authors state that the results for Africa are higher than expected, but seem in line with those of (Vishwakarma et al., 2023). The modelling conducted by (Rubin et al., 2023) appears to be more sophisticated and thus may be more reliable for regions without ground observations.

The authors persist in fitting (polynomial) curves to data. There is no reason to do this, there are no meaningful hypotheses being tested. If necessary, fit a linear trend to the most recent observations to determine whether there is an increasing or decreasing trend.

Formally, temporal trends fitted to repeated measures must consider temporal autocorrelation.

Fig. S9, S10. Please weight arrow width by coefficient magnitude.

Lines 261-278: These are rather bland conclusions (everyone should reduce their N emissions) and can be removed.

References

Rubin, H. J., Fu, J. S., Dentener, F., Li, R., Huang, K., & Fu, H. (2023). Global nitrogen and sulfur deposition mapping using a measurement–model fusion approach. Atmospheric Chemistry and Physics, 23(12), 7091–7102. https://doi.org/10.5194/acp-23-7091-2023

Vishwakarma, S., Zhang, X., Dobermann, A., Heffer, P., & Zhou, F. (2023). Global nitrogen deposition inputs to cropland at national scale from 1961 to 2020. Scientific Data, 10(1), 488. https://doi.org/10.1038/s41597-023-02385-8

Reviewer #2 (Remarks to the Author):

The revised manuscript shows significant improvement and addresses most of my previous comments. The key contribution of this study is its data-driven estimate of global nitrogen deposition. I recommend highlighting the estimated global nitrogen deposition of 92.7 Tg N for 2020 in the Abstract, as I believe this is the most crucial result.

However, I am concerned about the new framework introduced in the revised manuscript, which results in a 24.1 Tg N reduction from the previous estimate, representing a 21% decrease from 116.8 Tg N to 92.7 Tg N. This discrepancy suggests significant variations among the different frameworks used. It would be prudent to include a careful discussion on these discrepancies in global nitrogen deposition estimates across the various frameworks. Overall, I am pleased to see this work added to the literature.

(Italics are comments from reviewers)

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1. *The authors have improved the manuscript and removed a lot of unnecessary material. The results for areas with large amounts of observational data (North America, Europe, China), i.e. the human-modified areas, are likely to be robust. The estimation of economic development stage at which N emissions begin to decline is valuable and interesting. The switch from agricultural to industrial emissions is well known but this is a nice illustration.*

Response: We greatly appreciate your thoughtful review and are pleased that you find our revised manuscript significantly improved and our findings compelling.

2. *The results for areas with no observational data (Africa, Latin America, large parts of Southeast Asia, Australia) are based only on "normalized" satellite-derived column concentrations of NH3 and NO2 (Equation 1). What this normalization entails is not described. This model does not appear to take depositional pathways (wet or dry) or meteorological effects into account. This seems to be a weakness. Also, what does the 0.01 multiplier do?*

Response: We apologize for not adequately addressing this "normalized" method. We only applied this method for wilderness areas (not all regions without observational data). These wilderness areas are primarily located in high-latitude regions of the Northern Hemisphere (such as Siberia, Greenland), the Sahara Desert in Africa, the Qinghai-Tibet Plateau in China, and desert regions of Australia (Supplementary Figure 14). The NH_3 and NO_2 satellite column concentrations in these areas are very low, and we believe that human impact is minimal, resulting in low N deposition levels.

As you mentioned, due to the lack of direct observational data in wilderness areas, it is challenging to incorporate depositional pathways and meteorological influences into the prediction model for these regions. Therefore, we used a simpler linear model, assuming that areas with higher N satellite column concentrations have higher deposition fluxes. The 0.01 in the equation is a unit conversion factor that considers pre-industrial N deposition levels, which has been illustrated in the method section of revised manuscript.

3. *I would like to see a map of uncertainty estimation (e.g. 95% confidence intervals) for the gridded estimates.*

Response: Thank you for your constructive comments. We have supplied a map to show the relative uncertainty estimation in this revision (Supplementary Figure 13). The relative uncertainty was defined as the ratio of standard error to the mean value of three models.

4. *How does this manuscript compare to the results presented by (Rubin et al., 2023; Vishwakarma et al., 2023)? Does this manuscript provide any substantial advance over these results? The authors state that the results for Africa are higher than expected, but seem in line with those of (Vishwakarma et al., 2023). The modelling conducted by (Rubin et al., 2023) appears to be more sophisticated and thus may be more reliable for regions without ground observations.*

Response: Thank you for your suggestion. Rubin et al. (2023) used a model-data fusion approach to assess global atmospheric N and sulfur deposition. They employed the simulation results from 11 atmospheric chemistry transport models and combined them with 2010 observational data from major global atmospheric N deposition networks to correct the model results. They only reported N deposition results for 2010, and their study primarily applied the model-data fusion method to the dry and wet deposition of NH_4^+ and NO_3^-. However, for dry deposition components such as NH_3, HNO_3, and NO_2, they still relied solely on model simulations.

Although Vishwakarma et al. (2023) provided global atmospheric N deposition results for farmland from 1961 to 2020, they mainly used two N deposition datasets (the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) and Wang et al.) and two global farmland datasets (Land-Use Harmonization 2 (LUH2) and the History of the Global Environment database (HYDE)) to produce their global farmland N deposition dataset. They did not directly simulate or construct N deposition data but instead relied on two pre-existing model simulation products. While their results focused only on farmland, the N deposition flux and input for Africa were lower than our assessment of N deposition in the region.

Our Monitoring-based Global Nitrogen Deposition (MGND) not only covers all N deposition components but also includes temporal dynamics, allowing us to directly assess global N deposition trends based on observational data. As we emphasized, this is the only dataset independent of model simulations. While Rubin et al. (2023) used a model-data fusion approach, it is important to note that there are significant differences between the various model simulation results and they only applied this method to the regions with observation data. In regions lacking observational data, model results are also difficult to validate, so it is hard to definitively claim that model results are necessarily more accurate or reliable.

5. *The authors persist in fitting (polynomial) curves to data. There is no reason to do this, there are no meaningful hypotheses being tested. If necessary, fit a linear to the most recent observations to determine whether there is an increasing or decreasing trend. Formally, temporal trends fitted to repeated measures must consider temporal autocorrelation.*

Response: Thank you for your suggestion. We have removed the fitted curve in Figure 1 but kept it in Figure 2. We agreed that temporal trends fitted to repeated measures can consider temporal autocorrelation to pursue the overall predictive performance and explanatory power. However, the fitted curve in Figure 2 is not intended to test any hypothesis or predict future trend, but rather to more clearly illustrate the evolution of nitrogen deposition over the past 40 years in each region. This helps us categorize the dynamic changes in nitrogen deposition into three types: decline, transition, and increase. Additionally, since some regions have missing observational data for certain years, it can be difficult to discern trends directly from the scatter plot, which is why we chose to retain the fitted curve in Figure 2.

6. *Fig. S9, S10. Please weight arrow width by coefficient magnitude.*

Response: Thanks. Change made.

7. *Lines 261-278: These are rather bland conclusions (everyone should reduce their N emissions) and can be removed.*

Response: Thanks. We have removed these sentences.

References

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Response: Thanks. We have added the data in the Abstract in this revision.

9. *However, I am concerned about the new framework introduced in the revised manuscript, which results in a 24.1 Tg N reduction from the previous estimate, representing a 21% decrease from 116.8 Tg N to 92.7 Tg N. This discrepancy suggests significant variations among the different frameworks used. It would be prudent to include a careful discussion on these discrepancies in global nitrogen deposition estimates across the various frameworks. Overall, I am pleased to see this work added to the literature.*

Response: Thank you very much for your suggestion. In this revision, we have added a discussion on the impact of the modeling framework on the prediction results.

REVIEWERS' COMMENTS

Reviewer #1 (Remarks to the Author):

The authors have addressed my previous comments.