Peer Review File

The optimum nitrogen fertilizer rate for maize in the US Midwest is increasing

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This manuscript has been previously reviewed at another journal. This document only contains information relating to versions considered at Nature Communications.

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 1:

Reviewer comments:

Reviewer #4

(Remarks to the Author)

The authors have figured out my concerns in the revision. Here, I would like to suggest that the difference between the continuous maize system and maize-soybean system should be enough discussed, since the optimum nitrogen fertilizer rate for them was different, especially the tradeoff in Fig.5.

Reviewer #5

(Remarks to the Author)

Baum et al combined data from long-term and short-term nitrogen experiments combined with the crop growth modeling simulation model APSIM to assess trends in agronomic, economic and environmental optimal N rates (AONR, EONR and EnvONR) over the period 1991-2021. As expected agronomic optimal N rates, AONR, > economic optimal N rates, EONR > environmental optimal N rates, EnvONR. They showing that the increase in EONR is higher than AONR and EnvONR implying that the difference between agronomic and economic optimal N rates decreased over time while the difference in economic and environmental optimal N rates increased over time. They then show that reducing N rates from the economic to the environmental optimum based reduce maize productivity by 6% while the N loss reduction is small.

In general I find the paper interesting and their replies to the comments adequate. I have still some additional main comments and specific comments that are relevant in my view

Main comments

The abstract is not very informative and does not convey well the main message of this paper. I would think that the main message is that

• the difference between agronomic and economic optimal N rates decreased over time while the difference in economic and environmental optimal N rates increased over time.

The reduction in N rates from the economic to the environmental optimal N rate comes at a cost for yield at low N loss reduction

The first point is not mentioned and the last point comes with nuances due to two aspects

• this low N loss reduction is related to the assumed high societal costs of N losses. To me this part is not really so clear. Note e.g. that cost of huma health related to contaminated drinking water only play a role at high NO3 levels (above > 50 mg NO3/l) and it EONR prevents sch levels, further reductions do not imply any reduction in costs related to human health improvements. The point that I like to make is that the N loss reduction might be low but if the societal costs are really so high, it could be worth it but if it is an overestimate, the EnvONR would come much close to the EONR and the effect of reducing EONR to EnvONR on crop yield would be much less

• this low N loss reduction is based on the current NUE. The point of reconciling crop yield with environmental impacts has of course to do with increasing the NUE up to an attainable level (see e.g. Schulte-Uebbing and de Vries, 2021; Chang et al., 2021). I would discuss both aspects in the aper and shortly summarize in the abstract

NB: the extra space needed for this can come from removing the text "We call for improved and timely predictability of the optimum N rate across space and time as a solution to maximizing productivity, sustainability, and profitability. Present

findings serve as a benchmark to guide future N fertilizer rate research, recommendations, and policy". This is vague and not well related to the results .I can imagine that some text on the implications is needed and I would then rather say: "we call for an enhanced assessments of the environmental optimal N rate (as that part is most unreliable based on societal costs) to assess the need increase in N use efficiency considering the need to maintain or even increase the current crop yields"

References

Schulte-Uebbing L.F. and W. de Vries, 2021. Reconciling food production and environmental boundaries for agricultural nitrogen inputs in the European Union. Science of the total Environment 786 (2021) 147427. https://doi.org/10.1016/j.scitotenv.2021.147427 Chang, J., P. Havlík, D. Leclère, W. de Vries, H. Valin, A. Deppermann, T. Hasegawa and M. Obersteiner, 2021. Global food security risks associated with meeting regional nitrogen boundaries. Nature Food 2: 700–711.

https://doi.org/10.1038/s43016-021-00366-x

Specific comments

Line 70-71: Based on results of Wang et al., 2014; Martinez-Feria et al., 2018, the authors state that the EnvONR is typically lower than the AONR at a rate of 35-41%. I assume they mean that EnvONR is typically 35-41% lower than the AONR. I would then rather think that it is logic to give such results also for this study and then how the %difference changes in time from 1991-2021. And do this also for EONR. In general the paper does not give enough attention to the changing differences between the various rates in time that I see as a main result (see comment above).

Line 74-76: The authors state here that. "A summary of the potential cost of N loss can be found in Sobota et al. (2015), which attributes cost ranges associated with eutrophication, the increased health risk from human consumption, and nitrous oxide and NOX emissions to the atmosphere". To me this is not clear. To start with, NOx emissions from agriculture enhance the concentrations of particulate matter affecting human health but these emissions are limited. However, this is not true for NH3 emissions and both NOx and Nh3 enhance the concentrations of particulate matter affecting human health (see e.g. an overview of N impacts by de Vries, 2021). Why is NH3 emissions then not included in the assessment. Second, the increased risk of colon cancer due to nitrate contamination only occurs above a threshold level near 10 mg NO3-N/l and is that reached (see comments above)?

De Vries, W., 2021. Impacts of nitrogen emissions on ecosystems and human health: a mini review. Current Opinion in Environmental Science & Health 100249. https://doi.org/10.1016/j.coesh.2021.100249

Line 97-98. The authors hypothesize that over the last decades, the optimum N rate for maize production is increasing because N outputs to grain and the environment exceed the reduction in grain N concentration. To me this is not clear since the reduction in grain N concentration affects the N outputs to grain being the product of grain yield and N concentration. Do you now say that you expect that the increase in growth rate by roughly 1.2% per year is likely higher than the reduction in the concentration of grain N?. But can you not show this directly from data on trends in grain N? Or do you now know this and thus link it to the sum of N out and N leaching?

Line 103-106. The authors state that long-term experiments are prone to exaggerate maize's yield response to N as N treatments reach a steady-state due to consistent over-under fertilization. Why exaggerate? What is reaching a steady state: the soil N pool or the yield. has it to with N immobilization in short term experiments that do not occur any more after a long time period. This needs explanation.

Line 130-131. The authors find that the increase in EONR is comparable to the maize yield increase. This leads to the question, which data were used for the grain N concentrations in the modelling. Measured data? And did they possibly stay constant over time?.

Specific comments on Figures and supplementary tables

Figure 2 and Table S1.

• At the end of figure 1, I would add: The full regression equations are given in Table S1. Make the number of significant digits equals in figure (you use 3, e.g. 2.38x) and table (you use 4, e.g. 2.382x). I would for sure not use more than 3 (even that is high considering the uncertainty). Note that you do this in Figure S1, S4 and S5!

• In table S1, you need to skip "& 3". The linear equations for each optimum nitrogen (N) rate and crop rotation are related to Fig. 2 only. In fig 3, the full equations are given in the figure.

Figure 4 and Table S2.

• At the end of figure 4, I would add: The full regression equations are given in Table S4

• In both the figure and table I would use at maximum three significant digits. So 0.011; 0.110; 1.10, 11.0,

Reviewer #6

(Remarks to the Author)

The primary purpose of this review is to check the authors' responses to the previous Reviewer #2.

The numbers below correspond to each of Reviewer #2's general comments.

1. The inclusion of the limitation flagged by this reviewer in the introduction should be sufficient to address this concern, however, it is unclear to the reader if the authors have clearly described this limitation. What is meant by "over-under fertilization"? Why does this lead to a steady state? What is meant by "production fields"? Please clarify this sentence to address this comment.

2. Is this sensitivity analysis included in a supplementary file? If not, consider including. Based on the conclusions of the sensitivity analysis described by the authors, I am satisfied that using a fixed price is appropriate.

3. I am satisfied the authors with the authors' response to this comment.

4. I would argue that this recalculation did change the results fairly substantially (i.e., the EONR was increased by 11% from 2.58 to 2.85), and am satisfied that these years were removed from the analysis in response to Reviewer #2's comments.

5. This concern has not been sufficiently addressed. Suggest to explicitly include mention of weather effects in the final paragraph of the discussion in response to this comment. The authors should consider including a sentence regarding the effects of weather and potentially climate change, to tie in with mentions of climate change in the introduction and the first paragraph of the discussion.

6. Satisfied with this response.

7. The EnvONR is sufficiently described in the materials and methods.

8. I agree with the authors' response that variable rate N application is not within the scope of this publication. However, to address this comment, an acknowledgement of this challenge could be included as a sentence in the final paragraph of the discussion.

9. This comment has been sufficiently addressed.

Specific comment #8. Disagree with this response by the authors. Once an acronym has been defined, please use it throughout the text.

Specific comments from this reviewer: L88-89 - add 't' after 'simultaneous and' L102 - why introduce soil C research here? Is this important for this manuscript? L126 - is 'well known' necessary? Please strike. L201&207 - these lines have repetitive language. Please revise.

Version 2:

Reviewer comments:

Reviewer #5

(Remarks to the Author)

I really like the revision of the paper by Baum et al. First of all, they tackled my main comment very well, both in improving the abstract and by adding two Figure s (S6 and S7) in the supplementary material illustrating the points that I like to make.

My specific comments are also mostly well tackled. I have some problems with the answers discussed below

1 The answer related to the role of NH3 in human health impacts by PM. Mentioning that it is uncertain is true for all the societal impacts and the reference (Kelly et al., 2004) is very old. But I can imagine that the authors like to leave it out as the NH3 losses are small. I would then rephrase the addition as follows

"Note we exclude the potential cost of the formation of particulate matter PM10 and PM2.5 from NH3 volatilization from inorganic fertilizers in this analysis because NH3 volatilization is marginal within the N management practices used for maize production in the US Midwest (Woodley et al., 2020).

You might add: In addition there are large uncertainties in overall impact on NH3 on PM2.5/PM10 and thereby on human health but I am inclined to remove this text. There has been a nature paper on the high impacts of NH3 emissions on PM2.5 in Nature by Gu et al (2023_

Gu et al., 2023. Cost-effective mitigation of nitrogen pollution from global croplands. Nature Vol 613 | 5 January 2023 | 77

2 The answer related to the statement "We hypothesize that over the last decades, the optimum N rate for maize production is increasing because N outputs to grain and the environment exceed the reduction in grain N concentration". This statement remains strange as N outputs to grain is the product of crop yield and grain N concentration. I would say: "We hypothesize that over the last decades, the optimum N rate for maize production is increasing due to an increase in N outputs to grain, since crop yields increase likely exceed grain N concentration reductions, and an increase in N losses".

One last comment I would revise Fig S1-S5 as follows Fig S1- is Fig S5 Fig S2- is Fig S1 Fig S3- is Fig S2 Fig S4- is Fig S3 Fig S5- is Fig S4 This is the order in which they appear in the text.

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Response to reviewers' comments

Dear Editor, we thank you for the opportunity to revise our manuscript and re-submit. We addressed each reviewer's comment (see detailed responses below), and the manuscript has considerably improved. The major changes made are listed below:

- 1. Removed the year 2019 as suggested and rerun the analysis conclusions did not change
- 2. Added new text in MM for the price ratio and its impact on the optimum N rate
- 3. Highlighted more the impact of long-term experiments on the results
- 4. Added new supplementary materials for the regression models used

Please see our response to each reviewer comment below

Thank you.

Reviewers' Comments:

Reviewer #1:

Remarks to the Author:

The paper represents an impressive cooperative effort to establish time trends in N use in maize production and does it for optimum agronomic, economic and environment trends. Although the trends reported might be expected, the research provides quantitative trends in two common maize production systems and produces seemingly reliable estimates of N fertilizer use efficiency, N leaching and gaseous losses and net soil mineralization. The paper is not necessarily novel, certainly not a breakthrough, as this topic is heavly discussed in technical journals. The title of the paper seems obvious without the research results that verified the fact that more N fertilizer is needed to take advantage of the genetic improvement in maize yields taking place over the time. The average USA corn yields has been increasing at an annual rate of about 2 bu/acre for about 50 years. The factor that has changed the most is the cost of the fertilizer and the application cost, making the economic optimum go down somewhat, as the paper shows. Calculating an environmental optimum seems mute since the monetary values used can be almost anything, as Figure S4 demonstrates.

Response: We thank the reviewer for the positive comments and criticism, but we must clarify here that "applied N fertilizer rate" and "optimum N fertilizer rate" are two different terms. The reviewer was referring to the "farmer's applied N rates". Our study is about the "optimum N rate", which is strongly related to profitability and environmental sustainability. Our study is the first to report "optimum N rate" historical trends, hence, very novel and timely because it fills a significant knowledge gap and provides actionable data to support decision-makers, scientists, and N recommendation systems. The reviewer mentioned that the fertilizer cost is a major driver of the economic optimum N rate, we agree, but the selling price of corn is also a major driver. Interestingly, the ratio between fertilizer cost and corn price has been relatively stable over the years (see figure below). We updated the MM section to reflect better the cost of fertilizer and corn selling price and their influence on the optimum N rate as also suggested by other reviewers. We also added a new suppl figure (Fig. S7). Please also below in which we elaborate more on the cost of fertilizer (3rd comment).

Small plot research to determine fertilizer response usually has large year to year variations. Making the use of mean response over years as a recommendation for future yields almost sure not to be the true optimum for the year the fertilizer is applied because the weather and soil water dynamics are never repeated, thus resulting in the year-to-year variability in response as demonstrated. Using mean response recommendations implies that there is no interest in risk, and practically no farmer is willing to bear much risk at all. There is usually a yield goal involved in the decision and experience from past years is usually factored into the rates the farmers use, although modern farmers use approximately what the state extension specialists suggest. However, farmers now practically all use yield mapping to obtain the spatial variation of the yield, and it should be obvious that the optimum rate for the various yields is not the same in space and for real nitrogen loss reductions, the yield potential of the spaces in the field should have appropriate N rates for the yield variations.

Response: The reviewer brings up good points, which we agree with, and in the discussion section we already touch on many of those (e.g., yield as the driver for N-rate, variability in N rate across space, etc.). We fully agree that more research is needed on spatial variability, and we believe emerging technologies, such as remote sensing, may help in that respect; however, addressing spatial variability aspects is beyond the scope of this manuscript.

You demonstrate in the paper that APSIM (as calibrated) gave good results compared with measurements. Why not just use the simulation to determine the temporal change in yields and N inputs? It would be much less expensive, and it was one of the main purposes for developing such a robust model such as APSIM. You did not mention in the paper how APSIM was calibrated, what was changed besides N input to result in the temporal yield increase. I assume that it was related to plant population increases, better genetic materials and possibly a longer growing season with earlier sowing dates. Is there any experimental evidence that the N loses as simulated are correct? These details should be included in the paper.

Response: In this study we used APSIM to get insight into variables such as N loss and N mineralization (Fig. 4) that were not measured in the long-term experiments but are important to explain temporal trends in optimum N rate. We were able to simulate long-term grain yield increases by altering cultivar parameters and management inputs as described by Baum et al. (2023), but the accuracy in estimating the optimum N rate was not as good as the yield prediction because small over/underestimation in the yields can result in large over/underestimation of the optimum N rate, and thus we used original experimental data to develop temporal trends for the optimum N-rate. The capacity of APSIM to simulate N leaching has been verified in previous studies in the Corn Belt and thus we feel confident that the simulated results are correct.

In the MM section, we added the following text to support this "We simulated N loss as the amount of NO3- moving below the rooting zone (150 cm depth) and N2O emitted to the atmosphere per year, location, rotation and N-rate. Previous research in the US Midwest has shown good model agreement with simulated NO3- leaching from 56 site-years of data sourced from artificially subsurface-drained field experiments across the US Midwest (Pasley et al., 2022). Additionally, APSIM has been shown to simulate well N2O emissions from various N rates in maize-based cropping systems (Li et al., 2021). Furthermore, our N2O and NO3- estimates align with previously established efficiency factors. APSIM simulated N2O at an average efficiency factor of 2.14% of N2O per kg N applied, which is within an

established 1-3% efficiency factor of observed values (Bouwman & Boumans, 2002; Nash et al., 2015; Hergoualc'h et al., 2021)."

For the economic analysis, a constant was used for the prices of fertilizer and grain (line 285:5.6:1 N fertilizer: maize price (US\$ 0.88 kg-1: US\$ 0.16 kg-1)). More documentation is needed that the assumption is appropriate and that one price did not increase more than the other during the time of analysis, since this is a time trend paper.

Response: Thanks. We added more documentation in the MM as suggested and a supporting figure to explain the reasoning of using a constant price ratio. "While the N-fertilizer price and the corn price have changed over the years, the price ratio has remained fairly constant (Fig. S7) and therefore we used a constant price ratio in our analysis similar to previous studies exploring temporal dynamics in EONR (Cerrato and Blackmer, 1990; Sawyer et al., 2006; Clark et al., 2019). Furthermore, we performed a sensitivity analysis in which we changed the price ratio by 22% (from 4.3 to 6.8) and found that the 1999-2021 median EONR to change only by $\pm 2\%$, which is well below the variability in the optimum N rate obtained across sites (31%) and years (30%). For these reasons, we did not consider year-specific price values. "



Fig S7. The United States Department of Agriculture estimates for annual maize grain and anhydrous ammonia fertilizer prices. Scales maize grain price and anhydrous ammonia were adjusted by multiplying the maize grain price by the price ratio of 5.6 (price of anhydrous/grain).

Line 91 Close parenthesis not necessary

Response: Completed, thanks for the suggestion.

Reviewer #2:

Remarks to the Author:

This paper used long-term and short-term nitrogen experiments combined with crop growth modeling simulation to analyze optimal N rates (AONR, EONR and EnvONR), and drew the conclusion that EONR for US maize production has increased by 2.6 kg ha-1 year-1 from 1991 to 2021 (or 1.2% per year). They contributed this increase to increasing grain yield and N losses. They also showed that reducing ONR to account for the social cost of environmental nitrogen losses will reduce US maize productivity by 6% while having a minimal impact on nitrogen loss reduction.

I have several concerns for the study:

1. Using long-term experiments for this analysis has some limitations. The long-term experiments have the treatments at fixed locations, and there is an accumulative or carry over effect. This is different from real world production conditions, and the results may not be representative of real world conditions.

Response: We are aware of this limitation, and we took measures to minimize this concern by adding single-year, on-farm experiments to our analysis (Fig. 4). We also added text in the introduction to highlight this further "While long-term experiments are valuable resources for assessing the performance of alternative N management practices (Van Grinsven et al., 2013), they are also prone to exaggerate maize's yield response to N compared to production fields as both low and high N treatments reach a steady-state due to consistent over-under fertilization (Van Grinsven et al., 2022). To enhance our analysis towards creating a robust assessment of the temporal trends in EONR, we sourced additional single-year EONR data from the Corn N rate calculator (n=176 EONR values, Sawyer et al., 2006; Nafziger et al., 2022)."

2. The study focused on EONR, and used a fixed historical N fertilizer and maize prices (\$0.88 kg-1 and \$0.16 kg-1). The prices are changing a lot and the results based on fixed historical prices over a period of 20 years may not represent real world conditions.

Response: We agree that the prices change but the ratio remains relatively stable (see fig above). See also our response to reviewer 1. Historically, the cost of fertilizer and the selling price of corn follows the same trends over the years and the ratio remains constant.

3. The study only considered one-time preplant nitrogen application, which has been proved to have high risks of nitrogen losses if there is more rainfall in the spring. Many farmers in such regions are applying nitrogen fertilizers in splits, which can increase nitrogen use efficiency and reduce nitrogen losses. But this was not considered in this study.

Response: This is a good point, but it is not the case for our study region according to data from Cao et al. (2018), who found that a single application timing in either the fall or the spring was the predominate N application method over the last years in Iowa and Illinois. We added the following text in MM "Experiments followed the predominate farming practices within their surrounding regions, therefore, N fertilizer was applied as a single application roughly ±2 weeks of maize planting (Cao et al., 2018)."

4. The long-term experiments in Iowa received zero nitrogen fertilizer in all plots in 2017 and 2018,

which were excluded from the analysis, and the experimentation resumed under the same design in 2019. This could lead to higher EONR in 2019 and 2020, which could influence the long-term trend.

Response: To minimize this concern, we reran the analysis by excluding 2019 and updated the graphs. The results were affected very little by this update (please see updated figs).

5. Year-to-year weather variability has a significant impact on optimal nitrogen rates. Therefore, rather than keep increasing nitrogen application rates, it is more important to dynamically adjust optimal N rates according to different weather conditions.

Response: We fully agree with the reviewer's observations, and in our paper, we tried to highlight the year-to-year variability in Fig 2 by showing the box plots. Dynamically adjusting the optimum N rate is one of the conclusions of our manuscript, please see the last paragraph of the discussion.

6. The study is based on small plot experiments, which have limited spatial variability. In commercial production, there is large field-to-field and within-field spatial variability in soil-landscape conditions and optimal nitrogen rates, which was not considered in this study.

Response: We agree with his observation, and this extends to all field experiments. While we would love to have data by field zones to capture spatial variability, that was not possible in this research. Despite the small plots, our overall effort captured over 30 or more soil types, and represented well major corn production environments in the US Corn Belt.

7. How EnvONR was calculated is not clear. EnvONR was defined as the minimum N rate required to maximize environmental performance by considering the cost of N2O and NO3- leaching, added to N fertilizer cost. How to maximize environmental performance? It will be maximized if no N is applied, but that is obviously not the option. You provided two references, but it is better to provide some explanation in this manuscript, so readers don't have to read the references to understand it.

Response: We expanded the definition of the EnvONR in Fig 1 legend to make this clear. Basically, we calculated the EnvONR by accounting for both costs: N fertilizer + environmental cost, which was further clarified in the MM "The cost associated with the damages of NO_3^- leaching to the environment (\$41.15 kg⁻¹ N) was taken from Sobota et al. (2015), which includes many categories with the most important to be freshwater eutrophication, ground water N loading and the increased risk of colon cancer, nitrate contamination, damages of declining fisheries and degradation of recreational area. Given that this cost value is not yet established in the market, we performed a sensitivity analysis (\$1 to 70 ha⁻¹) to better understand its impacts (Fig. S4)"

8. The real challenge of N management is the large spatial and temporal variability in optimal N rates. How to determine the optimal N rate at the beginning of the growing season if you apply all the N before planting? Different weather conditions will result in very different optimal N rates. Also commercial fields have large within-field variability in optimal N rates. A uniform N rate across the field will unavoidably result in over- and under-applications in different parts of the field, no matter what optimal N rate you use. Response: Good points, fully agree. Expanding further on these points is beyond the scope of this paper neither we have the data to support such analyses. In our conclusion, we mention the need to improve predictability across time and space, in line with these comments.

9. It is important to know what the N rates most farmers are using, how are those farmer N rates compared with EONR?

Response: Thanks, we added the following text in the discussion to address reviewer's question "In this survey, maize producers in the US Midwest applied 207 to 280 kg N ha-1 to continuous maize and 186 to 235 kg N ha-1 to maize following soybeans. These ranges are comparable with the ranges reported in Fig. 2, but the mean values in the survey appear to be higher than the reported optimum N rates in Fig. 3. From this comparison, we cannot state that farmers overapply N fertilizer because these are two different databases with different fields, weather-years, management practices, and hybrids, but the increasing N-rate trend in the survey suggest that maize producers are adapting. Our results provide strong support for this increasing trend."

Specific comments:

1. Line 87: Lower grain N concentration will reduce grain N demand, increase N use efficiency, and thus will decrease the optimum N rate, I don't think it will increase optimum N rate.

Response: We agree, the sentence was updated to "will decrease the optimum N rate".

2. Line 91: no) after production.

Response: done, thank you

3. Line 104: -105: ...for the cost of environmental N losses 2... their costs.

Response: done, thanks.

4. Line 141-142: This confirm the importance of split N application, rather than applying all N in the spring before planting.

Response: We agree with the reviewer that split application will increase the fertilizer use efficiency of cropping systems. We added the following text in the discussion "Additionally, alterations in N management practices may reduce the environmental impact of N application. Split N application can reduce fertilizer-N losses, especially compared to a single fall application, while increasing fertilizer use efficiency to the environment by better synchronizing fertilizer N with crop demand (Gentry et al., 2023)."

5. Line 145: ((yield at EONR-yield at zero N)/EONR).

Response: Corrected clarification.

6. Line 147-151: A little confusing. You said the grain N concentration did not change over the study period but you also said it suggests a dilution of N in the grain over time.

Response: Thanks, we fixed that, the updated text reads "The amount of N exported at crop harvest, *i.e.*, grain yield x N concentration, increased by 0.73% per year and had a r=0.24 correlation with EONR, despite a slight dilution of the grain N concentration over the study period (Fig. 4H; Table S2)."

7.Line 170: Furthermore, using APSIM simulated N loss data. This is not a complete sentence.

Response: Thanks, fixed.

8. You have defined EONR, but you still use economic optimum N rate.

Response: that is correct, we try to minimize the use of acronyms within the paper to increase readability

9. Line 194: This assumption may not be true.

Response: we agree and there we changed the "will" to "may".

10. Line 265: Using grain yield at 0% moisture is not the common practice. Generally, grain yield is reported at 15.5% moisture. This can lead to different results.

Response: Corrected.

11. Line 165-266: The maximum N rates may not be high enough.

Response: Given the current results, we agree, especially in years where the optimum N rate exceeded the measured rate. However, when the experiments were designed a rate of 268 kg N/ha was thought to far exceed the optimum N rate. Ongoing research has been adjusted to account for higher N rates.

12. Line 321-328: Did you use EONR or MRTN?

Response: Functionally they are the same. We removed the "MRTN" abbreviation to avoid any confusion between the two.

Reviewer #3:

Remarks to the Author:

This article conducts a thorough analysis of the temporal patterns in the optimum nitrogen (N) rates for maize production in the US Midwest from 1991 to 2021. The study reveals an increasing trend in these optimum N rates over the specified period and strives to elucidate potential causative factors for this phenomenon. The authors highlight the notable trade-off between reduced fertilizer use, which could

mitigate environmental pollution, and the substantial yield reductions associated with such reductions, deeming it an undesirable outcome. The findings of this study present interesting insights that could offer valuable recommendations for guiding fertilizer use practices in the United States.

Moreover, the results contribute meaningful insights into determining optimal fertilizer application rates in various countries, rendering the article worthy of publication. Despite the merits, certain aspects of the article require substantial revisions before publication. The following comments outline specific areas that need attention and refinement

Response: We thank the reviewer for the positive comments on the manuscript.

Please clarify whether the agronomic optimum N rate depicted in Fig.1 will eventually converge to parallel lines rather than decline. I think that more nitrogen fertilizer input doesn't necessarily lead to an unlimited and linear increase in yield.

Response: The theoretical yield response to N rate should be quadratic, and yields will eventually decline at extremely high N rates, however, in real-life experiments, we barely see such quadratic responses because we don't apply too much excessive N rate (>400 kg N/ha). In our study, the quadratic-plateau model was the best fitting model in about 340 out of the 386 N-trials (=88% of the cases), and thus was used this model in conceptual figure 1 to best represent reality.

The explanation in lines 84-87 regarding the reasons for the increase in the optimum N rate may need reconsideration. It's essential to clarify whether these factors are simultaneous or if there exists an actual causal relationship.

Response: Thanks. The factors are simultaneous, and we revised the text in this paragraph to better communicate this "... These changes are simultaneous and currently remains unknown how these factors have interacted and affected the optimum N rates for maize over decades of farming..."

Similarly, reconsider and provide an explanation on whether a higher optimal nitrogen input in a crop rotation system can be directly equated to a higher efficiency of its utilization.

Response: Thanks for catching this. We fixed the sentence to "Lower grain N concentration will reduce grain N demand and optimum N rate while increasing N use efficiency (Sinclair and Rufty, 2012; Mueller et al., 2019; Tenorio et al., 2020)".

Please clarify why a reduction in the optimal N rate (from economic to environmental) leads to a nonlinear decrease in grain productivity. Consider whether this should be attributed to the actual applied N rate and provide a more detailed explanation for the observed trend.

Response: It is because of the shape of yield response to N-rate (see fig 1 - concept). Decreasing the N rate from the optimum N-rate decreases the yield, and the yield reduction becomes bigger as we move further from the optimum N-rate. We added this clarification in the results section.

Rethink the placement of important graphs that reveal key insights in the article. Consider whether they should be included in the main body or placed in the extended data.

Response: We feel the figures included in the manuscript are sufficient to support the conclusions and convey a clear message, however, we remain open to specific suggestions to alter as needed.

Some detailed suggestion as following:

Line 38: As mentioned earlier, please reconsider the accuracy of using "this was attributed to increasing grain yields and N losses."

Response: we changed that to "simultaneously with..."

Lines 113-116: Could you explain the rationale behind the statement, "the increase in the economic optimum N rate was 33% greater in the maize-soybean rotation compared to the continuous maize system," indicating "the well-known difference in the economic optimum N rate between crop rotations is closing"? The relations seem unclear.

Response: We added the following explanation "This is likely because of the greater increase in grain yield in the maize-soybean system compared to maize-maize." Please see further explanations in the 4th paragraph of the discussion. The rationale behind this statement is that N-rate recommendation for corn following soybean considers on average 50 lbs N / acre less than corn following corn (Pedersen and Lauer, 2002; Gentry et al., 2013; Puntel et al., 2016, 2019; Struffert et al., 2016; Bowles et al., 2020). This is also included in university extension recommendations to the farmers. Our research shows that the difference in optimum N-rate between rotations is getting lower year by year, which is a novel finding previously unknown.

Line 125: Please review the use of "however."

Response: Thanks, changed to "Additionally"

Line 135: Is it referring to R2?

Response: The reported "r" correlation is the square root of R2.

Line 188: Please complete the sentence.

Response: done, thanks for catching this.

Reviewer #4:

Remarks to the Author:

This study hypothesized that over the last decades, the optimum N rate for maize production) is increasing because N outputs to grain and the environment exceed the reduction in grain N concentration, and investigated the three forms of optimum nitrogen fertilizer rate for maize. They call for improved and timely predictability of the optimum N rate across space and time as a solution to

maximizing productivity, sustainability. The paper has a good logical flow and combined results from long-term experiments and database from short-term experiments. Here are two major concerns regarding methods and implication of the research.

Response: We thank the reviewer for the positive comments on the manuscript.

1) The authors need to crystalize how the agronomic optimum N rate (AONR), economic optimum N rate (EONR), and environmental optimum N rate (EnvONR) were obtained in the study.

Response: Done, please see updated Fig 1 with improved definitions of the optimums.

L280-283: For each yield response to N rate (n=386 combinations of locations and years and rotations), we fitted four descriptive models: quadratic plateau, quadratic, linear-plateau, and linear (Baum et al., 2023). The AONR was calculated from the best-fit model, and it was the inflection point for the quadratic-plateau, linear-plateau, and quadratic models and the maximum N-rate applied for the linear model. In previous research, the correlationship between yield and N application rate are always descript by quadratic equation, since the yield is always enhanced and then keep consistent or decrease a little bit, with the increasing N fertilizer. Here, the authors used four equations to show their relationship. I doubt about that which one could reflect the real relationship between them? We cannot judge it just according to the result from statistical analysis (for example R2), if it is the "best-fit model". We could get the optimum rate with scientific theory but the statistical analysis. Moreover, what is the standard for the "best-fit model". It will help the readers to understand it to show the figures.

Response: We understand the reviewer's point about the use of a single model instead of a multi-model approach. In this study we used the multi-model approach for two reasons 1) most used in the literature to estimate optimum N rate (Miguez and Poffenbarger, 2022; Cerrato and Blackmer, 1990; Bielenberg et al., 2023; Clark et al., 2019; Puntel et al., 2016; Baum et al., 2023) and 2) standard approach used in the N-rate calculator, a regional tool developed from six University Extension teams for maize N rate recommendations in the US Corn Belt. To address the very important point made by the reviewer, we ran an additional analysis with a single model, only the quadratic plateau because this is the predominant model among the 4 options (88% of the cases). We found no statistical difference in the slopes between the two approaches, please see the figure below, therefore we decided to keep the original analysis in the paper. Furthermore, we incorporated this extra analysis in the supp materials, together with the equations used. Please see new Supplement table (Table S4).



For the linear model, the maximum-N rate was chosen. It means that the designed N fertilizer rate might be low and cannot reach the inflection point, and the yield might increase continuously if more N fertilizer is supplied. So the AONR might be higher than the maximum N-rate in this experiment. Thus, the authors need to demonstrate the relationship between the yield and N fertilizer rate, and a reasonable model to descript it, obtain the optimum N rate.

Response: We agree with the reviewer about the limitations of the linear model and the assumptions regarding the optimum N rate. However, here we kept our analysis as consistent as possible with standard methods used in the N rate calculator and literature papers (Miguez and Poffenbarger, 2022; Cerrato and Blackmer, 1990; Bielenberg et al., 2023; Clark et al., 2019; Puntel et al., 2016; Baum et al., 2023) so our results can be comparable.

Furthermore, to address the reviewer's concern, we performed an additional analysis (see graph below). We found that the use of a quadratic model to extrapolate beyond the known (field applied) N rate to derive the optimum N rate is very risky, and the results may be unrealistic (e.g., optimum N rate of 500 kg N/ha), so we added the below figure to supplemental (Fig S6) materials to demonstrate this risk. Also, it should be noted here that the linear model provided the best fit among the 4 models in only 5% of the cases, which is a small number to have any significant effect on our results.

We made the following updates in the manuscript in response to reviewer's comment:

- Added the following text in the MM "To avoid extremely high values of the convergence point, potentially occurring when the optimal N rate exceeded the maximum N rate (i.e., 252 and 268 kg N ha-1 for the Illinois and Iowa locations), it was set to be the maximum N rate to avoid the uncertainty of extrapolating results beyond the tested range of N rates (Fig. S6)."
- 2. Add a new supplementary figure to show the result of the extrapolation (fig below)



Fig legend: Conceptual figure depicting two examples of the risk in estimating the agronomic optimum N rate when extrapolating yield response functions past the known N rates. The vertical and horizontal dashed lines represent the agronomic optimum N rate and the yield associated with them used in this study (blue line) and extrapolated (red line).

2) Research work concluded that "We call for improved and timely predictability of the optimum N rate across space and time as a solution to maximizing productivity, sustainability, and profitability". Actually, the optimum N rate was referred in some reports from time to time in the 20-30 years, so I don't think it's enough to call for optimum N rate across time, but it will be helpful if the optimum N rate could be listed in across time. It's not necessary to be in the US Midwest, it could happened in other region or country, but could testify the result of this research.

Response: We couldn't understand the reviewer's point here. Our conclusion is for improved predictability of optimum N rate across both time and space.

Specific Comments:

L51-52, "the use of N fertilizer has increased 5-fold from 1961 to 2010". The N fertilizer amount could be updated to recent years, since 2010 is already more than ten years ago.

Response: Updated, thanks for the suggestion

L36, n=562, it is the number of short term or total of long and short-term experiments. It should be write separately.

Response: Done, thanks

L36-37, the economic Opt N increased by 2.6 from 1991 to 2021, then how about the agronomy and Environ-Opt. N?

Response: All of them increased but we are limited by space in the abstract (max 150 words) to add all the numbers and all opt N rate definitions. We used the most cost term in the abstract.

L38, "reducing optimum N fertilizer rates", what is that?

Response: We updated the sentence to read "Furthermore, we provide evidence that reducing N rates below the economic optimum based on the societal cost of N losses will reduce US maize productivity by 6% while having a minimal impact on N loss reduction" To avoid confusion with differing optimal N rates in the abstract

L85-87, "Lower grain N concentration will reduce grain N demand while increasing N use efficiency (Sinclair and Rufty, 2012; Mueller et al., 2019; Tenorio et al., 2020) and thus will increase the optimum N rate.". It will be better if it could be explained that why the reduce gain N demand lead to increased NUE?

Response: Thanks, the sentence was rewritten to "Lower grain N concentration will reduce grain N demand and optimum N rate due to the plant sequestering more carbon per unit of N uptake ... "

Section "Quantifying the tradeoff between productivity and N fertilizer reductions". The research established the relationship between yield and N fertilizer reduction. However, the N fertilizer reduction is the defined as the difference value from economic to environmental optimum N rate, which is always lower than the application rate by farmers. So is it representative? The concept of insert figure in Fig.1 is great. It it could be quantified according to the result in this research, it will improve the novelty of this research.

Response: The reviewer brings a good point here that farmers, in some cases over-apply N-fert, above the EONR so a reduction of 40 lbs N/ac will not decrease yields because we are well above the EONR assuming farmers are over-applying. We agree, and we added the following text in the manuscript "While our results indicate increases in the optimal N rate over time, not every situation will call for applying more N, especially in fields that currently receive N fertilizer rate in excess of the optimum rate" in the discussion to depict that. Still the tradeoff graph has merit as it provides a benchmarking framework for decision makers to demonstrate that lowering N fertilization below the optimum will harm crop production.

The increased optimum N rate might be attributed to the increasing yield, because of the variety breeding, irrigation technique, or cultivation, and so on. So it should be discussed enough.

Response: Agreed – we added a sentence "The increase in the economic optimum N rate was in part explained by increasing grain yields, which in turn is caused by improved genetics, improved agronomic management, and environmental conditions (Messina et al., 2022; Rizzo et al., 2022; King et al., 2024)." to acknowledge this, however, getting deeper into the relative contribution of each factor to the grain yield increase is beyond the scope.

Response to reviewers' comments

Dear Editor, we thank you for the opportunity to revise our manuscript and re-submit. We addressed each reviewer's comment (see detailed responses below), and the manuscript has considerably improved. The major changes made are listed below:

- 1. Updated abstract, including the concluding statement as suggested
- 2. Add new text in the Results and Discussion for the different rates of increase in AONR, EONR, and EnvONR
- 3. Expanded text on crop rotation impact
- 4. Provided further evidence for the relative change in yield and relative grain N conc

Please see our response to each reviewer's comment below

Thank you.

REVIEWER COMMENTS

Reviewer #4 (Remarks to the Author):

The authors have figured out my concerns in the revision. Here, I would like to suggest that the difference between the continuous maize system and maize-soybean system should be enough discussed, since the optimum nitrogen fertilizer rate for them was different, especially the tradeoff in Fig.5.

Response: Thank you. As suggested, we added more text regarding the difference in optimum N rates between crop rotations.

Within the 2nd paragraph of the Introduction section we added ".....Typically, maize following soybean requires 40 kg N ha⁻¹ less N-fertilizer rate than maize-maize (Sawyer et al., 2006; Baum et al., 2023) because of the increased soil N mineralization caused by the amount and quality of the soybean residue (Green and Blackmer, 1995; Ruiz et al., 2024)." In the discussion section we added "....The reduction in maize yields would be greater on the continuous maize system because this system has a higher EONR and, hence, higher N losses than the maize-soybean system (Figs. 2&S5). Furthermore, planting soybeans prior to maize favors soil N mineralization and higher yields under zero N (Figs. 4b&f), which partially alleviates N fertilizer demand. For these reasons, there is a steeper yield penalty for underfertilizing a continuous maize system (Fig. 5) combined with a greater proportion of N losses (Fig. S5).".

Reviewer #5 (Remarks to the Author):

Baum et al combined data from long-term and short-term nitrogen experiments combined with the crop growth modeling simulation model APSIM to assess trends in agronomic, economic and environmental optimal N rates (AONR, EONR and EnvONR) over the period 1991-2021. As expected agronomic optimal N rates, AONR, > economic optimal N rates, EONR > environmental optimal N rates, EnvONR. They showing that the increase in EONR is higher than AONR and EnvONR implying that the difference between agronomic and economic optimal N rates decreased over time while the difference in economic and environmental optimal N rates increased over time. They then show that reducing N rates from the economic to the environmental optimum based reduce maize productivity by 6% while the N loss reduction is small. In general I find the paper interesting and their replies to the comments adequate. I have still some additional main comments and specific comments that are relevant in my view

Response: Thank you for finding our manuscript interesting and the very helpful suggestions.

Main comments

The abstract is not very informative and does not convey well the main message of this paper. I would think that the main message is that

• the difference between agronomic and economic optimal N rates decreased over time while the difference in economic and environmental optimal N rates increased over time.

• The reduction in N rates from the economic to the environmental optimal N rate comes at a cost for yield at low N loss reduction

The first point is not mentioned and the last point comes with nuances due to two aspects

Response: Thank you for the GREAT suggestions. We incorporated them in the revised manuscript. First, we updated the abstract by adding the following text "...By accounting for societal cost estimates for N losses, we estimate an environmental optimum N rate, which was also increased over time but at a lower rate than EONR. Furthermore, we provide evidence that reducing N rates from economic to environmental optimum N rate could reduce US maize productivity by 6% while slightly reducing N losses. Thus, we call for enhanced assessments and predictability of the economic and environmental optimum N rate to meet rising maize production while avoiding unnecessary N losses.".

Second, we added new text in the Results section to expand on the AONR, EONR, and EnvONR differences over time "....The different slopes indicate that the difference between EONR and AONR has decreased over time, while the difference between EONR and EnvONR has increased over time. Across rotations, the difference between AONR and EONR was 10 kg N/ha in 2020 compared to 22 kg N/ha in 2000, while the difference between EONR and EnvONR was 70 kg N/ha in 2020 compared to 46 kg N/ha in 2000. While the EONR can never equal the AONR due to fertilizer cost, the difference between EONR and EnvONR may shift in magnitude and direction depending on the societal cost of N losses."

Third, we added a new paragraph in the discussion to explain the reason for the different rates "The difference between the AONR and EONR decreased over time by 54%, from 22 to 10 kg N ha⁻¹ (Fig. 2), primarily due to grain yield increase and NUE improvement (Fig. S10). While the two optimal N rates will never be equal as long as there is an expense for fertilizer cost, it is encouraging that there is a growing return to N as N rates approach the AONR. Typically, N rates between the AONR and EONR have reduced NUE and profits (Nafziger et al., 2022). Conversely, we found that the difference between the EONR and EnvONR has increased by 34% (Fig. 2), indicating that EnvONR is increasing at a slower rate than EONR.

This is due to the increase in N loss (Fig. 4) and the EONR (Fig. S11) assuming the fixed cost for N losses. The mean difference between the EnvONR and EONR was 27% (Fig. S2), within the range reported in the literature (from 17 to 41%; Wang et al., 2014; Gourevitch et al., 2018; Martinez-Feria et al., 2018). While the differences between the EONR and EnvONR will, in part, be determined by uncertainty in the cost of N losses, our results suggest this difference will continue to grow as N losses continue to increase." Lastly, we added two new supplementary material figures, also based below.



Fig S6. Comparison of factors influencing the difference between the agronomic and economic optimum N rate (AONR and EONR, respectively) per crop rotation. These factors include yield at the AONR (top left panel), yield at zero nitrogen application (top right panel), the EONR (bottom left panel), and the fertilizer use efficiency (bottom right panel).



Fig S7. Comparison of factors influencing the difference between the economic and environmental optimum N rate (EONR and EnvONR, respectively) per crop rotation. These factors include the yield at the agronomic optimum N rate (top left panel), yield at zero N application (top right panel), EONR (middle left panel), fertilizer use efficiency (middle right panel), the sum of N losses at the EONR (i.e., nitrate leaching and N₂O emissions) (bottom left panel), and social cost of N losses (bottom right panel).

• this low N loss reduction is related to the assumed high societal costs of N losses. To me this part is not really so clear. Note e.g. that cost of huma health related to contaminated drinking water only play a role at high NO3 levels (above > 50 mg NO3/I) and it EONR prevents sch levels, further reductions do not imply any reduction in costs related to human health improvements. The point that I like to make is that the N loss reduction might be low but if the societal costs are really so high, it could be worth it but if it is an overestimate, the EnvONR would come much close to the EONR and the effect of reducing EONR to EnvONR on crop yield would be much less

Response: The N loss reduction amount has more to do with the limited amount of N that is lost below the EONR (see Fig. S5) than with the N loss cost. To clarify this further, we added a clarification in M&M under the optimum N rate calculation section: "....A higher cost of N losses will favor larger discrepancies between EnvONR and EONR." Also in the discussion section we added "...with lower social cost reducing the difference between the EnvONR and EONR, thus having a lower yield reduction between the two rates and vice versa".

• this low N loss reduction is based on the current NUE. The point of reconciling crop yield with environmental impacts has of course to do with increasing the NUE up to an attainable level (see e.g. Schulte-Uebbing and de Vries, 2021; Chang et al., 2021). I would discuss both aspects in the paper and shortly summarize in the abstract

Response: Thanks for the suggestion, and the references. We expanded on the improvement of nitrogen use efficiency for improved food security.

We added the following text to the discussion section ".....However, N losses in some hotspot regions exceed environmental thresholds, beyond which can be mitigated through technological improvements (Schulte-Uebbing and de Vries, 2021). In such regions, other strategies must be utilized to mitigate environmental damage, such as redistribution of N inputs to more suitable production areas or alleviating crop demand by reducing food waste......." We also added ".....A solution proposed by Chang et al. (2021) suggests sifting the international focus from the demand-side of global markets to the supply-side by setting goals for regional improvement in nitrogen use efficiency. They found improved nitrogen use efficiency to be the most effective strategy for improving food security, while considering N boundaries. We call for enhanced assessments of the environmental optimal N rate and increasing nitrogen use efficiency strategies considering the need to maintain or even increase the current crop yields.....".

NB: the extra space needed for this can come from removing the text "We call for improved and timely predictability of the optimum N rate across space and time as a solution to maximizing productivity, sustainability, and profitability. Present findings serve as a benchmark to guide future N fertilizer rate research, recommendations, and policy". This is vague and not well related to the results. I can imagine that some text on the implications is needed and I would then rather say: "we call for an enhanced assessments of the environmental optimal N rate (as that part is most unreliable based on societal costs) to assess the need increase in N use efficiency considering the need to maintain or even increase the current crop yields"

Response: Thank you

References

Schulte-Uebbing L.F. and W. de Vries, 2021. Reconciling food production and environmental boundaries for agricultural nitrogen inputs in the European Union. Science of the total Environment 786 (2021) 147427. <u>https://doi.org/10.1016/j.scitotenv.2021.147427</u>

Chang, J., P. Havlík, D. Leclère, W. de Vries, H. Valin, A. Deppermann, T. Hasegawa and M. Obersteiner, 2021. Global food security risks associated with meeting regional nitrogen boundaries. Nature Food 2: 700–711. <u>https://doi.org/10.1038/s43016-021-00366-x</u>

Response: Thank you. The suggested references have been cited and greatly improved the discussion section and the message.

Specific comments

Line 70-71: Based on results of Wang et al., 2014; Martinez-Feria et al., 2018, the authors state that the EnvONR is typically lower than the AONR at a rate of 35-41%. I assume they mean that EnvONR is typically 35-41% lower than the AONR. I would then rather think that it is logic to give such results also for this study and then how the %difference changes in time from 1991-2021. And do this also for EONR. In general the paper does not give enough attention to the changing differences between the various rates in time that I see as a a main result (see comment above).

Response: Thanks, please see our response to the first comments. We added new text for the different rates in Results ".....The different slopes indicate that the difference between EONR and AONR has decreased over time, while the difference between EONR and EnvONR has increased over time. Across rotations, the difference between AONR and EONR was 10 kg N ha-1 in 2020 compared to 22 kg N ha-1 in 2000, while the difference between EONR and EnvONR was 70 kg N ha-1 in 2020 compared to 46 kg N ha-1 in 2000. While the EONR can never equal the AONR due to fertilizer cost, the differences between EONR and EnvONR may shift in magnitude and direction depending on the societal cost of N losses."

Furthermore, we added, "The difference between the AONR and EONR decreased over time by 54%, from 22 to 10 kg N ha-1 (Fig. 2), primarily due to grain yield increase and nitrogen use efficiency improvement (Fig. S6). While the two optimal N rates will never be equal as long as there is an expense for fertilizer cost, it is encouraging that there is a growing return to N as N rates approach the AONR. Typically, N rates between the AONR and EONR have reduced nitrogen use efficiency and profits (Nafziger et al., 2022). Conversely, we found that the difference between the EONR and ENOR has increased by 34% (Fig. 2), indicating that EnvONR is increasing at a slower rate than EONR. This is due to the increase in N losses (Fig. 4) and the EONR (Fig. S7) assuming a fixed cost for N losses. The mean difference between the EnvONR and EONR was 27%, within the range reported in the literature (from 17 to 41%; Wang et al., 2014; Gourevitch et al., 2018; Martinez-Feria et al., 2018). While the differences between the EONR and EnvONR will, in part, be determined by uncertainty in the cost of N losses, our results suggest this difference will continue to grow as N losses continue to increase."

Line 74-76: The authors state here that. "A summary of the potential cost of N loss can be found in Sobota et al. (2015), which attributes cost ranges associated with eutrophication, the increased health risk from human consumption, and nitrous oxide and NOX emissions to the atmosphere". To me this is not clear. To start with, NOx emissions from agriculture enhance the concentrations of particulate matter

affecting human health but these emissions are limited. However, this is not true for NH3 emissions and both NOx and Nh3 enhance the concentrations of particulate matter affecting human health (see e.g. an overview of N impacts by de Vries, 2021). Why is NH3 emissions then not included in the assessment. Second, the increased risk of colon cancer due to nitrate contamination only occurs above a threshold level near 10 mg NO3-N/l and is that reached (see comments above)?

De Vries, W., 2021. Impacts of nitrogen emissions on ecosystems and human health: a mini review. Current Opinion in Environmental Science & Health 100249. https://doi.org/10.1016/j.coesh.2021.100249

Response: Our results don't reflect the watershed level, and it is therefore impossible for us to know the nitrate concentrations in the water. However, a report by the Iowa Environmental Council has shown that over 12,000 private wells in rural areas across Iowa (i.e., the main maize producing state in the US) have a nitrate concentration exceeding 5mg/l. Therefore, any additional leaching regardless of amount will potentially lead to increased human health concerns. Moreover, the cost associated with human health concerns is low compared to the ecological cost associated with eutrophication, and degrading ecosystem services. To address this, we added "A summary of the potential cost of N loss can be found in Sobota et al. (2015), which attributes cost ranges associated with eutrophication, the increased **potential** health risk from human consumption, and nitrous oxide and NOX emissions to the atmosphere"

In terms of NH3 volatilization from inorganic fertilizer, we didn't include it because the amount is very small given current maize management practices in the US Midwest (i.e., N-fertilizer is injected in the soil and includes an inhibitor). However, we added "Note we exclude the potential cost of particulate matter PM10 and PM2.5 from for the estimation of the EnvONR in this analysis due to inconstancies within the literature about the overall impact on human health from inorganic nitrogen fertilizers (Kelly et al., 2004). As well as the impact of NH3 volatilization from inorganic fertilizers, because NH3 volatilization is marginal within the N management practices used for maize production in the US Midwest (Woodley et al., 2020)." in the materials and methods to express this exclusion from our analysis.

Line 97-98. The authors hypothesize that over the last decades, the optimum N rate for maize production is increasing because N outputs to grain and the environment exceed the reduction in grain N concentration. To me this is not clear since the reduction in grain N concentration affects the N outputs to grain being the product of grain yield and N concentration. Do you now say that you expect that the increase in growth rate by roughly 1.2% per year is likely higher than the reduction in the concentration of grain N?. But can you not show this directly from data on trends in grain N? Or do you now know this and thus link it to the sum of N out and N leaching?

Response: yes, the relative change in grain yield is 3-fold higher than the relative change in grain N conc (please see table below with associated references and calculations) according to literature data. Note that in our analysis, we did not have measured grain N conc, so we used modeling to estimate grain N conc (see our response to a following comment, below).

Category	Units	King et al.,	Duvick and	DeBruin et al.,
		2024	Cassman, 1999	2017
Study period	Years	1980 → 2020	1930 → 1990	1934 → 2013
Grain yield	Mg/ha	11.1 → 15.3	5.0 → 11.5	5.5 → 14.9

Grain N conc.	g/kg	1.45 → 1.2	1.65 → 1.41	1.64 → 1.27		
Absolute Rate of change						
Grain Yield	Kg/ha/yr	105	108	109		
Grain N conc.	g/kg/yr	-0.006	-0.004	-0.005		
Relative rate of change						
Grain yield	%/yr	0.8	1.3	1.2		
Grain N conc.	%/yr	-0.47	-0.26	-0.32		

References

-King et al., Genetic gains in short-season corn hybrids: Grain yield, yield components, and grain quality traits. *Crop Sci.* 64, 710-725 (2024).

-Duvick, D.N., Cassman, K.G., 1999. Post-green revolution trends in yield potential of temperate maize in the north-central United States. Crop Sci. 39, 1622–1630.

-DeBruin et al., Grain yield and nitrogen accumulation in maize hybrids released during 1934 to 2013 in the US Midwest. *Crop Sci.* 57, 1431-1446 (2017).

Line 103-106. The authors state that long-term experiments are prone to exaggerate maize's yield response to N as N treatments reach a steady-state due to consistent over-under fertilization. Why exaggerate? What is reaching a steady state: the soil N pool or the yield. has it to with N immobilization in short term experiments that do not occur any more after a long time period. This needs explanation.

Response: Thanks, we reworded the sentence as follows to add more clarity "......While long-term experiments are valuable resources for assessing the performance of alternative N management practices (Van Grinsven et al., 2013), they are also prone to overestimate maize's yield response to N because the zero N treatment reaches a different steady-state compared to commercial fields without legacy zero N treatments (Van Grinsven et al., 2022)".....

Line 130-131. The authors find that the increase in EONR is comparable to the maize yield increase. This leads to the question, which data were used for the grain N concentrations in the modelling. Measured data? And did they possibly stay constant over time?.

Response: The grain N concentrations were simulated using the APSIM model, thus is an emergent property of the simulation. However, the change in concentration was nonsignificant for the continuous maize rotation and very small for the maize-soybean rotation (Table S2). Therefore, as grain yields increased, the total amount of grain N exported increased as well. Please see lines 156 to 160.

Specific comments on Figures and supplementary tables

Figure 2 and Table S1.

• At the end of figure 1, I would add: The full regression equations are given in Table S1. Make the number of significant digits equals in figure (you use 3, e.g. 2.38x) and table (you use 4, e.g. 2.382x). I would for sure not use more than 3 (even that is high considering the uncertainty). Note that you do this in Figure S1, S4 and S5!

Response: Done, text added to Fig 2. We did the digit adjustment in Figure 4. However, we maintained the regression coefficient of Table S2 in its original format to not lose accuracy when equations are reproduced in Excel.

• In table S1, you need to skip "& 3". The linear equations for each optimum nitrogen (N) rate and crop rotation are related to Fig. 2 only. In fig 3, the full equations are given in the figure.

Response: Thanks for catching this. We checked and we noticed that in Fig. 3 we provide the overall regression equation (long and short term experiments together). Therefore, we added in the legend "The full regression equations for the single-year lines are given in Table S1." and maintained the "& 3" in the Table S1. Thank you.

Figure 4 and Table S2.

• At the end of figure 4, I would add: The full regression equations are given in Table S4

Response: Added, thanks.

• In both the figure and table I would use at maximum three significant digits. So 0.011; 0.110; 1.10, 11.0, 110. So change in figure a slope of 121.63 to 122 and in equation a regression of y=121.630x – 233920 to y =122x – 233920. These numbers after the comma suggest a reliability which is not there.

Response: We kept the equation coefficients in the original form because we noticed that rounding the numbers as suggested altered the results. For example, taking the above-proposed equation, we found a 7% overestimation of the yield level due to simplification of numbers. We used Excel to test this. Therefore, no changes were made.

Reviewer #6 (Remarks to the Author):

The primary purpose of this review is to check the authors' responses to the previous Reviewer #2.

The numbers below correspond to each of Reviewer #2's general comments.

1. The inclusion of the limitation flagged by this reviewer in the introduction should be sufficient to address this concern, however, it is unclear to the reader if the authors have clearly described this limitation. What is meant by "over-under fertilization"? Why does this lead to a steady state? What is meant by "production fields"? Please clarify this sentence to address this comment.

Response: Thanks, we reworded the sentence as follows to add more clarity "While long-term experiments are valuable resources for assessing the performance of alternative N management practices (Van Grinsven et al., 2013), they are also prone to overestimate maize's yield response to N because the zero N treatment reaches a different steady-state compared to commercial fields without legacy zero N treatments (Van Grinsven et al., 2022)".

2. Is this sensitivity analysis included in a supplementary file? If not, consider including. Based on the conclusions of the sensitivity analysis described by the authors, I am satisfied that using a fixed price is appropriate.

Response: Thanks, we added the below figure to the supplemental materials to portray the sensitivity analysis (please see new Fig S10).



Fig S10: One-to-one comparison of using a traditional fertilizer to grain price ratio (i.e., 5.6) to an adjusted price ratio. The solid black line represents the one-to-one line between the EONR given the traditional and the adjusted price ratios, whereas the dashed lines represent ±5% change from the one-to-one line.

3. I am satisfied the authors with the authors' response to this comment.

Response: Thank you

4. I would argue that this recalculation did change the results fairly substantially (i.e., the EONR was increased by 11% from 2.58 to 2.85), and am satisfied that these years were removed from the analysis in response to Reviewer #2's comments.

Response: Thank you

5. This concern has not been sufficiently addressed. Suggest to explicitly include mention of weather effects in the final paragraph of the discussion in response to this comment. The authors should consider including a sentence regarding the effects of weather and potentially climate change, to tie in with mentions of climate change in the introduction and the first paragraph of the discussion.

Response: Thanks, we added the following "Year-to-year weather variability is a major factor influencing the EONR (Fig. 2; Puntel et al., 2019; Baum et al., 2024); consequently, as climate change is expected to increase weather variability, annual variability of the EONR may also increase".

6. Satisfied with this response.

Response: Thank you

7. The EnvONR is sufficiently described in the materials and methods.

Response: Thank you

8. I agree with the authors' response that variable rate N application is not within the scope of this publication. However, to address this comment, an acknowledgement of this challenge could be included as a sentence in the final paragraph of the discussion.

Response: Thanks. We updated the MS by adding "To that end, improved prediction of the EONR can further enhance the effectiveness of variable rate applicators, especially in large commercial fields where a uniform EONR may not be sufficient across differing soil types." to the last paragraph of the discussion.

9. This comment has been sufficiently addressed.

Response: Thank you

Specific comment #8. Disagree with this response by the authors. Once an acronym has been defined, please use it throughout the text.

Response: Fixed. All occurrences of "economic optimum N rate" in the main text have been abbreviated after first use.

Specific comments from this reviewer: L88-89 - add 't' after 'simultaneous and'

Response: Thanks, the word "it" was inserted after 'simultaneous and'.

L102 - why introduce soil C research here? Is this important for this manuscript? Response: Thanks, we agree it's not necessary here. We removed it from the text

L126 - is 'well known' necessary? Please strike.

Response: Removed, Thank you

L201&207 - these lines have repetitive language. Please revise. Response: Fixed, "suggesting that farmers are adapting" was removed. Reviewer #5 (Remarks to the Author):

I really like the revision of the paper by Baum et al. First of all, they tackled my main comment very well, both in improving the abstract and by adding two Figure s (S6 and S7) in the supplementary material illustrating the points that I like to make.

Response: Thank you

My specific comments are also mostly well tackled. I have some problems with the answers discussed below

1 The answer related to the role of NH3 in human health impacts by PM. Mentioning that it is uncertain is true for all the societal impacts and the reference (Kelly et al., 2004) is very old . But I can imagine that the authors like to leave it out as the NH3 losses are small. I would then rephrase the addition as follows "Note we exclude the potential cost of the formation of particulate matter PM10 and PM2.5 from NH3 volatilization from inorganic fertilizers in this analysis because NH3 volatilization is marginal within the N management practices used for maize production in the US Midwest (Woodley et al., 2020). You might add: In addition there are large uncertainties in overall impact on NH3 on PM2.5/PM10 and thereby on human health but I am inclined to remove this text. There has been a nature paper on the high impacts of NH3 emissions on PM2.5 in Nature by Gu et al (2023_

Gu et al., 2023. Cost-effective mitigation of nitrogen pollution from global croplands. Nature Vol 613 | 5 January 2023 | 77

Response: Done, thank you for the suggestion.

2 The answer related to the statement "We hypothesize that over the last decades, the optimum N rate for maize production is increasing because N outputs to grain and the environment exceed the reduction in grain N concentration". This statement remains strange as N outputs to grain is the product of crop yield and grain N concentration. I would say: "We hypothesize that over the last decades, the optimum N rate for maize production is increasing due to an increase in N outputs to grain, since crop yields increase likely exceed grain N concentration reductions, and an increase in N losses".

Response: Thanks the sentence was reworded to "We hypothesize that over the last decades, the optimum N rate for maize production is increasing due to an increase in N outputs to grain and N losses, since increased crop yields likely exceed reductions in grain N concentration."

One last comment I would revise Fig S1-S5 as follows Fig S1- is Fig S5 Fig S2- is Fig S1 Fig S3- is Fig S2 Fig S4- is Fig S3 Fig S5- is Fig S4 This is the order in which they appear in the text.

Response: Thanks, everything has been checked and the figure order is OK.