Web links to the author's journal account have been redacted from the decision letters as indicated to maintain confidentiality.

This manuscript has been previously reviewed at another Nature Research journal. This document only contains reviewer comments and rebuttal letters for versions considered at Communications Earth & Environment.

6th Apr 21

Dear Dr Feinberg,

Your manuscript titled "Risk of declining atmospheric selenium and sulfur inputs to agricultural soils" has now been seen by our reviewers, whose comments appear below. In light of their advice I am delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment under the open access CC BY license (Creative Commons Attribution v4.0 International License).

We therefore invite you to revise your paper one last time to address the remaining concerns of our reviewers, in particular those regarding validation of your model output and assumptions. At the same time we ask that you edit your manuscript to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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We hope to hear from you within two weeks; please let us know if you need more time.

Best regards,

Dr Clare Davis

Associate Editor
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REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

This is an interesting paper, suggesting a counterintuitive result: reducing acid rain (sulfate fluxes) will decrease productivity in agricultural land, plus the introduction of atmospheric Se deposition as an important driver....

Please do mention acid rain and the impacts of that, even if you think the ag impacts are important.

For the Se modeling, this is quite new and interesting, but poorly explained. please explain what the assumptions were on your sources, and how you obtained the source strengths you include.

I do not see a comparison of available Se in situ concentration or deposition data across the globe. I see just a few comparisons. Can you show more comparisons to convince us you are doing a good job?

I also wonder whether soil particles carry Se also. If I just google for crustal amounts, it suggests Se in soil should be about 1ppm. With that value that would imply that Se deposition close to desert dust or agricultural lands would be dominated by the Se in soils and how that moves around. Deposition estimates of dust in North Africa, for example, is about 50g/m2/year, so that would be 50 ug/m2/year of Se….this would be important. Plus then the estimates that agricultural lands would move around Se? please include these in a revised manuscript would be helpful.

Reviewer #2 (Remarks to the Author):

This paper examines the potential trends in atmospheric deposition of selenium and sulfur and future socio-economic scenarios. These trends are important to understand as selenium and sulfur are important nutrients either for plant grown or human and livestock health. Overall, this is a very clearly written paper with well supported conclusions, and I have only minor comments.

Main text:

1) Page 1, Line 30: The use of "nutrients" could be clarified as it is not clear if you mean the nutrients for plant growth or nutrients for human health.
2) Page 1, Line 35 (and elsewhere): The use of the term safety seems a bit odd as safety usually refers to the absence of bacteria, viruses, etc. rather than nutrient content.
3) Page 2, Line 28: SSP5-8.6 should be SSP5-8.5, I believe.
4) Page 2, Line 40: It might be helpful to clarify here why wet deposition of selenium is generally greater than dry deposition.
5) Page 3, Lines 18-20: The linkage between the deposition decrease and the prevalence of emissions is not clear to me – perhaps this could be worded a bit differently.
6) Page 4, Line 5: How did you arrive at the > 25% level? How sensitive are your findings to this? How many grid cells did this yield?
7) Page 6: Line 10: What was the height of the first layer in the modeling? The grid cell size is large compared to typical chemical transport modeling and I would expect strong sub-grid spatial gradients in concentrations and deposition. Capturing precipitation patterns in complex terrain with a large grid size can be challenging. Did you examine any downscaled climate runs to determine if grid scale had any effects?
8) Figures: Consider the colors used for the plots. For all of the figures except 3, the colors were very difficult to see and distinguish.

Supplemental Information:

Page 3, European deposition observations: It is not clear how the measurements and modeling were paired temporally.

Page 6, USGS stream data: Were the stated trends assessed to determine if they were statistically significant?
Figure S15: Consider using more distinct colors for the lines in the plot.

Reviewer #3 (Remarks to the Author):

Atmospheric selenium chemistry, further selenium deposition and fate in soil-plants systems (including crops biofortification) have received a considerable attention in the last decades. Nevertheless, this study reports on a novel point of view where the risk of declining atmospheric selenium resulting from reductions in coal-generated energy was estimated through a modelling approach. In that frame, the comparison with atmospheric S is very opportune.
I reviewed that paper as a generalist in biogeochemistry. The paper is very well structured and written. I especially appreciated the use of modelling as a tool helpful for integrating individual information's based on recent and more ancient sources, monitoring or data analysis conducted in other fields. For sure, an approach to encourage for other trace elements. The main message is convincing, and well supported by model outputs. The inevitable uncertainties left in the model were well described, notably thanks to a detailed Supplementary Information.
The role of terrestrial ecosystems in the recycling of atmospheric Se (in addition to anthropogenic or geogenic Se supply) is central and extent of Se retention or mobility in soil may vary depending on edaphic conditions, explaining why balance between inflow and outflow is also not easy to characterize at large scales. This paper carefully illustrated that complexity by comparing Se mass balances in different US watersheds.
The lag between atmospheric deposition and runoff to the rivers still remains few documented, which makes the risk of atmospheric Se decline, here estimated at a global scale, difficult to objectify at a local scale. Gaps and needs in that domain of selenium terrestrial biogeochemistry could be emphasized, for example by listing the most important processes, notably those leading to labile/non-labile pools, or involved in inorganic/organic cycles of selenium. In other terms, I found the Fig.3 very simplified in the light of the current knowledge, notably about the influence of organic matter turnover that can alter the Se cycle dynamics, especially in a context of climate change.
Overall, this is an interesting and well-conducted modelling approach to address the initial question: What is the possible extent of decline of Se atmospheric supply in this century? The conclusions are of interest to different scientific disciplines involved in selenium biogeochemistry and food chain management. The current draft require minor changes.

Minor comments/suggestions
P2 L17 : "… reduced soil Se retention associated with climate change." - Quite vague statement; the introduction of the mechanism(s) involved would be informative.
P4 L19-20 : "…, retention of Se in soils will decrease due to aridification with climate, …" – idem, it would be opportune to introduce a more explicit information on the mechanism involved.
P4 L28-30: "A significant fraction of fertilized S and Se will not be assimilated by crops and can leach into surface waters." - That statement could be nuanced by indicating that Se budgets and mass balances at different scales (field plots, watersheds ...) are still few documented in the literature for various environmental conditions.
P5 L17-18: "… a critical research need would be to conduct mass balance calculations for Asian and European agricultural catchments." – Obviously, agricultural area are at the heart of questions about selenium in the foodchain. On long time scale, budgets in agricultural catchments are not easy to

quantify due to numerous potential changes in practices and land use. Semi-natural areas, non perturbed by changes in land use or management practices, could serve as a reference in balance calculations and must not be neglected in catchment studies.

P6 L36-38: It could be mentioned somewhere if soil occupation was maintained constant for the period of simulations in all the scenarios tested.

Author response to all referees' comments on COMMSENV-21-0109-T, "Risk of declining atmospheric selenium and sulfur inputs to agricultural soils"

We would like to thank the three reviewers for their feedback on the manuscript. We very much appreciate their time and interest in the manuscript. We have considered these comments carefully and present our responses below, as well as changes to the main text (reviewer comments are in blue and author responses are in black).

Response to Reviewer #1

This is an interesting paper, suggesting a counterintuitive result: reducing acid rain (sulfate fluxes) will decrease productivity in agricultural land, plus the introduction of atmospheric Se deposition as an important driver....
Please do mention acid rain and the impacts of that, even if you think the ag impacts are important.

Thank you for pointing this out; we agree that it is important to emphasize the consequence of coal combustion for acid rain and climate. We have added to the introduction (Page 2, Lines 4–6 & Lines 9–11) additional sentences acknowledging the impacts of acid rain, as well as the importance of reducing coal emissions for climate change mitigation:
"*Until the 1990s, high levels of atmospheric S deposition in industrialized countries caused acidification of aquatic systems, decline of fish populations, and degradation of forests (13,14) … Shifts away from coal energy generation are also essential for meeting the climate change mitigation goals of the Paris Agreement (15).*"

For the Se modeling, this is quite new and interesting, but poorly explained. please explain what the assumptions were on your sources, and how you obtained the source strengths you include.

The current study builds upon results established by two previous peer-reviewed publications from our group (Feinberg et al., https://doi.org/10.5194/acp-20-1363-2020, 2020a; Feinberg et al., https://doi.org/10.1021/acs.est.0c01408, 2020b). Feinberg et al. (2020a) contains a very detailed description of the assumptions in our Se model and presents a global sensitivity analysis of the model. Feinberg et al. (2020b) focuses on constraining the magnitude of Se emission sources using Bayesian inverse modelling. The method for deriving the Se source strengths is elaborated in Feinberg et al. (2020b), which we state in the methods (Page 7, Lines 11–13). We believe that the summarized description of the model in the methods section is suitable for the reader to understand the approach. We reference both of these foundational modelling papers throughout the methods section so that interested readers can refer to the detailed information in these publications.

I do not see a comparison of available Se in situ concentration or deposition data across the globe. I see just a few comparisons. Can you show more comparisons to convince us you are doing a good job?

In the current study, we show that the model matches well with Se deposition trends in Canada (Fig. 1B) and Europe (Supplementary Figure 8). In a previous study, we have validated the model with a much larger observational dataset of airborne particulate Se (Feinberg et al., https://doi.org/10.1021/acs.est.0c01408, 2020b). The observational dataset includes measurements from five aerosol networks and 232 papers that we compiled from the literature, totaling over 600 measurement locations (see Figure 1 in Feinberg et al., 2020b). Overall, our model matched very well with the observational dataset (see Figures 3 and 4 in Feinberg et al., 2020b), predicting 85% of observed annual means within a factor of 2 and displaying a good correlation with observations (R^2 = 0.66). To emphasize this point, we have added the following sentence to Page 3, Lines 21–23: *In a previous study comparing the model with a larger observational dataset for particulate Se, we found that 85% of modelled Se concentrations are within a factor of 2 of observations and the model captures the observed decline in particulate Se in North America (10).*

I also wonder whether soil particles carry Se also. If I just google for crustal amounts, it suggests Se in soil should be about 1ppm. With that value that would imply that Se deposition close to desert dust or agricultural lands would be dominated by the Se in soils and how that moves around. Deposition estimates of dust in North Africa, for example, is about 50g/m2/year, so that would be 50 ug/m2/year of Se….this would be important. Plus then the estimates that agricultural lands would move around Se? please include these in a revised manuscript would be helpful.

Dust is expected to be a more minor source of atmospheric Se, estimated by previous budget studies to be less than 0.3% of the total Se emissions (Mosher and Duce, 1987; Nriagu, 1989; Wen and Carignan, 2007). The reason for this is that the Se concentration in the Earth's crust is on average very low, 0.05 ppm (Plant et al., 2014). Using this average concentration, in a previous study (Feinberg et al., https://doi.org/10.1021/acs.est.0c01408, 2020b) we calculated the mean flux of Se from the bedrock through soil formation, determining an annual source of 3.5 μg m^{-2}. This is much smaller than the global mean deposition flux to continents, 87 μg m^{-2}. However, the reviewer is correct that crustal sources could play more of a role in specific regions, e.g., areas with black shale bedrock where geological Se concentrations are high (Plant et al., 2014). Since we did not focus our analysis on these regions, and we have found a limited role for crustal sources on a global scale, in this paper we do not consider these sources.

Response to Reviewer #2

This paper examines the potential trends in atmospheric deposition of selenium and sulfur and future socio-economic scenarios. These trends are important to understand as selenium and sulfur are important nutrients either for plant grown or human and livestock health. Overall, this is a very clearly written paper with well supported conclusions, and I have only minor comments.

Thank you.

Main text:

1) Page 1, Line 30: The use of "nutrients" could be clarified as it is not clear if you mean the nutrients for plant growth or nutrients for human health.

"Nutrients" here refers to both nutrients for plant growth and/or human health. Sulfur is a nutrient for plant growth and human/animal health, while selenium is a nutrient for human/animal health. Since this is explained in the sentences that follow in the introduction, we decided to leave as is.

2) Page 1, Line 35 (and elsewhere): The use of the term safety seems a bit odd as safety usually refers to the absence of bacteria, viruses, etc. rather than nutrient content.

Thank you for this point. To avoid confusion, we have replaced this term throughout with the terms nutrition or micronutrient deficiency (Page 1, Line 27; Page 1, Line 37; Page 6, Line 2)

3) Page 2, Line 28: SSP5-8.6 should be SSP5-8.5, I believe.
Corrected.

4) Page 2, Line 40: It might be helpful to clarify here why wet deposition of selenium is generally greater than dry deposition.

Thanks for this suggestion. We have added the following to the text (Page 3, Lines 6–8): *Since atmospheric Se is mainly transported in submicron particles, the dominant deposition pathway is wet deposition (approximately 80% of total deposition) (10).*

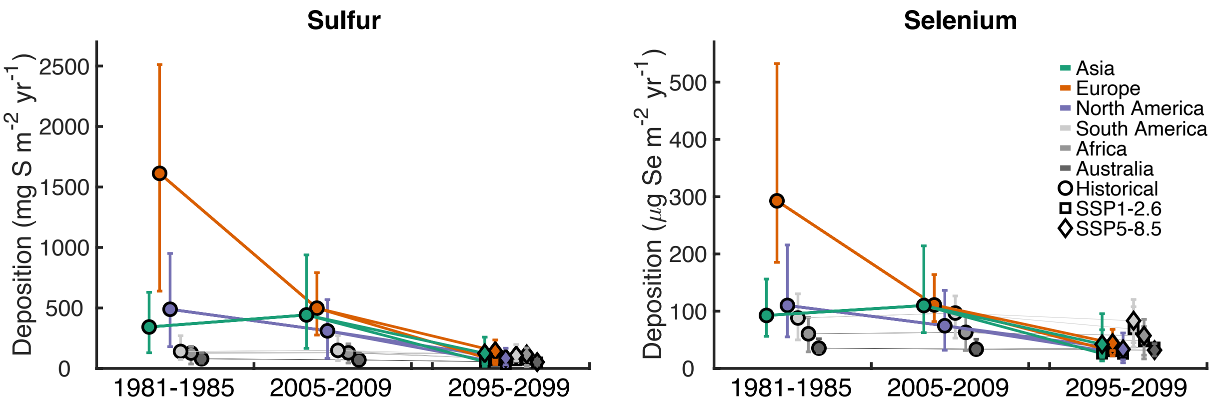
5) Page 3, Lines 18-20: The linkage between the deposition decrease and the prevalence of emissions is not clear to me – perhaps this could be worded a bit differently.

The anthropogenic emissions were mainly located in Asia, North America, and Europe in the recent period. Therefore, when anthropogenic emissions decrease in the future, these continents also show the greatest decrease in Se deposition. To ensure clarity, we have amended this sentence to (Page 3, Lines 32–34): *The projected decrease in deposition is particularly noteworthy over Asia, North America, and Europe, because anthropogenic emissions are mainly located in these continents.*

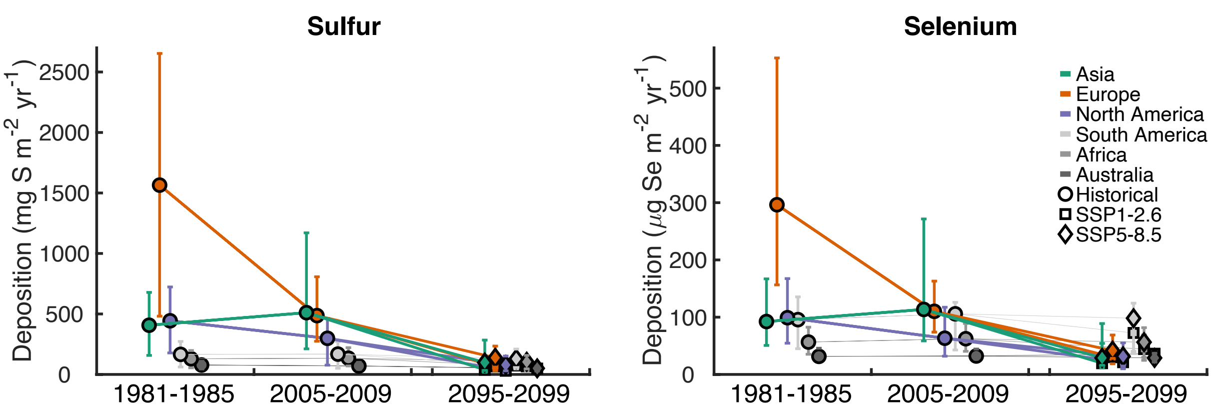
6) Page 4, Line 5: How did you arrive at the > 25% level? How sensitive are your findings to this? How many grid cells did this yield?

We used the >25% level to be consistent with previous work in the literature that investigated the effect of climate change on soil Se levels in agricultural areas (Jones et al., 2017). This yields a reasonable number of grid cells for each continent in order to calculate statistics: Asia (295 grid cells), Europe (124), North America (116), South America (132), Africa (200), and Australia (63). We tested the plot for different thresholds (>10% and >50%) and the results were almost identical (see plots below for >10% and >50%).

>10% threshold for agricultural grid cells



>50% threshold for agricultural grid cells



7) Page 6: Line 10: What was the height of the first layer in the modeling? The grid cell size is large compared to typical chemical transport modeling and I would expect strong sub-grid spatial gradients in concentrations and deposition. Capturing precipitation patterns in complex terrain with a large grid size can be challenging. Did you examine any downscaled climate runs to determine if grid scale had any effects?

Thank you for the question, we also think a lot about this issue and aim to only apply our model to problems where it would be suitable. Given that SOCOL-AER is a global model, it could have difficulty capturing smaller-scale temporal and spatial gradients, particularly in mountainous regions. However, our study focuses on regional to continental scale changes in deposition averaged over 5-year simulation periods. We therefore would argue that the deposition trends are dominated by emission changes, meaning that resolution would not impact the main conclusions of our paper. We would be very interested in incorporating the Se model in a regional transport model, in order to better understand shorter-term field measurements from mountainous areas, as well as to better evaluate the model's accuracy. We did not examine any downscaled climate runs for this study, but we agree that such an approach would be required to refine the predictions for specific locations. To answer your first question, the height of the first layer in the model varies spatially and temporally depending on the orography and local meteorology, but for standard surface pressure it would be around 70 m.

8) Figures: Consider the colors used for the plots. For all of the figures except 3, the colors were very difficult to see and distinguish.

Thank you for pointing this out. We have adjusted the line colours in Fig. 2 and Fig. 4 to increase the contrast.

Supplemental Information:

Page 3, European deposition observations: It is not clear how the measurements and modeling were paired temporally.

We clarified this with an additional sentence: *For each location, we average the model over the same years that were available from the observations.*

Page 6, USGS stream data: Were the stated trends assessed to determine if they were statistically significant?

Thanks for this important question. The flow-normalized flux trends from the USGS stream data are calculated using Weighted Regressions on Time Discharge and Season (WRTDS) from the R EGRET package (Hirsch et al., 2015a). Bootstrap methods have been developed to assess the statistical significance of these trends and produce confidence intervals for WRTDS estimates (Hirsch et al., 2015b). These methods were released as an R package, EGRETci, in 2019 and we use it to assess the statistical significance of our results. We calculated 90% confidence bands (5^{th} percentile – 95^{th} percentile) around the flow-normalized flux trends in Figure 4, which we show below. We believe that such a figure would be too confusing for the main manuscript. However, it illustrates that indeed for the three watersheds in the northeastern US (Susquehanna River, Delaware River, and Ohio River) the riverine Se and sulfate trends have evolved separately from the orthophosphate and nitrate trends. In the Mississippi River and Platte River watersheds, Se and sulfate trends more closely follow the trends of orthophosphate and nitrate. Using the EGRETci package, we find that the likelihood that mentioned riverine Se trends are negative to be very likely (>90%): 97% likelihood for the Susquehanna River, 92% for the Delaware River, 99% for the Ohio River, and 97% for the San Joaquin River. Likewise, the riverine trends in Se and sulfate at the Mississippi River and Platte River basin are all very likely (>90%) to be increasing: 90% for Se at Mississippi River, 99% for sulfate at Mississippi River, 99% for Se at Platte River, and 90% for sulfate at Platte River. We have added statements regarding the significance of the riverine Se trends throughout the discussion (Page 5, Lines 17, 26, 34):

-Watersheds in the Northeastern USA show declines ***(likelihood of decreasing trend > 92%)*** *in Se stream fluxes (–15 to –28%) that follow steeper Se deposition declines in these watersheds (–39 to –58%).*
-In contrast, basins in the Midwestern USA show increasing trends in the riverine S and Se ***(likelihood of increasing trend > 90%)*** *that more closely follow nitrate and orthophosphate trends and are inconsistent with decreasing deposition trends.*
*- In the San Joaquin River Basin, an area known for high Se concentrations in agricultural irrigation drainage water causing toxicity in animals (37), riverine Se fluxes have strongly decreased (–73%) from 2008 to 2019 (****likelihood of decreasing trend = 97%).***

We have added the following to the methods section to describe the approach (Page 9, Lines 10–12): *Statistical significance of the derived WRTDS trends was assessed by calculating the likelihood of increasing or decreasing trends with a bootstrap approach (67), using methods available in the R package EGRETci.*

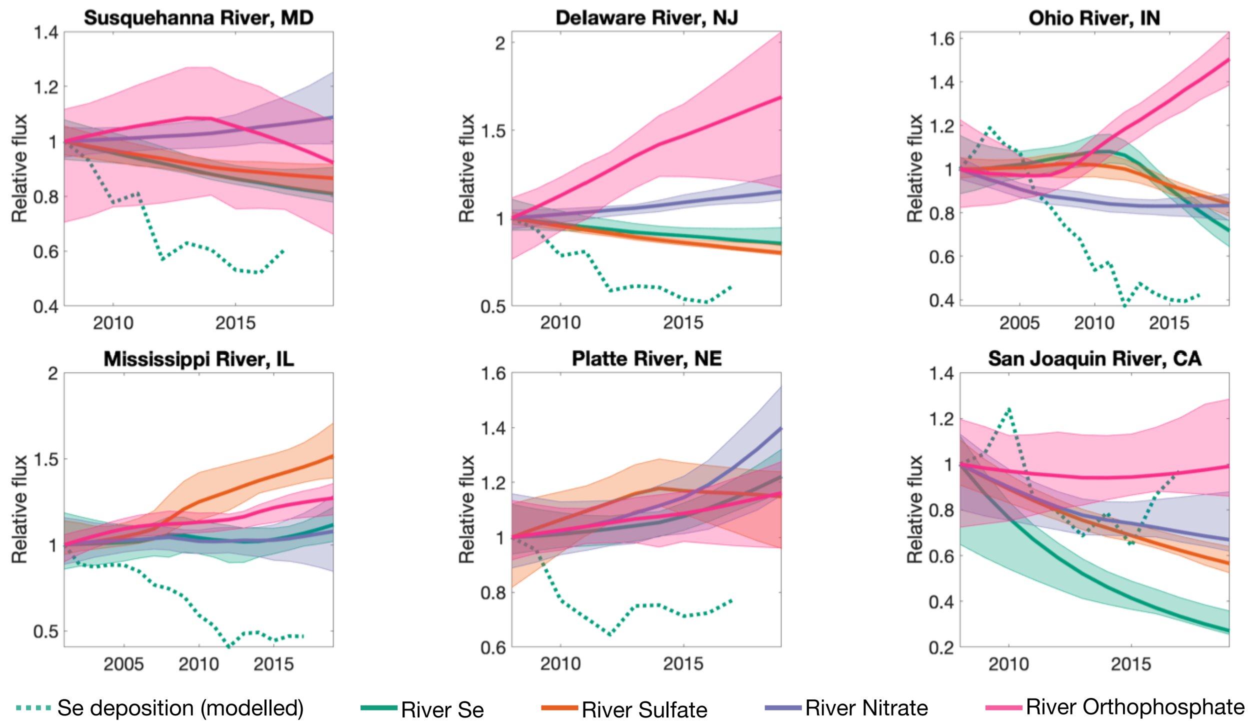


Figure S15: Consider using more distinct colors for the lines in the plot.

We have adjusted the line colours to increase the contrast.

Response to Reviewer #3

Atmospheric selenium chemistry, further selenium deposition and fate in soil-plants systems (including crops biofortification) have received a considerable attention in the last decades. Nevertheless, this study reports on a novel point of view where the risk of declining atmospheric selenium resulting from reductions in coal-generated energy was estimated through a modelling approach. In that frame, the comparison with atmospheric S is very opportune.

I reviewed that paper as a generalist in biogeochemistry. The paper is very well structured and written. I especially appreciated the use of modelling as a tool helpful for integrating individual information's based on recent and more ancient sources, monitoring or data analysis conducted in other fields. For sure, an approach to encourage for other trace elements. The main message is convincing, and well supported by model outputs. The inevitable uncertainties left in the model were well described, notably thanks to a detailed Supplementary Information.

The role of terrestrial ecosystems in the recycling of atmospheric Se (in addition to anthropogenic or geogenic Se supply) is central and extent of Se retention or mobility in soil may vary depending on edaphic conditions, explaining why balance between inflow and outflow is also not easy to characterize at large scales. This paper carefully illustrated that complexity by comparing Se mass balances in different US watersheds.

The lag between atmospheric deposition and runoff to the rivers still remains few documented, which makes the risk of atmospheric Se decline, here estimated at a global scale, difficult to objectify at a local scale. Gaps and needs in that domain of selenium terrestrial biogeochemistry could be emphasized, for example by listing the most important processes, notably those leading to labile/non-labile pools, or involved in inorganic/organic cycles of selenium. In other terms, I found the Fig.3 very simplified in the light of the current knowledge, notably about the influence of organic matter turnover that can alter the Se cycle dynamics, especially in a context of climate change.

We thank the reviewer for their comments and agree that Fig. 3 is very simplified in terms of soil processes. Our intention is to focus the diagram on the results from the current study on atmospheric deposition changes from present to the future, and how this would affect the overall mass balance of the soil/plant system. It would be difficult to include different S and Se soil pools within the diagram, while maintaining readability and accuracy. Since this is a topic which should be afforded more nuance, we instead add the following statement in the caption, referencing to comprehensive reviews on S and Se cycling in soils: *Soil processes are simplified in the diagram and are discussed more comprehensively in previous reviews (Dinh et al., 2019; Li et al., 2017; Kovar and Grant, 2011; Scherer, 2009).*

Overall, this is an interesting and well-conducted modelling approach to address the initial question: What is the possible extent of decline of Se atmospheric supply in this century? The conclusions are of interest to different scientific disciplines involved in selenium biogeochemistry and food chain management. The current draft require minor changes.

Minor comments/suggestions

P2 L17 : "… reduced soil Se retention associated with climate change." - Quite vague statement; the introduction of the mechanism(s) involved would be informative.
We have included an additional sentence to summarize the mechanism from Jones et al. (2017) (Page 2, Lines 24–26): *The driving mechanism for soil Se losses is that increasing aridification shifts the speciation of Se to more oxidized species that are weaker bound in soil and leach more readily during precipitation events.*

P4 L19-20 : "…, retention of Se in soils will decrease due to aridification with climate, …" – idem, it would be opportune to introduce a more explicit information on the mechanism involved.

We have decided to leave as is, since a full discussion of the included mechanisms of reduced Se retention are discussed within the referenced paper (Jones et al., 2017), and we have now included a brief summary of the mechanism in the Introduction (previous response, Page 2, Lines 24–26).

P4 L28-30: "A significant fraction of fertilized S and Se will not be assimilated by crops and can leach into surface waters." - That statement could be nuanced by indicating that Se budgets and mass balances at different scales (field plots, watersheds ...) are still few documented in the literature for various environmental conditions.

Thanks for this point. We added several sentences to expand on this issue, as well as mentioning here that there are other agricultural management practices that could decrease the required amount of fertilizer inputs to soils (Page 5, Lines 4–11): *The extent to which S and Se are mobilized from soil systems would depend on the local environmental conditions (e.g., temperature, precipitation, and land use) and soil properties (e.g., pH, organic carbon content, and clay content) (17). In general, there is a strong need for studies that assess S and Se mass balances at a variety of scales (field plots, watersheds, and larger regions) and geographic locations. Other agricultural management practices could also be employed to increase the efficiency of S and Se uptake by plants, such as conventional breeding or genetic engineering (34,35), thereby reducing the required amount of fertilizer inputs.*

P5 L17-18: "… a critical research need would be to conduct mass balance calculations for Asian and European agricultural catchments." – Obviously, agricultural area are at the heart of questions about selenium in the foodchain. On long time scale, budgets in agricultural catchments are not easy to quantify due to numerous potential changes in practices and land use. Semi-natural areas, non perturbed by changes in land use or management practices, could serve as a reference in balance calculations and must not be neglected in catchment studies.

We agree with your point and we removed the word "agricultural" to generalize our statement.

P6 L36-38: It could be mentioned somewhere if soil occupation was maintained constant for the period of simulations in all the scenarios tested.

The biogenic emissions of Se from soils is included as a climatology, meaning that there is a seasonal cycle in emissions, but these emissions do not change interannually. Our emissions are scaled from the spatial distribution of volatile organic carbon emissions from the MEGAN-MACC inventory (Sindelarova et al., 2014). MEGAN-MACC parametrizes volatile organic carbon emissions as a function of light, temperature, leaf area index, carbon dioxide, plant species distributions, and land cover. More information about the calculation of terrestrial biogenic Se emissions can be found in the Supplementary Information of Feinberg et al., https://doi.org/10.1021/acs.est.0c01408 (2020b).

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