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31st Oct 23

Dear Mr Cusworth,

Your manuscript titled "The growing legacy of plastic use" has now been seen by 2 reviewers, and we include their comments at the end of this message. They find your work of interest, but some important points are raised. We are interested in the possibility of publishing your study in Communications Earth & Environment, but would like to consider your responses to these concerns and assess a revised manuscript before we make a final decision on publication.

We therefore invite you to revise and resubmit your manuscript, along with a point-by-point response that takes into account the points raised. Please highlight all changes in the manuscript text file.

We are committed to providing a fair and constructive peer-review process. Please don't hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the referees' comments (which should be in a separate document to any cover letter), a tracked-changes version of the manuscript (as a PDF file) and the completed checklist:

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We hope to receive your revised paper within six weeks; please let us know if you aren't able to submit it within this time so that we can discuss how best to proceed. If we don't hear from you, and the revision process takes significantly longer, we may close your file. In this event, we will still be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

Please do not hesitate to contact us if you have any questions or would like to discuss these revisions further. We look forward to seeing the revised manuscript and thank you for the opportunity to review your work.

Best regards,

Alienor Lavergne, PhD
Associate Editor
Communications Earth & Environment

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- If a dataset has a Digital Object Identifier (DOI) as its unique identifier, we strongly encourage including this in the Reference list and citing the dataset in the Data Availability Statement.

DATA SOURCES: All new data associated with the paper should be placed in a persistent repository where they can be freely and enduringly accessed. We recommend submitting the data to discipline-specific, community-recognized repositories, where possible and a list of recommended repositories is provided at <http://www.nature.com/sdata/policies/repositories>><http://www.nature.com/sdata/policies/repositories>.

If a community resource is unavailable, data can be submitted to generalist repositories such as <https://figshare.com/>>figshare or <http://datadryad.org/>>Dryad Digital Repository. Please provide a unique identifier for the data (for example a DOI or a permanent URL) in the data availability statement, if possible. If the repository does not provide identifiers, we encourage authors to supply the search terms that will return the data. For data that have been obtained from publically available sources, please provide a URL and the specific data product name in the data availability statement. Data with a DOI should be further cited in the methods reference section.

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REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

Cusworth et al. discuss the increasing impact of plastic usage, with a particular emphasis on agricultural plastics at a highly specific research site. It is essential to note that the ramifications of plastics extend far beyond soil environments. The paper's title could be revised to better encapsulate this focus:

"Assessing the Expanding Environmental Footprint of Agricultural Plastics: A Time Series Analysis of the Rothamsted Sample Archive in the UK"

Line 19: While the data is valuable, it has been previously reported in multiple sources. The primary emphasis should be placed on the agricultural context, encompassing atmospheric transport. Additionally, I recommend incorporating data about plastic fibers.

Line 23: It is not entirely accurate to state that plastics are irreversible contaminants in soil. It is crucial to provide details on degradation rates and mass flow, and to clarify that plastic degradation is a multifaceted process involving biodegradation, chemical weathering, and potential biomineralization, going beyond just physical fragmentation. Plastics might also fully biodegrade.

Line 25: Please refer to the "Current Communication Earth and Environment" for more comprehensive information on plastics in agriculture. This source contains extensive details on sources and sinks that should be cited. <https://www.nature.com/articles/s43247-023-00982-4>

Line 39: Mention that the examples provided are not exhaustive, as there are numerous other applications and uses of additives within the industry.

Line 50: To maintain a balanced perspective, this paper should include a discussion of the ecological benefits of plastics. Banning plastics outright can have severe repercussions on current agricultural practices and ecosystems.

Line 62: The statement about contamination should be further elaborated, explaining why it is unavoidable. Procedures for blank samples should be provided.

Figure 1: Revise the figure legend for better clarity. Simplify the representation of "2.21 FYM" for reader-friendliness. Provide an explanation for why the "081" treatment is at the same level as the manure treatment, as this needs clarification.

Line 85: Incorporate statistical analysis in this section. With only four samples available for comparison between FYM and 081, it may not be sufficient to make a compelling case.

Line 87: Avoid speculative statements without supporting evidence.

Line 98 and following: The speculative nature of the content in this section is concerning. There should be more documentation for the research site, including information about its location and proximity to roads.

Line 108: Remove claims of irreversibility regarding plastics. The degradation process depends on the type of plastics and degradation rates. The subsequent statements should be more specific and supported by evidence.

The method section lacks a comprehensive discussion of quality control. Address issues with the Nile red (NR) staining method, highlighting its cost-effectiveness and simplicity, but also noting the need for quality control. The outcome of microplastic examination with NR is significantly affected by the method and efficacy of organic matter removal. Shruti et al made clear that the lack of proper validation and quality risks to generate data that will have limited use for comparisons and exposure assessment. (<https://doi.org/10.1016/j.jhazmat.2021.127171>). Their review shows that organic debris hinders microplastic quantification and necessitates additional procedures to ensure the accuracy of NR staining data. For instance, obtaining results for unstained samples through light microscopy and subsequently correlating the data post-staining. Alternatively, one may integrate NR staining with FTIR or Raman to confirm quantification. You can also compare microplastic-spiked samples that contain a specific number of NR-stained plastic particles to examine recovery rates using visible or automated particle counting methods. This way, you get to determine the precision of microplastic separation methods and the effectiveness of the NR method applied in the study. The NR fluorescence methodology in conjunction with these approaches is necessary for significant insight into

the certainty of microplastic quantification. However, there is no established method to ensure that organic contaminants other than plastics are removed from environmental samples.

Reviewer #2 (Remarks to the Author):

Overall, this is a clearly written paper with the potential to make an important contribution to the literature on the impacts of plastic use in agriculture. As a social scientist who studies agricultural plastic pollution, I appreciated that the paper was accessible to a broad audience.

I have a few suggestions for improvements:

(1) The authors should consider adding a sentence to explain how exactly inorganic fertilisers and farmyard manure can be direct or indirect sources of plastic residues.

(2) The authors define microplastics, but should consider also defining macroplastics and nanoplastics. Were nanoplastics too small to measure in the study?

(3) The authors define microplastics as "any polymer between 1 micrometer and 5 mm." They then state that the soil samples showed a "significant increase in microplastic concentrations and particles less than 10 micrometers." Later in the paper, they explain that a particle was identified as a microplastic if the "particle size was greater than 10 micrometers." These three statements in combination are very confusing. Why not use the official definition of microplastics in the study? Why not include the actual size range used in the study (rather than just "less than" or "greater than")? There needs to be more consistency/clarity in the microplastic definition/measurement.

(4) The year ranges jump around a great deal in the discussion of the study results: 1846-1914, 1900-1960, 1966-2022, 1966-2010, 1997-2005, and 2010-2022. One of the years (1960) is not one of the 18 time points. I would suggest presenting the results in a more chronological order with discussion of 3-4 time periods in turn (rather than jumping around in time).

Author comments and changes made to the manuscript

We are grateful for the comments from both of the referees and have provided responses below. Thank you.

Referee 1

General comments - Cusworth et al. discuss the increasing impact of plastic usage, with a particular emphasis on agricultural plastics at a highly specific research site. It is essential to note that the ramifications of plastics extend far beyond soil environments. The paper's title could be revised to better encapsulate this focus: "Assessing the Expanding Environmental Footprint of Agricultural Plastics: A Time Series Analysis of the Rothamsted Sample Archive in the UK".

Author response: Thank you for your comments on the manuscript and your suggested changes.

We agree that the title of the manuscript should be changed to better reflect the focus of the research. According to the style and formatting guide, manuscript titles should be 15 words or fewer. Therefore, we have used an adapted version of the title you suggested: "Assessing the expanding environmental footprint of agricultural plastics: a time series analysis".

Line 19: While the data is valuable, it has been previously reported in multiple sources. The primary emphasis should be placed on the agricultural context, encompassing atmospheric transport. Additionally, I recommend incorporating data about plastic fibers.

Author response: We thank the reviewer for this comment and agree. Given that the scope of the research we have added a statement about the proportion of plastics used in agriculture on line 21 - 22. We have added a couple of sentences to reflect the important of atmospheric transport and deposition in agricultural soils. As particle shape is not a key focus of this research we have referred to fibers briefly. These changes have been made between line 50 – 56.

Line 23: It is not entirely accurate to state that plastics are irreversible contaminants in soil. It is crucial to provide details on degradation rates and mass flow, and to clarify that plastic degradation is a multifaceted process involving biodegradation, chemical weathering, and potential biomineralization, going beyond just physical fragmentation. Plastics might also fully biodegrade.

Author response: We have made changes to reflect your comment and have described microplastic pollution as 'poorly reversible' instead as stated by MacLeod et al. 2021. On line 24 we already state that plastic degradation pathways may be microbial, chemical and mechanical. To clarify this, we have added an additional statement to provide more detail on degradation pathways and made reference to biodegradable plastics on line 24 – 30.

Line 25: Please refer to the "Current Communication Earth and Environment" for more comprehensive information on plastics in agriculture. This source contains extensive details

on sources and sinks that should be cited. <https://www.nature.com/articles/s43247-023-00982-4>

Author response: We thank the reviewer for the link the suggested reference. This is a great resource and we have now cited the work of Hofmann et al. in this paper. We have referred to numerous sources on line 42 – 45 of the original manuscript. As suggested, we have included a more complete list of microplastic sources to agricultural soils between these lines.

Line 39: Mention that the examples provided are not exhaustive, as there are numerous other applications and uses of additives within the industry.

Author response: This has been clarified on line 62.

Line 50: To maintain a balanced perspective, this paper should include a discussion of the ecological benefits of plastics. Banning plastics outright can have severe repercussions on current agricultural practices and ecosystems.

Author response: We agree and have added a paragraph on lines 72 - 80 to provide a balanced perspective and state why removing plastics is not the solution and may do more harm than good.

Line 62: The statement about contamination should be further elaborated, explaining why it is unavoidable. Procedures for blank samples should be provided.

Author response: We have added a note about unavoidable contamination from lab environments on line 95 – 96. A couple of sentences have also been added to the methods section to describe the procedure for blank samples.

Figure 1: Revise the figure legend for better clarity. Simplify the representation of "2.21 FYM" for reader-friendliness. Provide an explanation for why the "081" treatment is at the same level as the manure treatment, as this needs clarification.

Author response: We have removed the plot numbers from the figure legend in Figure 1 and inserted these in the figure caption.

Line 85: Incorporate statistical analysis in this section. With only four samples available for comparison between FYM and 081, it may not be sufficient to make a compelling case.

Author response: Given the nature of these classic long-term experiments and the lack of replication, they are challenging to analyse statistically. With just four time points we feel that statistical analysis of this subset could be misleading. Instead we have changed the wording of our statements on line 120 to be recognise this limitation.

Line 87: Avoid speculative statements without supporting evidence.

Author response: On lines 122 – 128 we have clarified our original statement and provided additional information and supporting references.

Line 98 and following: The speculative nature of the content in this section is concerning. There should be more documentation for the research site, including information about its location and proximity to roads.

Author response: We have changed the text between lines 139 – 144 to be less speculative and provided additional references. We have added more information about the site and the location to the methods (line 164 – 169). It is unexpected that microplastic pollution from roads has a significant impact at this site. Instead, tyre wear mentioned is referring to farm machinery.

Line 108: Remove claims of irreversibility regarding plastics. The degradation process depends on the type of plastics and degradation rates. The subsequent statements should be more specific and supported by evidence.

Author response: We have changed our statement in line with our response to line 23.

Methods: The method section lacks a comprehensive discussion of quality control. Address issues with the Nile red (NR) staining method, highlighting its cost-effectiveness and simplicity, but also noting the need for quality control. The outcome of microplastic examination with NR is significantly affected by the method and efficacy of organic matter removal. Shruti et al made clear that the lack of proper validation and quality risks to generate data that will have limited use for comparisons and exposure assessment. (<https://doi.org/10.1016/j.jhazmat.2021.127171>). Their review shows that organic debris hinders microplastic quantification and necessitates additional procedures to ensure the accuracy of NR staining data. For instance, obtaining results for unstained samples through light microscopy and subsequently correlating the data post-staining. Alternatively, one may integrate NR staining with FTIR or Raman to confirm quantification. You can also compare microplastic-spiked samples that contain a specific number of NR-stained plastic particles to examine recovery rates using visible or automated particle counting methods. This way, you get to determine the precision of microplastic separation methods and the effectiveness of the NR method applied in the study. The NR fluorescence methodology in conjunction with these approaches is necessary for significant insight into the certainty of microplastic quantification. However, there is no established method to ensure that organic contaminants other than plastics are removed from environmental samples.

Author response: We thank the reviewer for their comprehensive comment about quality control. Our quality control procedures are now fully outlined and we have added multiple statements throughout the methods section. We were already aware of this excellent reference and have now used this to support some of the new statements. We appreciate the comments about using FTIR or Raman to improve quantification, this would be an opportunity for

future research. However, we believe that digestion and density separation are effective methods used to sufficiently remove organic matter, this method has been used by a number of highly-cited studies: <https://doi.org/10.1007/s10661-019-7786-4>, <https://doi.org/10.1016/j.scitotenv.2019.05.456>, <https://doi.org/10.1021/acs.est.8b01517>.

Referee 2

General comments - Overall, this is a clearly written paper with the potential to make an important contribution to the literature on the impacts of plastic use in agriculture. As a social scientist who studies agricultural plastic pollution, I appreciated that the paper was accessible to a broad audience.

Author response: We thank the reviewer for their comments and are glad that they felt the manuscript is suitable for a broad audience.

Comment 1: The authors should consider adding a sentence to explain how exactly inorganic fertilisers and farmyard manure can be direct or indirect sources of plastic residues.

Author response: We thank the reviewer for the comment. We have added a couple of sentences between line 46 – 50 to differentiate between the two.

Comment 2: The authors define microplastics, but should consider also defining macroplastics and nanoplastics. Were nanoplastics too small to measure in the study?

Author response: We have added the definitions of macroplastics and nanoplastics to the manuscript on line 38 - 40. Nanoplastics were too small to measure in this study – but this is a great opportunity for future research.

Comment 3: The authors define microplastics as "any polymer between 1 micrometer and 5 mm." They then state that the soil samples showed a "significant increase in microplastic concentrations and particles less than 10 micrometers." Later in the paper, they explain that a particle was identified as a microplastic if the "particle size was greater than 10 micrometers." These three statements in combination are very confusing. Why not use the official definition of microplastics in the study? Why not include the actual size range used in the study (rather than just "less than" or "greater than")? There needs to be more consistency/clarity in the microplastic definition/measurement.

Author response: We have opted to remove our discussion of particles < 10 um apart from our comment on line 96 about contamination. After we define macro-, micro- and nanoplastics, we have added a statement to state the actual size range used in the study on lines 40 - 42. The methodology and instrumentation can only confidently detect microplastics of a size greater than 10 micrometres, so particles between 1um – 10um are not captured, this is much clearer now.

Comment 4: The year ranges jump around a great deal in the discussion of the study results: 1846-1914, 1900-1960, 1966-2022, 1966-2010, 1997-2005, and 2010-2022. One of the years (1960) is not one of the 18 time points. I would suggest presenting the results in a more chronological order with discussion of 3-4 time periods in turn (rather than jumping around in time).

Author response: Thank you for raising this comment. We agree with the reviewer and have made changes to the first 3 periods to: 1846-1914, 1914-1966 and 1966-2022. We believe that these are 3 key periods; no microplastics, detection of microplastics, increase in microplastics. We have changed the order of our discussion to make it more chronological but since we discuss comparisons between treatments as a primary focus this is not always possible.

23rd Nov 23

Dear Mr Cusworth,

Your manuscript titled "The growing legacy of plastic use" has now been seen by our reviewers, whose comments appear below. In light of their advice we are 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. 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.

Please note that it may still be possible for your paper to be published before the end of 2023, but in order to do this we will need you to address these points as quickly as possible so that we can move forward with your paper.

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

Best regards,

Alienor Lavergne, PhD
Associate Editor
Communications Earth & Environment

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors answered all questions.

Reviewer #2 (Remarks to the Author):

I am pleased with the revised manuscript. The authors have adequately addressed my comments/concerns. I look forward to seeing the final published version of the paper. Great job!