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

6th May 20

Dear Dr. Koren,

Your manuscript titled "Airborne microplastic particles in the marine atmosphere" has now been seen by two reviewers, and I 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.

Please note that for publication in Communications Earth & Environment to be appropriate, we would need you to:

* provide compelling evidence for non-negligible amounts of airborne marine plastics in the open ocean, away from the coast, including full details on the methodology
* discuss the findings in the context of the latest literature more comprehensively and specifically

If you are able to meet those thresholds, we 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) and the completed checklist:
[link redacted]
** This url links to your confidential home page and associated information about manuscripts you may have submitted or be reviewing for us. If you wish to forward this email to co-authors, please delete the link to your homepage first **

We hope to receive your revised paper within three months; 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 will 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.

We understand that due to the current global situation, the time required for revision may be longer than usual. We would appreciate it if you could keep us informed about an estimated timescale for resubmission, to facilitate our planning. Of course, if you are unable to estimate, we are happy to accommodate necessary extensions nevertheless.

Please do not hesitate to contact me 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,

Heike Langenberg, PhD

Chief Editor
Communications Earth and Environment

On Twitter: @CommsEarth

EDITORIAL POLICIES AND FORMATTING

We ask that you ensure your manuscript complies with our editorial policies. Please ensure that the following formatting requirements are met, and any checklist relevant to your research is completed and uploaded as a Related Manuscript file type with the revised article.

Editorial Policy: Policy requirements

Furthermore, please align your manuscript with our format requirements, which are summarized on the following checklist:
Communications Earth & Environment formatting checklist

In the event that the manuscript is accepted we will be providing further guidance on formatting but please ensure that the manuscript generally complies with our house style at this stage. The main points are as follows:

* ABSTRACT: less than 150 words and accessible. It should include the background and context of the work, the phrase ‘Here we’ (present/show/suggest) to indicate where the description of your own work starts, and then the methods/data, main results and conclusions of the paper.

*The text must be split into:
- INTRODUCTION (<1000 words): includes the background and rationale for the work. The final paragraph should be a brief summary of the main results and conclusions. The results of the current study should only be discussed in this final paragraph.
- RESULTS: split into subheaded sections; ensure that the subheadings are no longer than 60 characters including spaces.
- DISCUSSION: without subheadings.
- METHODS: split into subheaded sections; ensure that the subheadings are no longer than 60

characters including spaces

* SUPPLEMENTARY INFORMATION should be organised logically, with all items labelled as one of the following item types, and cited in the main article:
- Supplementary Figures, labelled and referred to as i.e. Supplementary Figure 1 throughout both the Supplementary Information and the main text
- Supplementary Tables, labelled as above
- Supplementary Notes, labelled as above
- Supplementary Discussion
- Supplementary Methods
- Supplementary References

Data: All Communications Earth & Environment manuscripts must include a section titled "Data Availability" at the end of the Methods section or main text (if no Methods). More information on this policy, and a list of examples, is available at http://www.nature.com/authors/policies/data/data-availability-statements-data-citations.pdf.
In addition, Communications Earth & Environment endorses the principles of the Enabling FAIR data project (http://www.copdess.org/enabling-fair-data-project/). We ask authors to make the data that support their conclusions available in permanent, publically accessible data repositories. (Please contact the editor if you are unable to make your data available).
In particular, the Data availability statement should include:
- Unique identifiers (such as DOIs and hyperlinks for datasets in public repositories)
- Accession codes where appropriate
- If applicable, a statement regarding data available with restrictions
- 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.

If a community resource is unavailable, data can be submitted to generalist repositories such as figshare or 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.

Please refer to our data policies at http://www.nature.com/authors/policies/availability.html.

REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

The manuscript presents a very interesting study on a topic of high relevance. Results are novel, as studies of atmospheric microplastics are scarce, and in particular we lack knowledge on open ocean and remote ocean atmospheric plastic load. These results need to be made available to the scientific community. The study methodology is sound, the techniques applied robust and widely recognized and applied in this field. The blanks and contamination control well performed and reported. The figures are informative and there is a comprehensive literature, which however I found sometimes a bit outdated. I would suggest to review it including more recent studies in marine microplastics research because the field is rapidly expanding and in this respect decade-old studies might be too obsolete in the light of new measurements, definitions, reports.
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Specific comments below

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If particles are quite big, can you say with high confidence that they should have an origin in the immediate water below or within a short distance?
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• Line 38-39 other recent and relevant references worth considering here
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o Worm B, Lotze HK, Jubinville I, Wilcox C, Jambeck J (2017) Plastic as a Persistent Marine Pollutant. Annual Review of Environment and Resources 42(1):1-26. doi:10.1146/annurev-environ-102016-060700
• Line 39-40 more than MP degradation I would say persistence is a concern because MPs do not disappear, but rather gets smaller and therefore incorporated easily in an increasing number of environmental matrices / marine compartments. It is thus difficult (impossible) to remove them, and their toxicity, when more smaller particles are present, will affect a greater fraction of marine organisms from any point of view: physical (ingestion), chemical (leaching), etc..
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• Line 48 Liu et al. 2019 is also over remote parts of the ocean or at least part of it, or?
Methods and SI
• Line 163: why 0.8 µm poresize? Just wondering if there is a specific reason.
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• Pag. 4 SI: “the cellulose…surface” this sentence is incomplete.
• Pag. 6 SI: I understand that you had to rule these particles out. But ship contamination may be one of the most relevant sources in the open ocean when considering that particles larger than 5 µm are maybe too big for being transported over large distances in a certain amount of time. You briefly say (line 122, main text) that some particles may originate from other boats but what if this is the most relevant source? It would be important to include the degree of biofouling of the particles found, or some observations in this respect.
• Pag. 7 SI: “Most importantly..none of the particles” you could add “besides polyester, cellulose and Mortoperm blue”
Results and discussion
• Line 89 Biofouling plays an important role. Size is relevant for biofouling and sinking processes too. Do you think particles >5 µm were more inclined to stay in the sea-surface microlayer or sink? I think it would be interesting to add some discussion referring to these points.
• Line 92: why are 48 hours chosen? Is it comparable to marine aerosols sampling protocols over that region? How long do particles >5 µm stay in the atmosphere? Some insights into physical

properties of aerosols I think would be interesting, just as comparison, given the fact that these are two classes of completely distinct particles, but may there be some behavioral similarities?

• Line 106: morphological scanning: what is the average concentration per volume of air of the particles collected, if this is possible to retrieve from the analysis? How many particles per filter were found (where you found plastics)? Did you find a similar degree of biofouling by observing seawater and air samples? Were you in a high/low contaminated region in terms of seawater concentration? You were in an area close/or passed by the North Atlantic Gyre. Maybe some information on current knowledge of the geographic area in terms of plastic concentration in the seawater should be added, since there are datasets available (e.g. Sea Semester Study - Lavender Law, Osle et al. 2019)

• Line 120: I do not fully agree here. Particles, especially if they stay in the sea-surface, will likely ultimately sink because in the surface microlayer they may encounter high amounts of organic matter and thus get incorporated, increase density, etc..(e.g. Galgani and Loiselle 2019, doi:https://doi.org/10.3390/geosciences9020066).

• Did you observe biofilm-like features on the AMP? If not, it may also be very likely that they have recently derived from land or other boats. Hints/ideas on processes that may keep and control particles on the surface should be added. There are quite a bunch of studies on processes making particles sink or dealing with vertical distribution e.g.:

o Kaiser D, Kowalski N, Waniek JJ (2017) Effects of biofouling on the sinking behavior of microplastics. Environmental Research Letters 12 (12):124003. doi:10.1088/1748- 9326/aa8e8b,

o Koelmans AA, Kooi M, Law KL, van Sebille E (2017) All is not lost: deriving a top-down mass budget of plastic at sea. Environmental Research Letters 12 (11):114028. doi:10.1088/1748-9326/aa9500,

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How would you explain that these particles remain on the surface ocean? What about AMP sources? Bubble bursting, are they transported from below, or do they reside in the surface microlayer?

• Line 124: measured means found?

• Line 135: the smaller the particle, the more likely it is to be ingested and sink, incorporated into aggregates. How would you relate it to your results? How does the distribution of MP in the marine environment relate to your results? Some suggestions should be given here. An increase in small plastic fragments facilitates the interaction with microbes and particles' incorporation into aggregates (Galloway et al. 2017, nteractions of microplastic debris throughout the marine ecosystem. Nature Ecology & Evolution 1:0116. doi:10.1038/s41559-017-0116)

• Line 142: Specify severe consequences.

• Line 144:Products like Mortoperm blue?

• Line 146: Royer et al., 2018 Production of methane and ethylene from plastic in the environment. PLOS ONE 13 (8):e0200574. doi:10.1371/journal.pone.0200574 it may be relevant for your discussion on the effect of PE leachates in air and at the interface water-air when exposed to solar radiation

• Line 146: greater harm is too vague

• Line 148-150 Very general, please specify, give some examples of what possible harm/effects. There are studies on the effects on primary and secondary producers. E.g.:Tetu SG, Sarker I, Schrameyer V, Pickford R, Elbourne LDH, Moore LR, Paulsen IT (2019) Plastic leachates impair growth and oxygen production in Prochlorococcus, the ocean's most abundant photosynthetic bacteria. Communications Biology 2 (1):184. doi:10.1038/s42003-019-0410-x

• Also, a major byproduct of irradiation of PE, PP and EPS is DOC and it may have important relations to bacteria and organic matter even in aerosols, see: Z Zhu L, Zhao S, Bittar TB, Stubbins A, Li D (2020) Photochemical dissolution of buoyant microplastics to dissolved organic carbon: Rates and microbial impacts. Journal of Hazardous Materials 383:121065. doi:https://doi.org/10.1016/j.jhazmat.2019.121065
• Line 152: what is it meant by "devastating effects"? Some communities may benefit from plastic leachates. This sentence is not clear nor well explained. See also Kühn S, Bravo Rebolledo EL, van Franeker JA (2015) Deleterious Effects of Litter on Marine Life. In: Bergmann M, Gutow L, Klages M (eds) Marine Anthropogenic Litter. Springer International Publishing, Cham, pp 75-116. doi:10.1007/978-3-319-16510- 3_4
• Line 154: the paper by Liu et al (2019) also detects AMP. Maybe rephrase or stress the novelty of your results. You may add that you have been sampling in the North Atlantic Gyre or close by, one of the highest accumulation zones for plastic debris… Lines 154-156: what possible effects would you suggest? The conclusion here is quite weak.
• Reference 42 is missing in the references list

Reviewer #2 (Remarks to the Author):

Review of 'Airborne microplastic particles in the marine atmosphere' by Trainic and co-workers. Trainic and colleagues report on airborne microplastic data from the Tara Pacific expedition in the North Atlantic Ocean. The authors used Raman spectroscopy to identify the chemical nature of particles (> 5 um) collected from the atmosphere and seawaters and found the overlap of polymer types between two environmental compartments. Therefore, the authors claim that the ocean is a source of airborne microplastics. While the overall topic of ocean plastic is interesting, the contribution of Trainic and colleagues is not novel as they stated and lack a number of criteria that are important for a well-constrained scientific publication:
Novelty – airborne microplastic from the oceanic atmosphere is not a new topic and has been already surveyed by some research. The recent papers on this matter are from Bergmann et al. (2019, Science Advances) who showed that MP prevails in snow from the European sampling sites to the Arctic. Liu et al. (2019, EST) did a survey of airborne MP in the Pacific Ocean. Despite the authors claimed that this study is over the remote ocean, the major polymers (PE, PP, PS, Figure 1A) in this study was found in coastal and off-coastal samples, which is similar to Liu et al.
Open questions –
Introduction:
Line 38: "transport pathogens" is not rigorous. "potential pathogens" is proper.
Line 47-48: based on your sampling area and positive samples, it is not the first study for this topic.
Methods:
1)it is very good that these authors employed two-step measurements. I did not find any actions about checking your filter papers prior to being used for sampling. Based on my experiences, plastics especially fibrous particles are commonly presented on new filters.
2) did you have replicates of your samples? If it does, please clarify this.
3) It is very blurred that how the filters were scanned by Raman spectroscopy. First of all, it looks like a non-consistent scanning strategy was employed in this study. The particles usually present patchy

patterns on filters. If you employed a subsampling method (especially not consistent subsampling approaches), the results are largely biased. This problem is worse if you focus on the smaller MP. This research claimed to focus on MP > 5um, which is a relatively lower size fraction for MP research. Unexpectedly, I did not find any detailed information about the MP size (besides the Table 1).

Results and discussion:

1) Line-62-63: Again, the research theoretically focused on AMP as small as 5 um depending on the Raman spectroscopy. But their methods and results did not support this statement.

2) It is difficult to conclude that Samples 38-39 sharing PE and PP with aerosol samples provide evidence for the marine source of the AMP. No direct evidence present here. Pursing the origins of MP is very important and hard in this field. “The marine source of the AMP” seems to be the sale point in this manuscript, but your evidence is not enough and solid. Furthermore, the Raman spectrum of PE between the seawater and aerosol samples is different at the lower range.

Finally, the text flow is not straight forward. I had to read some text passages a few times and/or to flip back and forth to understand how the experiments and analyses were set up.

COMMSENV-20-0111

The manuscript presents a very interesting study on a topic of high relevance. Results are novel, as studies of atmospheric microplastics are scarce, and in particular we lack knowledge on open ocean and remote ocean atmospheric plastic load. These results need to be made available to the scientific community. The study methodology is sound, the techniques applied robust and widely recognized and applied in this field. The blanks and contamination control well performed and reported. The figures are informative and there is a comprehensive literature, which however I found sometimes a bit outdated. I would suggest to review it including more recent studies in marine microplastics research because the field is rapidly expanding and in this respect decade-old studies might be too obsolete in the light of new measurements, definitions, reports.
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Specific comments below

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Results and discussion

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Answer to reviewers: revisions for the manuscript: 'Airborne microplastic particles in the marine atmosphere' in *Communications Earth & Environment*

On behalf of the co-authors of this manuscript, we would like to thank the reviewers for their helpful and thorough review of our work. We believe we were able to address the editor's and reviewers' concerns, and provide compelling evidence for non-negligible amounts of airborne marine plastics in the open ocean, away from the coast, including full details on the methodology, and discuss the findings in the context of the latest literature more comprehensively and specifically.

Below please find our point-by-point response to each of the reviewers' comments.

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The manuscript presents a very interesting study on a topic of high relevance. Results are novel, as studies of atmospheric microplastics are scarce, and in particular we lack knowledge on open ocean and remote ocean atmospheric plastic load. These results need to be made available to the scientific community. The study methodology is sound, the techniques applied robust and widely recognized and applied in this field. The blanks and contamination control well performed and reported. The figures are informative and there is a comprehensive literature, which however I found sometimes a bit outdated. I would suggest to review it including more recent studies in marine microplastics research because the field is rapidly expanding and in this respect decade-old studies might be too obsolete in the light of new measurements, definitions, reports.

Overall the text present is well written, the style and language are appropriate and no major grammar mistakes are present. I have however some concerns on its current form: I think both the introduction and discussion need major improvement, and therefore, suggest major revision of the manuscript. I had the impression that very general statements are made throughout the manuscript, which however remain very vague and superficial despite citing may papers. Maybe all the relevant references are cited but they do not find a sufficiently explained corresponding discussion and conceptualization in the text. I think there the authors should really focus on

explaining processes, impacts, mechanisms, or propose ones. As an example, line 147 “great harm” and below “devastating effects” do not make much sense in my opinion.

Specific comments below

Introduction

In general I think the introduction is too general and vague, lacking important information, and needs improvement. I think you should explain why finding MP in the atmosphere might be relevant. For example, could they transport contaminants/other microorganisms (and relative effects) over large areas quicker than oceanic currents? Via atmospheric transport, could they easily reach remote, pristine areas (e.g. protected areas).

Thank you very much. The introduction was changed according to the reviewer’s suggestions. Information about the implications of the presence of AMP was added, lines 51-54: “Marine AMP may have implications on marine ecology; facilitated by the winds, AMP can spread potential pathogens and persistent organic pollutants adsorbed onto their surface [1] by an order of magnitude faster than the ocean currents [2,3]. Furthermore, the components adsorbed to the AMP are exposed to UV radiation and oxidation, which can increase their toxicity to marine biota [4-7].”

In the size of 5-10 µm, could they have effects similar to PM10 particulates if the ocean is a (secondary) source of these contaminants? Being the analysis focused on >5 µm particles, how would you compare their residence time and behavior with respect to larger marine aerosols? If particles are quite big, can you say with high confidence that they should have an origin in the immediate water below or within a short distance?

Thank you very much for raising this important issue. To fully address these questions, we have performed an elaborate estimation of the expected residence times for the sizes of the AMP we found by calculating their terminal velocities. The full calculations of the terminal velocities can now be found in the SI methods section:

“AMP terminal velocity calculations

AMP terminal velocity was estimated using the well-established theory and measurements of settling velocity of water droplets and ice crystals with needle shapes, in the range ~10 to 100 μm [8]. The particles shape, sizes and densities that are close to water, allowed us to use this theory.

The terminal velocity of elongated particles in such size range, scales to the equivalent sphere of the same mass but with radius that scales to the largest dimension of the particle. This implies that the density of the equivalent sphere will be reduced by the ratio (α) of the real volume (V_r) of particle and the volume (V_s) of the equivalent sphere:

$$\alpha = \frac{V_r}{V_s}$$

Then, to a good approximation for the AMP terminal velocity (μ) could be scaled as:

$$\mu = \alpha \mu_s$$

Where μ_s is the terminal velocity of a sphere (droplet) with diameter of the polymers major axis.

More specifically: The real volume of the MP particle can be approximated as a spheroid by $V_r = \frac{4}{3}\pi a b^2$, where a is the major and b is the minor axis and equivalent sphere volume is $V_s = \frac{4}{3}\pi a^3$. Assuming that the MP density is close to water density, $\rho_{mp} \approx \rho_w$, yields:

$$\alpha = \frac{V_r}{V_s} = \left(\frac{b}{a}\right)^2$$

The terminal velocity of spheres can be described as a power-low function of the radius r, $V_t = \kappa r^{\eta}$ where, κ and η depend on the sizes. Droplets with radii $r < 40\mu m$ are in the laminar regime for which $\kappa \approx 1.19x10^6\ cm^{-1}s^{-1}$, and $\eta = 2$. Droplets in the range of $40 < r < 600\mu m$ are in the intermediate regime between laminar and turbulent with $\kappa \approx 8x10^3\ s^{-1}$ and $\eta = 1$ [9,10].

Applying the above scaling to the measured AMP particles yields settling velocity in the range of 0.001 - 0.2 m sec^{-1} for the particles. For the most elongated cases, despite having long major axis, their settling velocities are in the range of 0.001 m sec^{-1} (0.1 cm sec^{-1})."

We added the conclusions from the calculations in several places in the Results and Discussion section of the main text:

Lines 103-120: "Next, we assessed the atmospheric residence times of the AMP we found by calculating their settling (terminal) velocities (Table 1; see SI methods for the detailed calculations). The residence time of atmospheric particles is likely to be affected by their atmospheric terminal velocities. Slower velocities (≤0.01m sec^{-1}) imply higher likelihood to be lifted from near the water surface by eddies and to be carried by the surrounding air [11]. We found AMP settling velocities in the range of 0.001 - 0.2 m sec^{-1} for the particles (Table 1, SI methods). By combining the HYSPLIT back trajectories with the obtained residence times, we can evaluate the source of the particles. Particles 2, 17, 30 and 38 were clearly a product of local emission, with residence times of minutes to several hours (Table 1). The rest, with residence times of 1-2 days, combined with the HYSPLIT back trajectories, also appear to have been formed in the ocean. This suggests that the AMP found in this study are emitted from the seawater, indicating wind-related processes via bubble bursting.

AMP particles with irregular and elongated shapes can have long atmospheric residence times. For the most elongated AMP, despite having long major axis, their atmospheric settling velocities are in the range of 0.001 m sec^{-1}, equivalent to typical submicron particles, which are suspended in the marine atmosphere in a timescale of days, and dominate the marine boundary layer [12]. We summarize that the AMP we found can remain in the atmosphere up to ~2 days, and for a typical marine wind speed of 8 ms^{-1} can be transported to 100's -1000's km. However, AMP with settling velocities in the order of ~0.1 m sec^{-1} will remain in the atmosphere for ~5 min and travel ~ 3km [12]."

Lines 176-184: ". Given that some AMP can persist in the air for days according to our terminal velocity calculations, allowing long exposure times to UV radiation and oxidation, they can become highly toxic [4-7] before being re-deposited into the seawater, where they can cause more harm to the marine ecosystem than non-airborne microplastic particles. The ability of microplastic to carry pathogens [1,4,5,13-21] may also increase the harm posed by AMP to the marine

ecosystem, as airborne microplastic can carry pathogens further than typical ocean currents. According to our calculations some AMP can travel 100's to 1000's of km, rendering distant marine ecosystems in danger of infection by these pathogens, provided they remain infective in the atmosphere, as shown possible in several studies [22,23]."

• Line 34 you could define primary and secondary MP, as probably you detected secondary ones in your samples.

Thank you. The following sentence was added to lines 32-34: "Primary microplastic is produced either for direct use or as a precursor for other products, while secondary microplastics are formed in the environment from breakdown of larger plastic material, especially marine debris [4,5]."

• Line 35-36 vague sentence: specify, give examples of reasons of concern, physically, chemically, biologically…like Ingestion, habitat damage, entangling, transport of invasive species, etc..

Thank you. We elaborate on the specific potential damage of MP in lines 36-39: "MP may pose a risk to aquatic environments due to their documented ubiquity in the marine ecosystems, their long lifetime in the environment, their ability to transport potential pathogens and persistent organic pollutants [1], their role as a new source of toxic compounds in the marine environment, and their propensity to be ingested by diverse organisms [4,5,7,20,21,24-28]."

• Line 38-39 other recent and relevant references worth considering here

o Burns EE, Boxall ABA (2018) Microplastics in the aquatic environment: Evidence for or against adverse impacts and major knowledge gaps. Environmental Toxicology and Chemistry 37 (11):2776-2796. doi:10.1002/etc.4268

o Villarrubia-Gómez P, Cornell SE, Fabres J (2018) Marine plastic pollution as a planetary boundary threat – The drifting piece in the sustainability puzzle. Marine Policy 96:213- 220. doi:https://doi.org/10.1016/j.marpol.2017.11.035

o Hammer J, Kraak MHS, Parsons JR (2012) Plastics in the Marine Environment: The Dark Side of a Modern Gift. In: Whitacre DM (ed) Reviews of Environmental Contamination and Toxicology. Springer New York, New York, NY, pp 1-44. doi:10.1007/978-1-4614- 3414-6_1

o Worm B, Lotze HK, Jubinville I, Wilcox C, Jambeck J (2017) Plastic as a Persistent Marine

Pollutant. Annual Review of Environment and Resources 42(1):1-26. doi:10.1146/annurev-environ-102016-060700

Thank you. All references suggested by the reviewer were added, as well as additional references which were very recently published, lines 34-39: “Marine MP are a subject of elaborate research, and a matter of great concern in the fields of marine pollution, chemistry and ecology [28-39]. MP may pose a risk to aquatic environments due to their documented ubiquity in the marine ecosystems, their long lifetime in the environment, their ability to transport potential pathogens and persistent organic pollutants [1], their role as a new source of toxic compounds in the marine environment, and their propensity to be ingested by diverse organisms [4,5,7,20,21,24-28].” (See reference list below).

• Line 39-40 more than MP degradation I would say persistence is a concern because MPs do not disappear, but rather gets smaller and therefore incorporated easily in an increasing number of environmental matrices / marine compartments. It is thus difficult (impossible) to remove them, and their toxicity, when more smaller particles are present, will affect a greater fraction of marine organisms from any point of view: physical (ingestion), chemical (leaching), etc..

Thank you very much for the clarification. We have changed the sentence, lines 40-45: “Marine microplastic persistence is a major source of concern, since plastic entering marine ecosystems doesn’t degrade, but rather undergoes fragmentation through mechanical erosion and photooxidation via interaction with ultraviolet (UV) radiation, resulting in fragmentation to nano-plastic sizes (< 1 µm) [4]. The micro and nano-plastic fragments are easily incorporated in an increasing number of environmental matrices, making them impossible to remove. The effect of MP toxicity on marine biota increases with decreasing size, since smaller sizes facilitate ingestion, as well as chemical processes such as leaching [4,28].”

• Line 44: a more recent reference for plastic abundance. Erni-Cassola G, Zadjelovic V, Gibson MI, Christie-Oleza JA (2019) Distribution of plastic polymer types in the marine environment; A meta-analysis. Journal of Hazardous Materials 369:691-698. doi:https://doi.org/10.1016/j.jhazmat.2019.02.067

Also, consider that only 1% of plastic introduced in marine systems is observed floating at sea

(Oberbeckmann and Labrenz 2020, Marine Microbial Assemblages on Microplastics: Diversity, Adaptation, and Role in Degradation. Annual Review of Marine Science 12 (1):null. doi:10.1146/annurev-marine-010419-010633)

Thank you. The reference was added, lines 74-76, and 84-85: "We detected AMP in the marine atmosphere from the airborne samples collected in the North Atlantic Ocean, and found that most of them correspond to the most ubiquitous MP found in the marine environment [32,40-44] (see Figure 1, Table 1)…

PS, PE, and PP were previously identified as common plastic pollutants in the waters of remote ocean locations and shores worldwide [32,40-42,44,45]."

Lines 36-40: "MP may pose a risk to aquatic environments due to their documented ubiquity in the marine ecosystems, their long lifetime in the environment, their ability to transport potential pathogens and persistent organic pollutants [1,46], their role as a new source of toxic compounds in the marine environment, and their propensity to be ingested by diverse organisms [4,5,7,20,21,24-28]. "

Lines 181-183: "The ability of microplastic to carry potential pathogens [1,4,5,13-21,46] may also increase the harm posed by AMP to the marine ecosystem, as airborne microplastic can carry pathogens further than typical ocean currents."

• Line 48 Liu et al. 2019 is also over remote parts of the ocean or at least part of it, or?

Yes. They define a remote region at 10°-20° from the continental coast, which is indeed remote, but it is located near the Mariana Islands, which are populated. They could not, therefore, claim a remote oceanic source for their particles, and indeed have not done so. On the contrary, they claim their atmospheric plastic source is terrestrial, since they were either in the vicinity of a continent or an island. While two of our samples were taken relatively close to shore (~5°), most are located between 10° and 45° from any land (continents or populated islands). Therefore, we are the first to sample in the open ocean/ remote ocean.

Methods and SI

• Line 163: why 0.8 µm poresize? Just wondering if there is a specific reason.

There is a technical reason. We used vacuum pumps in order to draw air onto our filters. We found that the same filters with lower power size decrease the air flowrate substantially. Therefore we chose the pore size that inspires the least resistance on the system while still allowing us to collect submicron particles as well.

• Pag. 3 SI: “as the plastic debris…” incomplete sentence

Corrected.

• Pag. 4 SI: “the cellulose…surface” this sentence is incomplete.

Corrected.

• Pag. 6 SI: I understand that you had to rule these particles out. But ship contamination may be one of the most relevant sources in the open ocean when considering that particles larger than 5 µm are maybe too big for being transported over large distances in a certain amount of time. You briefly say (line 122, main text) that some particles may originate from other boats but what if this is the most relevant source? It would be important to include the degree of biofouling of the particles found, or some observations in this respect.

Thank you very much. Contamination from other boats could certainly be a major source of airborne marine microplastic. However, we didn’t find any plastic on the surfaces of our boat, except for polyester (which wasn’t found in the atmospheric samples). We found cellulose, (which isn’t plastic), and organic dyes, which are used for plastics but are not themselves considered as plastic. So, the R/V Tara in itself is not a source. We did find plastic in the seawater, some of which corresponds to the plastic in our airborne samples. Therefore, boat contamination isn’t ruled out. As for biofouling, we do not currently have data about the degree of biofouling, but we plan to analyze the chemical and biological compounds adsorbed onto the microplastic particles in future work, which will be dedicated to that. Not only in the sense of biofouling, but also the adsorption of organic material onto the plastic.

• Pag. 7 SI: “Most importantly..none of the particles” you could add “besides polyester, cellulose and Mortoperm blue”

Thank you. For clarity, we chose to keep the sentence as originally written.

Results and discussion

• Line 89 Biofouling plays an important role. Size is relevant for biofouling and sinking processes too. Do you think particles >5 µm were more inclined to stay in the sea-surface microlayer or sink? I think it would be interesting to add some discussion referring to these points.

Large particles, in the order of mm's stay on the sea-surface in high concentrations, as reported in the literature, and also observed by our seawater measurements presented in Figure 1, given as seawater microplastic particle (MP) log abundance per m^3. The AMP we observed are polymers with low densities (lower than that of the seawater), more likely to remain in the sea surface microlayer, and are more readily emitted into the atmosphere (the densities of the plastics we found are presented in Table 1).

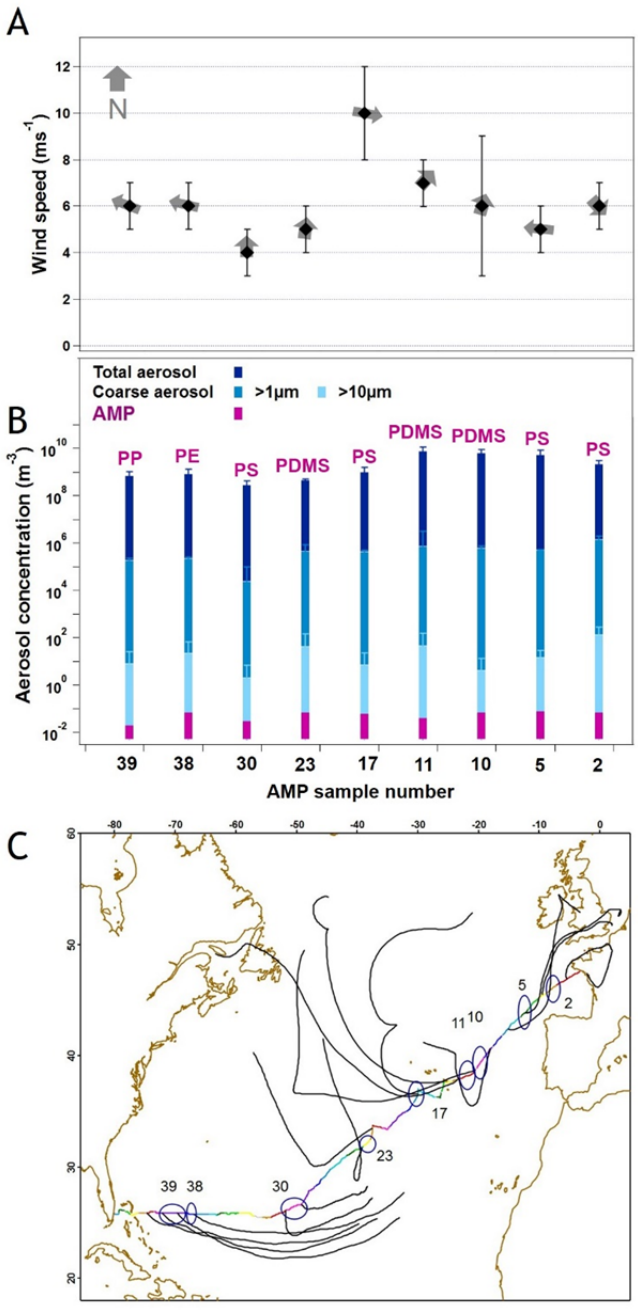
• Line 92: why are 48 hours chosen? Is it comparable to marine aerosols sampling protocols over that region? How long do particles >5 µm stay in the atmosphere? Some insights into physical properties of aerosols I think would be interesting, just as comparison, given the fact that these are two classes of completely distinct particles, but may there be some behavioral similarities?

Thank you, these are all crucial points, and therefore, in order to answer them properly we added a full calculation of the expected residence time of the particles through estimation of their terminal velocity (see above; manuscript SI methods section). 48 hours are ≥ the residence time expected for these type/ size of particles (and for aerosol particles in general), but we want to be 'on the safe side' to make sure, as much as possible, that we obtain the full trajectory that the particles went through (see manuscript Table 1; Figure 2C).

This is elaborated in the text, see lines 159-162: "Furthermore, the specific densities of most AMP we found, and specifically polyethylene and polypropylene found in the seawater and in AMP samples 38 and 39, are lower than the specific density of seawater (Table 1), and are therefore likely to accumulate in the sea surface microlayer and thus can be readily aerosolized."

• Line 106: morphological scanning: what is the average concentration per volume of air of the particles collected, if this is possible to retrieve from the analysis? How many particles per filter were found (where you found plastics)?

Thank you very much for this question. We changed Figure 2 entirely, and added the AMP concentration per volume of air to address this important point:



" Figure 2. AMP and total aerosol concentrations related to wind and air mass back trajectories. A. Average wind speed and direction (grey arrows) [47] during AMP sample collection B. AMP, total and coarse mode aerosol concentrations per m^3 for each sample. AMP are marked in magenta, total aerosol are dark blue, and coarse mode aerosol are marked in cyan and blue

for >10 µm and >1 µm, respectively. Sample numbers are presented in the direction following the R/V *Tara* transect shown on the map C. Map of 48 hr HYSPLIT back trajectory ensembles for each point along the R/V *Tara* transect in the Atlantic Ocean where AMP were detected [47]. These are average trajectories of the HYSPLIT 'Ensemble option' that were calculated based on an endpoint at 250 m height, the minimum height for the optimal configuration of the ensemble. The vast majority of the air masses originate from the ocean for the full extent of the 48 hr. "

A discussion of the results presented in Figure 2 is found in the paper in lines 93-103: "AMP concentrations are presented in Figure 2, together with total and coarse mode aerosol concentrations [47]. Wind speed and direction prevailing during AMP sampling periods are presented for reference. AMP concentrations are in the order of 1-10% of the coarse mode aerosol concentrations for sizes > 10 µm. The AMP concentrations obtained here are in good agreement with concentrations attributed to marine source AMP, which were obtained from measurements conducted at the French Atlantic coast [48]. Based on their coastal measurements, the authors suggested that AMP can be formed via bubble bursting, which may also occur in the open ocean.

In order to investigate the open ocean as a source of the AMP obtained in our study, we conducted 48 h back trajectories at 250m altitude (HYSPLIT) for each point along the *Tara* North Atlantic transect where AMP were detected. The vast majority of the air masses passed over the ocean during the full 48 hr trajectories (see Figure 2C)."

Did you find a similar degree of biofouling by observing seawater and air samples?

Were you in a high/low contaminated region in terms of seawater concentration?

Thank you for this important point. As mentioned above, we do not currently have data about the degree of biofouling, but plan to analyze the compounds adsorbed onto the microplastic particles in future work. As for the seawater concentrations – they varied throughout the course of the transect, as presented in Figure 1A (full circles on the map).

You were in an area close/or passed by the North Atlantic Gyre. Maybe some information on current knowledge of the geographic area in terms of plastic concentration in the seawater

should be added, since there are datasets available (e.g. Sea Semester Study - Lavender Law, Osle et al. 2019)

Thank you very much. We mention the current knowledge on marine MP in the introduction, including the latest estimates on the concentrations of MP in the oceans. Lines 49-50: "Estimates of recent years suggest that ~5.25 trillion plastic fragments are currently floating on the oceans' surface [25]."

As for the concentration in our transect, we present our own dataset, obtained by R/V Tara throughout the transect, in Figure 1.

• Line 120: I do not fully agree here. Particles, especially if they stay in the sea-surface, will likely ultimately sink because in the surface microlayer they may encounter high amounts of organic matter and thus get incorporated, increase density, etc..(e.g. Galgani and Loiselle 2019, doi:https://doi.org/10.3390/geosciences9020066).

They will ultimately sink, that is a valid possibility. However, that process takes time, and the evidence that microplastic remains at the sea surface is demonstrated by the high concentrations of MP found all over the Atlantic ocean (as well as the Pacific), even in the most remote areas, due to their low densities, mostly lower than those of seawater.

• Did you observe biofilm-like features on the AMP? If not, it may also be very likely that they have recently derived from land or other boats. Hints/ideas on processes that may keep and control particles on the surface should be added. There are quite a bunch of studies on processes making particles sink or dealing with vertical distribution e.g.:
o Kaiser D, Kowalski N, Waniek JJ (2017) Effects of biofouling on the sinking behavior of microplastics. Environmental Research Letters 12 (12):124003. doi:10.1088/1748- 9326/aa8e8b,
o Koelmans AA, Kooi M, Law KL, van Sebille E (2017) All is not lost: deriving a top-down mass budget of plastic at sea. Environmental Research Letters 12 (11):114028. doi:10.1088/1748-9326/aa9500,
o Kooi M, Nes EHv, Scheffer M, Koelmans AA (2017) Ups and Downs in the Ocean: Effects of Biofouling on Vertical Transport of Microplastics. Environmental Science & Technology 51 (14):7963-7971. doi:10.1021/acs.est.6b04702
How would you explain that these particles remain on the surface ocean? What about AMP

sources? Bubble bursting, are they transported from below, or do they reside in the surface microlayer?

MP are found in the ocean surface (Ref 1-15 in the paper; and the data from the current Tara expedition, Figure 1A) due to their low densities, which allow them to be buoyant. As the reviewer stated, and was demonstrated in previous studies, they will be subject to biological and chemical adsorption, which will eventually increase their density and cause them to sink. However, the amounts of plastic found in the ocean surface are irrefutable. Those MP can be readily emitted into the atmosphere. The bubble bursting process may incorporate particles from below the surface, when the bubble is rising, or from the microlayer itself, when the bubble reaches the surface. The smaller/lighter the particle, the more readily it can be incorporated into the rising bubble.

• Line 124: measured means found? Yes, corrected.

• Line 135: the smaller the particle, the more likely it is to be ingested and sink, incorporated into aggregates. How would you relate it to your results? How does the distribution of MP in the marine environment relate to your results? Some suggestions should be given here. An increase in small plastic fragments facilitates the interaction with microbes and particles' incorporation into aggregates (Galloway et al. 2017, nteractions of microplastic debris throughout the marine ecosystem. Nature Ecology & Evolution 1:0116. doi:10.1038/s41559-017-0116)

True, however, it is a continuous process where new 'small MP' keep being formed by the breakdown of larger plastic material, especially marine debris ([4,5]). See lines 34-36 and 42-44 in the paper:

"Primary microplastic is produced either for direct use or as a precursor for other products, while secondary microplastics are formed in the environment from breakdown of larger plastic material, especially marine debris [4,5]."

"Marine microplastic persistence is a major source of concern, since plastic entering marine ecosystems doesn't degrade, but rather undergoes fragmentation through mechanical erosion and photooxidation via interaction with ultraviolet (UV) radiation, resulting in fragmentation to nano-plastic sizes (< 1 µm) [4]."

• Line 142: Specify severe consequences.

We elaborate on this point and specify the severe consequence in the paragraph blow, lines 175-187: “It is known that micro and nano-plastics, particularly polystyrene, polyethylene and polypropylene (all found in our AMP samples), accumulate persistent organic pollutants [49] as well as heavy metals [40,50,51]. The products of plastic degradation, particularly polystyrene, found in our AMP samples, are toxic [7,51-53]. Given that some AMP can persist in the air for days according to our terminal velocity calculations, allowing long exposure times to UV radiation and oxidation, they can become highly toxic [4-7] before being re-deposited into the seawater, where they can cause more harm to the marine ecosystem than non-airborne microplastic particles. The ability of microplastic to carry potential pathogens [1,4,5,13-21,46] may also increase the harm posed by AMP to the marine ecosystem, as airborne microplastic can carry pathogens further than typical ocean currents. According to our calculations some AMP can travel 100’s to 1000’s of km, rendering distant marine ecosystems in danger of infection by these pathogens, provided they remain infective in the atmosphere, as shown possible in several studies [22,23]. Therefore, the effect of AMP can be devastating for marine microbial ecology, and must be thoroughly investigated [24,28].”

• Line 144:Products like Mortoperm blue?

No, Mortoperm blue is an additive, an organic dye used to paint plastic compounds. The products were previously studied [54].

• Line 146: Royer et al., 2018 Production of methane and ethylene from plastic in the environment. PLOS ONE 13 (8):e0200574. doi:10.1371/journal.pone.0200574 it may be relevant for your discussion on the effect of PE leachates in air and at the interface water-air when exposed to solar radiation

Thank you, reference was added to lines 177-178: “The products of plastic degradation, particularly polystyrene, found in our AMP samples, are toxic [7,51-53].”

• Line 146: greater harm is too vague

There are studies on the effects on primary and secondary producers. E.g.:Tetu SG, Sarker I, Schrameyer V, Pickford R, Elbourne LDH, Moore LR, Paulsen IT (2019) Plastic leachates impair

growth and oxygen production in Prochlorococcus, the ocean's most abundant photosynthetic bacteria. Communications Biology 2 (1):184. doi:10.1038/s42003-019-0410-x

• Also, a major byproduct of irradiation of PE, PP and EPS is DOC and it may have important relations to bacteria and organic matter even in aerosols, see: Z Zhu L, Zhao S, Bittar TB, Stubbins A, Li D (2020) Photochemical dissolution of buoyant microplastics to dissolved organic carbon: Rates and microbial impacts. Journal of Hazardous Materials 383:121065. doi:https://doi.org/10.1016/j.jhazmat.2019.121065

Thank you. The text was corrected, and the suggested references were added, lines 178-190: "Given that some AMP can persist in the air for days according to our terminal velocity calculations, allowing long exposure times to UV radiation and oxidation, they can become highly toxic [4-7] before being re-deposited into the seawater, where they can cause more harm to the marine ecosystem than non-airborne microplastic particles. The ability of microplastic to carry potential pathogens [1,4,5,13-21,46] may also increase the harm posed by AMP to the marine ecosystem, as airborne microplastic can carry pathogens further than typical ocean currents. According to our calculations some AMP can travel 100's to 1000's of km, rendering distant marine ecosystems in danger of infection by these pathogens, provided they remain infective in the atmosphere, as shown possible in several studies [22,23]. Therefore, the effect of AMP can be devastating for marine microbial ecology, and must be thoroughly investigated [24,28].

This study is the first to detect and identify AMP in the remote marine atmosphere. The implications of these findings on AMP emissions and their fate, the possible effects on the marine atmosphere and on marine microbial ecology, as suggested above, require further investigation [24,28]."

• Line 148-150 Very general, please specify, give some examples of what possible harm/effects.

Corrected, lines 182-186: "The ability of microplastic to carry potential pathogens [1,4,5,13-21,46] may also increase the harm posed by AMP to the marine ecosystem, as airborne microplastic can carry pathogens further than typical ocean currents. According to our calculations some AMP can travel 100's to 1000's of km, rendering distant marine ecosystems in danger of infection by these pathogens, provided they remain infective in the atmosphere, as shown possible in several studies [22,23]. "

• Line 152: what is it meant by “devastating effects”? Some communities may benefit from plastic leachates. This sentence is not clear nor well explained. See also Kühn S, Bravo Rebolledo EL, van Franeker JA (2015) Deleterious Effects of Litter on Marine Life. In: Bergmann M, Gutow L, Klages M (eds) Marine Anthropogenic Litter. Springer International Publishing, Cham, pp 75-116. doi:10.1007/978-3-319-16510- 3_4

The devastating effects refer to the two cases presented in the previous sentences: 1) the interaction of AMP with UV radiation, resulting in toxic compounds, to which the microorganisms can then be exposed, and 2) pathogens attached to the AMP which may infect distant ecosystems, lines 177-186:

“The products of plastic degradation, particularly polystyrene, found in our AMP samples, are toxic [7,51-53]. Given that some AMP can persist in the air for days according to our terminal velocity calculations, allowing long exposure times to UV radiation and oxidation, they can become highly toxic [4-7] before being re-deposited into the seawater, where they can cause more harm to the marine ecosystem than non-airborne microplastic particles. The ability of microplastic to carry potential pathogens [1,4,5,13-21,46] may also increase the harm posed by AMP to the marine ecosystem, as airborne microplastic can carry pathogens further than typical ocean currents. According to our calculations some AMP can travel 100’s to 1000’s of km, rendering distant marine ecosystems in danger of infection by these pathogens, provided they remain infective in the atmosphere, as shown possible in several studies [22,23].”

• Line 154: the paper by Liu et al (2019) also detects AMP. Maybe rephrase or stress the novelty of your results. You may add that you have been sampling in the North Atlantic Gyre or close by, one of the highest accumulation zones for plastic debris… Lines 154-156: what possible effects would you suggest? The conclusion here is quite weak.

In this concluding paragraph we refer to the possible effects suggested above - 1) the interaction of AMP with UV radiation, resulting in toxic compounds, to which the microorganisms can then be exposed, and 2) pathogens attached to the AMP which may infect distant ecosystems.

The sentence was corrected, lines 188-190: “The implications of these findings on AMP emissions and their fate, the possible effects on the marine atmosphere and on marine microbial ecology, as suggested above, require further investigation [24,28].”

• Reference 42 is missing in the references list

Corrected

Reviewer #2 (Remarks to the Author):

Review of 'Airborne microplastic particles in the marine atmosphere' by Trainic and co-workers. Trainic and colleagues report on airborne microplastic data from the Tara Pacific expedition in the North Atlantic Ocean. The authors used Raman spectroscopy to identify the chemical nature of particles (> 5 um) collected from the atmosphere and seawaters and found the overlap of polymer types between two environmental compartments. Therefore, the authors claim that the ocean is a source of airborne microplastics. While the overall topic of ocean plastic is interesting, the contribution of Trainic and colleagues is not novel as they stated and lack a number of criteria that are important for a well-constrained scientific publication:
Novelty – airborne microplastic from the oceanic atmosphere is not a new topic and has been already surveyed by some research. The recent papers on this matter are from Bergmann et al. (2019, Science Advances) who showed that MP prevails in snow from the European sampling sites to the Arctic. Liu et al. (2019, EST) did a survey of airborne MP in the Pacific Ocean. Despite the authors claimed that this study is over the remote ocean, the major polymers (PE, PP, PS, Figure 1A) in this study was found in coastal and off-coastal samples, which is similar to Liu et al.

Thank you very much for raising this important issue. Bergmann et al. have indirect evidence of long range transport of AMP, the question of the marine source is not addressed. Liu et al measured at a remote region, at 10°-20° from the continental coast, but it is located near the Mariana Islands, which are populated. They could not, therefore, claim a remote oceanic source for their particles, and indeed have not done so. On the contrary, they claim their atmospheric plastic source is terrestrial, since they were either in the vicinity of a continent or an island. While two of our samples were taken relatively close to shore (~5°), most are located between 10° and 45° from any land (continents or populated islands). Therefore, we are the first to sample in the open ocean/ remote ocean.

Open questions –
Introduction:

Line 38: “transport pathogens” is not rigorous. “potential pathogens” is proper. Corrected.

Line 47-48: based on your sampling area and positive samples, it is not the first study for this topic.

Thank you, this is an important point to clarify. As stated above, we are currently the first to sample in the open/ remote ocean, and the first to link the seawater MP to airborne MP in the open ocean. We emphasize it in the manuscript in the results and discussion section:

Lines 63-64: “The main goals of this study were to determine whether airborne microplastic particles (AMP) are present in the open ocean, to chemically identify them, and to investigate their possible sources. ”

Lines 102-104: “In order to investigate the open ocean as a source of the AMP obtained in our study, we conducted 48 h back trajectories at 250m altitude (HYSPLIT) for each point along the *Tara* North Atlantic transect where AMP were detected.”

Lines 153-158: “These evidence suggest that AMP can be produced in the open ocean through wind related processes forming sea spray aerosol particles, a different source and formation mechanism than the terrestrial and coastal AMP detected to date [39,48,55-65]. It also means that the ocean can be a constant global source of AMP, which may be transported to coastal regions and remote locations. However, we cannot dismiss the possibility that some AMP particles may have originated from other boats, continents or islands [65], and transported into the remote marine atmosphere.”

Methods:

1)it is very good that these authors employed two-step measurements. I did not find any actions about checking your filter papers prior to being used for sampling. Based on my experiences, plastics especially fibrous particles are commonly presented on new filters.

Thank you. The following sentence was added to the SI for clarification:

“The handling blanks include the following treatment for filters: new filters taken out from the case, filters inserted and removed from the filter holders without aerosol collection, filters placed in the vicinity of the sample filter during microscopy and/ or Raman analysis.”

2) did you have replicates of your samples? If it does, please clarify this.

We did not have replicates for the polycarbonate samples, which were used for measuring microplastic. The full setup includes 3 different filters, each used for a different purpose, as elaborated in Figure 4 in the paper: "**Figure 4:** Illustration of the atmospheric aerosol collection system onboard the R/V Tara. The system is composed of an inlet, positioned 15m above sea level, which is connected to a flow splitter leading to the filter holders. **Each filter holder contains a different filter, for biological analysis, for chemical characterization, and for morphology using SEM.** Air is being pulled from the inlet through all filter holders by a vacuum pump. The air flow rate through each filter was approximately 20 LPM (liter per minute), as measured by a flow-meter [47]." The filter we used was the polycarbonate for morphological analysis. It is a smooth surface, which allows particle detection and analysis in Raman spectroscopy.

3) It is very blurred that how the filters were scanned by Raman spectroscopy. First of all, it looks like a non-consistent scanning strategy was employed in this study. The particles usually present patchy patterns on filters. If you employed a subsampling method (especially not consistent subsampling approaches), the results are largely biased. This problem is worse if you focus on the smaller MP. This research claimed to focus on MP > 5um, which is a relatively lower size fraction for MP research. Unexpectedly, I did not find any detailed information about the MP size (besides the Table 1).

Thank you. Table 1 summarizes the size of all validated AMP found in the study. As for the scanning strategy, we altered the text in order to better clarify it (SI methods): "An area of ~ 870 mm^2 was scanned in search of particles in each filter (total of 43 filters). The area scanned was the same distance from the edge of the filter for each of the 43 filters. Upon finding a particle or group of particles, we obtained Raman spectra for an area of 0.018 mm^2 in each scan of the particle finder software. Depending on the amount of particles found during the search executed for the entire filter (the ~ 870 mm^2 scan), 30-100 scans were performed using the software, resulting in a total scanned area between 0.54-1.8 mm^2."

Results and discussion:

1) Line-62-63: Again, the research theoretically focused on AMP as small as 5 um depending on the Raman spectroscopy. But their methods and results did not support this statement.

This important point requires better explaining, which we now added. While we detect particles as small as 5 µm or even smaller, we did not find AMP in such small sizes. We found particles

from 1 µm up to 100's µm that were defined as contamination since they were found in the blanks (SI methods): "The size range of the particles, which were found in the blanks and/ or airborne samples was between ~1 µm – 100's µm."

2) It is difficult to conclude that Samples 38-39 sharing PE and PP with aerosol samples provide evidence for the marine source of the AMP. No direct evidence present here. Pursing the origins of MP is very important and hard in this field. "The marine source of the AMP" seems to be the sale point in this manuscript, but your evidence is not enough and solid. Furthermore, the Raman spectrum of PE between the seawater and aerosol samples is different at the lower range.

We thank the reviewer for this important comment. Further to our new residence time calculations, combined with the HYSPLIT back trajectories, and the chemical analysis we were able to provide more compelling evidence for the marine origin of the AMP. Therefore, we made extensive changes to the paper, presented now in lines 102-158:

"In order to investigate the open ocean as a source of the AMP obtained in our study, we conducted 48 h back trajectories at 250m altitude (HYSPLIT) for each point along the *Tara* North Atlantic transect where AMP were detected. The vast majority of the air masses passed over the ocean during the full 48 hr trajectories (see Figure 2C). Next, we assessed the atmospheric residence times of the AMP we found by calculating their settling (terminal) velocities (Table 1; see SI methods for the detailed calculations). The residence time of atmospheric particles is likely to be strongly affected by their atmospheric terminal velocities. Slower velocities (≤0.01m sec^{-1}) imply higher likelihood to be lifted from near the water surface by eddies and to be carried by the surrounding air [11]. We found AMP settling velocities in the range of 0.001 - 0.2 m sec^{-1} for the particles (Table 1, SI methods). By combining the HYSPLIT back trajectories with the obtained residence times, we can evaluate the source of the particles. Particles 2, 17, 30 and 38 were clearly a product of local emission, with residence times of minutes to several hours (Table 1). The rest, with residence times of 1-2 days, combined with the HYSPLIT back trajectories, also appear to have been formed in the ocean. This suggests that the AMP found in this study were emitted from the seawater, indicating wind-related processes via bubble bursting.

AMP particles with irregular and elongated shapes can have long atmospheric residence times. For the most elongated AMP, despite having long major axis, their atmospheric settling

velocities are in the range of 0.001 m sec^{-1}, equivalent to typical submicron particles, which are suspended in the marine atmosphere in a timescale of days, and dominate the marine boundary layer [12]. We summarize that the AMP we found can remain in the atmosphere up to ~2 days, and for a typical marine wind speed of 8 ms^{-1} can be transported to 100's -1000's km. However, AMP with settling velocities in the order of ~0.1 m sec^{-1} will remain in the atmosphere for ~5 min and travel ~ 3km [12]

To further explore the role of the open ocean as a source of AMP, we compared the chemical composition of the AMP samples with microplastic particles collected from seawater sampled at the same approximate location to where the AMP were sampled. We chose AMP samples collected at 26°N 66°W - 26°N 68°W and 26°N 68°W - 26°N 72°W, (samples 38 and 39, respectively; Figure 1A), where the HYSPLIT trajectories indicate the air masses are of purely oceanic source. Microplastic particles collected from the seawater on the 22.06.2016 at the area of 26°N 68°W, were compared with airborne sample collected at 26°N 66°W - 26°N 68°W (sample 38) containing polyethylene AMP, which was collected on the same day, and with airborne sample collected at 26°N 68°W - 26°N 72°W (sample 39) containing polypropylene AMP, which was collected the next day (23.06.2016) (see SI methods, and Figure 1A).

The seawater microplastic particles selected for analysis and comparison with the AMP underwent thorough morphological scanning in order to assure their identification as plastic before micro-Raman spectroscopy measurements (see SI methods).
Eight microplastic particles were analyzed from the seawater sample 26°N 68°W, seven of which contained polyethylene or polypropylene (Figure 3). Five were identified as polyethylene, out of which two were polyethylene compositions (one with Cobalt phthalocyanine and the second with acrylic acid), and two were identified as polypropylene (see Figure 3).

The oceanic origin of the air masses as indicated by the HYSPLIT back trajectories together with the chemical evidence from samples 38-39 and their corresponding seawater MP samples strongly suggest marine source of the AMP. Additionally, AMP sample 38 has a calculated terminal velocity of 0.2 m sec^{-1}, and 39 of 0.003 m sec^{-1}, yielding atmospheric residence times of ~5min and ~48hr, respectively (Table 1). This strongly suggests that both were produced in the ocean. AMP sample 38 was most likely locally produced, and sample 39 with a potential residence time of 48hr, may have been produced far from the sample collection point (the

residence time only indicates the maximal time a particle can spend in the atmosphere, and therefore could have also been emitted very shortly before sampling), but is also likely of oceanic source, as the full 48hr duration of the HYSPLIT back trajectory was spent over the ocean. Furthermore, the specific densities of most AMP we found, and specifically polyethylene and polypropylene found in the seawater and in AMP samples 38 and 39, are lower than the specific density of seawater (Table 1), and are therefore likely to accumulate in the sea surface microlayer and thus can be readily aerosolized.

These evidence suggest that AMP can be produced in the open ocean through wind related processes forming sea spray aerosol particles, a different source and formation mechanism than the terrestrial and coastal AMP detected to date [39,48,55-65]. It also means that the ocean can be a constant global source of AMP, which may be transported to coastal regions and remote locations. However, we cannot dismiss the possibility that some AMP particles may have originated from other boats, continents or islands [65], and transported into the remote marine atmosphere. ”

Table 1: The sample (filter) numbers in which AMP was detected, the types of plastic polymers detected by micro-Raman spectroscopy and their density, diameters and calculated equivalent sphere, AMP terminal settling velocities, and estimated residence times.

Filter number	Polymer detected	Polymer density (gr/cm^3)	Particle major and minor axis (μm)	μ_s = Terminal velocity of droplet/ sphere ($m\ s^{-1}$)	$\alpha=(b/a)^2$	μ= AMP terminal velocity of ($m\ s^{-1}$)	AMP residence time *
2	Polystyrene	0.96–1.04	66, 53	0.13	0.64	0.08	~ 10 min
5	Polystyrene	0.96–1.04	25, 18	0.017	0.52	0.009	~ 1 day
10	Polydimethylsiloxane	0.97	22, 17	0.014	0.6	0.008	~ 1 day

	(PDMS)						
11	Polydimethylsiloxane (PDMS)	0.97					
17	Polystyrene	0.96–1.04	124, 45	0.23	0.13	0.03	~ 5 hr
23	Polydimethylsiloxane (PDMS)	0.97	17, 13	0.01	0.58	0.006	~ 1 day
30	Polystyrene	0.96–1.04	100, 70	0.22	0.49	0.11	~ 5 min
38	Polyethylene	0.88–0.96	102, 97	0.22	0.90	0.20	~ 5 min
39	Polypropylene	0.86-0.95	172, 17	0.26	0.01	0.003	~ 2 days

*Based on the calculations in [12] (Table 8 in the chapter)

Finally, the text flow is not straight forward. I had to read some text passages a few times and/or to flip back and forth to understand how the experiments and analyses were set up.

Thank you, we revised the paper, and hope that the text flow is now improved.

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28th Sep 20

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

Best regards,

Heike Langenberg, PhD

Chief Editor
Communications Earth and Environment

On Twitter: @CommsEarth

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

Dear authors,

I think the manuscript has greatly improved and I thank you for including the estimation of AMP residence times and the calculation of the terminal velocities in the SI. This is indeed an important information. Overall I think you properly addressed the suggestions provided and in this new form the manuscript reads more complete.
I think the manuscript is an important contribution to the field, but I also think it still misses important points.
By reading the manuscript again I had the impression that it reports another description of a dataset, in another sampling context (although novel, as the authors describe), while the important implications of the study are just briefly mentioned. Actually, the implications would add to the novelty of it. Therefore I still have some suggestions on the discussions/results.

1) Since the major threat of AMP individuated by your paper considers pathogens and POPs, I think you should spend a few more words on how these particles may enter the marine environment and thus, present an ecological threat. Since, as you pointed out, most of them will be buoyant and stay in the sea-surface micro layer, in this respect I strongly suggest to refer to the study of Wurl and Obbard, 2004, "A review of pollutants in the sea-surface microlayer (SML): a unique habitat for marine organisms" as this points to important toxic effects of a varied suite of contaminants and ecological consequences. This aspect, in my opinion, is still missing in the paper. A stronger link to

adverse effects should be added, as the sources themselves may determine certain types of effect. These particles you describe are not inert at all as they undergo multiple processes that make them more dangerous, and this danger possibility should be better explained. For example, because the micro layer hosts some species' larvae. Thus, AMP, POPs and other pollutants may pose a serious risk to the species' survival. It is not the purpose of the study, but still I think you should explore these consequences when you describe adverse effects or toxicity, since adverse effects of pollutants have been explored, and you may thus establish an additional link to it (especially in lines 178-184 and 188-190).

2) In line 114, when you state that the particles found were likely emitted from the seawater through bubble bursting, would you define it as a resuspension effect? Do you think these particles might then have been in the ocean for a long time? Also lines 153-158. I suggest to add some discussion in this respect. For instance, see Lebreton et al. 2019 “A global mass budget for positively buoyant macroplastic debris in the ocean. Scientific Reports 9, 12922.”

3) You found that the particles are mostly derived from the ocean. I am unsure about the title then - airborne - since these particles are found in the marine atmosphere but have an oceanic origin. Just a thought.

Authors' responses:

On behalf of the co-authors of this manuscript, we would like to thank the editor and reviewers for their helpful comments. Please find enclosed our point-by-point response.

Reviewers' comments:

1) Since the major threat of AMP individuated by your paper considers pathogens and POPs, I think you should spend a few more words on how these particles may enter the marine environment and thus, present an ecological threat. Since, as you pointed out, most of them will be buoyant and stay in the sea-surface micro layer, in this respect I strongly suggest to refer to the study of Wurl and Obbard, 2004, "A review of pollutants in the sea-surface microlayer (SML): a unique habitat for marine organisms" as this points to important toxic effects of a varied suite of contaminants and ecological consequences. This aspect, in my opinion, is still missing in the paper. A stronger link to adverse effects should be added, as the sources themselves may determine certain types of effect. These particles you describe are not inert at all as they undergo multiple processes that make them more dangerous, and this danger possibility should be better explained. For example, because the micro
layer hosts some species' larvae. Thus, AMP, POPs and other pollutants may pose a serious risk to the species' survival. It is not the purpose of the study, but still I think you should explore these consequences when you describe adverse effects or toxicity, since adverse effects of pollutants have been explored, and you may thus establish an additional link to it (especially in lines 178-184 and 188-190).

Thank you. We added a more thorough explanation regarding microplastic effect on microbial ecology (lines 190-195):
'The presence of nano and micro-plastic in the marine atmosphere may have implications on the atmospheric composition, and may also increase microplastic toxicity due to exposure to UV radiation in the atmosphere [1,2,36], with possible severe consequences for marine microbial ecology. The consequences may include increased stress response, and a decrease in metabolism, growth, and reproduction, which are some of the phenomena observed in aquatic organisms interacting with microplastics [61].'

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Thank you. We agree that the AMP is most likely a suspension of a degraded macroplastic that was introduced into the ocean at an earlier stage. We don't have the means to know the source of the primary plastic. It could be particles that were suspended earlier, and are now re-suspended, or plastic that got into the oceans by other means. Moreover, we currently do not have a way to estimate the amount of time that the MP spent in the seawater prior to emission to the atmosphere, and cannot be sure that we can define the bubble bursting as a re-suspension effect, although it may be the case. We would define the AMP emission as part of the sea spray aerosol emission process. We elaborated on the process of emission of AMP from the seawater, to further clarify this point (lines 164-169):

'Our findings indicate a new possible source and formation mechanism for AMP, different to those found for the terrestrial and coastal AMP detected to date [13,22-33]. We have evidence from the open ocean, which suggest that AMP is emitted from the seawater into the atmosphere through wind related processes. MP is accumulated in the ocean surface microlayer, where it can be scavenged by rising air bubbles. The bursting of the bubbles injects the MP into the atmosphere in a process associated with sea spray aerosol formation [51,53].'

3) You found that the particles are mostly derived from the ocean. I am unsure about the title then - airborne - since these particles are found in the marine atmosphere but have an oceanic origin. Just a thought.

Thank you. We considered changing the word 'airborne' but found that it might be less accurate to write 'aerosolized' for example, since it is a suggested mechanism based on all the evidence we found, but other sources can't be completely ruled out. We would like to avoid writing a speculation, however strongly supported, in the title. The title was changed instead to: "Airborne microplastic particles detected in the remote marine atmosphere".