Response to Reviewers - GMD manuscript egusphere-2025-1575

Description and evaluation of airborne microplastics in the United Kingdom Earth System Model (UKESM1.1) using GLOMAP-mode

Thank you to the two reviewers who have provided their time and constructive reviews of this manuscript. To address the reviews, our microplastic simulations have been re-run with updated emissions and an improved representation of microplastic fibres, with all figures and tables updated. We believe this results in a more realistic treatment of microplastics in our model. Reviewer comments are below in black, responses in blue, and text changes to the manuscript in red.

Reviewer 1:

Major Comments:

Comment: Table 1: please explain your definition of plastics, since usually plastics are not soluble. Please explain where you get the data for soluble plastics, since the only measurements I have seen in the literature are insoluble particles. Please show your comparisons to soluble plastics also, separately. Please discuss this more in the paper if there are no measurements why you chose to include them.

Response: Apologies for the confusion. In this case 'soluble' and 'insoluble' are model-related descriptors that in terms of microplastics represent hydrophilic and hydrophobic particles respectively. Section 2.4 and the Table 1 caption have been edited to make this clear, and text throughout the results and discussion section has also been changed to reinforce this.

Tracked changes lines 105 - 106: "Aerosols in the soluble modes are hydrophilic and can be incorporated into cloud droplets and affect the formation of clouds"

Tracked changes lines 107 "Aerosols in the insoluble modes are hydrophobic and do not act as CCN."

Tracked changes Table 1 caption: "Aerosols in UKESM1.1 soluble modes are hydrophilic, and aerosol in UKESM1.1 insoluble modes are hydrophobic."

Tracked changes lines 223 – 224: "microplastics can be transferred to the model's soluble modes and become hydrophilic…"

Plus, more instances of 'hydrophobic' and 'hydrophilic' as a microplastic descriptor added through the text.

Comment: "The microplastic inventory is based on airborne microplastic deposition measurements collected across 11 National Park and Wilderness sites between 2017 and 2019 in the Western USA (Brahney et al., 2020)." Please explain how you use observations from the western united states to estimate global emissions. Please indicate why we should believe these extrapolations.

We have now extended section 2.2 and explain clearly how the observations were used along with the optimisation method.
- To be precise, nobody should believe in any published emission dataset for microplastics yet, and as indicated in the original publication, emissions are associated with uncertainty that is on the same range as the emissions.
- A recent paper by Evangelou et al. (Environ. Sci. Technol. 2024, 58, 9741−9749) shows that emissions in Africa are realistic, though it does not consider such large sizes.

Response: The Lagrangian particle dispersion model FLEXPART is used to calculate the sources of microplastic emissions from Brahney et al., (2020). Emissions patterns from other sectors are then used to estimate global microplastics emissions. The updated emissions use the latest knowledge in this space to try make the best

assumptions regarding emissions, although we acknowledge there is going to be uncertainties here. To constrain microplastic emissions using top-down methods, we need high spatial and temporal resolution observations collected for at least a year (to constrain potential seasonal variations), analysed with robust measuring techniques that have been intercompared with several others and found to give similar results. At present, neither such highly resolved observations exist, nor analytical methods are robust enough. However, the Brahney et al. (2020) dataset has the closest characteristics needed for inverse modelling (top-down), as samples were collected for over 1 year (long duration), in Western USA (small regional domain comprising many sampling points) and in weekly or bi-weekly frequency, using a relatively robust technique of micro-FTIR. The observations of Brahney et al. have been separated into 5 size-bins; the largest bin (100-250 um) has very large extent and inevitably the largest microplastic emissions mass. To our knowledge, there are no other measurements that could validate them. However, over the small domain of Western USA, the agreement of this size bin against the respective observation reaches R2 of 0.6 for small standard deviations (see Figure 1 in Evangeliou et al., 2022). Section 2.2 has been expanded with further detail on how these emissions were produced and how they have been updated since Evangeliou et al., (2022).

Evangeliou, N., Tichý, O., Eckhardt, S., Zwaaftink, C. G., and Brahney, J.: Sources and fate of atmospheric microplastics revealed from inverse and dispersion modelling: From global emissions to deposition, Journal of Hazardous Materials, 432, 128 585, https://doi.org/https://doi.org/10.1016/j.jhazmat.2022.128585, 2022.

Tracked changes lines 130 – 154: “The updated microplastic inventory is based on airborne microplastic deposition measurements collected across 11 National Park and Wilderness sites between 2017 and 2019 in the Western USA (Brahney et al., 2020). So far, this is the only consistent measurement dataset suitable for top-down estimates, because it comprises weekly to bi-weekly samples from background sites over a long period. The dataset is limited by its small spatial coverage, as well as the analysis method used for identification of microplastics, which restricts the size range of observed microplastics to 4 µm. The updated emissions inventory used a new version of the Lagrangian particle dispersion model FLEXPART, version 11 (Bakels et al., 2024). FLEXPART version 11 uses an updated settling scheme that, unlike other models, differentiates between spherical and non-spherical shapes (e.g fragments and fibres), that have been reported to disperse differently (Tatsii et al., 2024). The optimization procedure of measurements from Brahney et al. (2020) and source-receptor matrices were based on the Gibbs sampling method (Gelfand, 2000). We constructed a hierarchical Bayesian model, whose parameters are optimized using the Gibbs sampler. Gibbs sampling is beneficial for optimization in high-dimensional or complex problems where traditional methods struggle. It simplifies the process by sampling from conditional distributions, avoiding full joint evaluations. This makes it especially effective for Bayesian models and correlated parameters. Unlike gradient-based methods, it handles multimodality well. Its modularity also allows easy integration with other sampling strategies. This method was used to estimate microplastics emissions in the form of samples from posterior distributions, quantifying effectively uncertainties of estimated mean and median values. The respective posterior emissions estimated at a domain that cover most of the US (yet unpublished) were then extrapolated globally using emission patterns of other sectors. Sea spray, agriculture (plastic nets), resuspension from mineral dust in bare soil and road dust were assumed to be the main sources of microplastic fragments. For the microplastic fibres, their main source was assumed to be largely from clothing and linked to the distribution of the global population. Thus, fibre emissions are absent from the ocean. Yang et al. (2025) assessed the oceanic emission potential of microplastics and found that 100 µm long microplastic fibres of various widths did not produce an oceanic emissions flux due to size, density and shape.”

Tracked changes lines 155 – 170: “In the original publication, emissions of microplastics were estimated to be equal to 0.82 Tg y^{-1} for sizes between 5 – 25 µm, while fibres were 6.5 Tg y^{-1} for all sizes (10 – 3000 µm). In the updated inventory, microplastic emissions at the smallest and most highly dispersed size bin (5-25 µm) were 0.74 Tg y^{-1}, in contrast to 0.82 Tg y^{-1} in the original inventory, with greater differences at the largest sizes. Microfibre emissions are kept the same. The difference in the updated inventory is that the positions of ocean gyres (‘great garbage patches') are now considered alongside the sea spray inventory when determining oceanic emissions for microplastic fragments (Isobe et al., 2021), as well as the shape-oriented dispersion that is obtained using the newer model version of FLEXPART v11. Furthermore, the high emissions observed across polar regions in Evangeliou et al. (2022) have been reduced in the upgraded version, using the sea-ice extent adopted from the fifth generation

ECMWF (European Centre for Medium-range Weather Forecasts) atmospheric reanalysis produced by the Copernicus Climate Change Service (C3S; Hersbach et al., 2020). The new emission inventory is more realistic and in line with the latest knowledge, such as new insights into low oceanic microplastic emissions (Yang et al., 2025). To create emissions data files for UKESM1.1-AMIP the updated inventory was re-gridded with a resolution of 1.25◦ latitude × 1.85◦ longitude. One year of emissions data is available for 2018, based on when the airborne microplastic deposition measurements were collected (Brahney et al., 2020). Figure 1 shows microplastics emissions for both fragments and fibres using the updated emissions inventory.'

Comment: Figure 2: Why are there no fiber emissions from the ocean? What assumption does that derive from and how important is that?

Response: For the microplastic fibres, their main source is assumed to be largely from clothing and textiles, and linked to the distribution of the global population, which is why they don't have oceanic emissions. Global oceanic fibres would be hard to estimate due to a lack of reference studies, but we would expect oceanic fibre emissions to be lower as compared to fibre emissions in regions of high population density. Yang et al. (2022) looked at the emission of 100 µm long microplastic fibres of various widths from the ocean, and suggest that they did not emit due to size, density and shape.

Shanye Yang, Tao Zhang, Yuqi Gan, Xiaohui Lu, Hong Chen, Jianmin Chen, Xin Yang, and Xiaofei Wang *Environmental Science & Technology Letters* 2022 *9* (6), 513-519 DOI: 10.1021/acs.estlett.2c00214

Tracked changes lines 150 – 154: "For the microplastic fibres, their main source was assumed to be largely from clothing and linked to the distribution of the global population. Thus, fibre emissions are absent from the ocean. Yang et al., (2022) assessed the oceanic emission potential of microplastics and found that 100 µm long microplastic fibres of various widths did not produce an oceanic emissions flux due to size, density and shape."

Comment: Why are there such large emissions of microplastics from Africa? I think you might want to re-look at your emission scheme since you have as large of emissions from desert regions as population centers. Please show any data in this region that would justify these large emissions.

Response: Microplastic emissions for fragments in this region is associated with an emission inventory of mineral dust calculated using the FLEXPART DUST model in Evangeliou et al., (2022). Our emissions of microplastics fibres, based on population density, show sensibly low emissions over low populated African regions. Evangelou et al., (2024) used measurements of microplastic mass fractions in soil, and modelled mineral dust emission to similarly estimate high microplastic emissions over North African desert regions. Other evidence of emissions sources in these regions include mismanaged plastic waste that has the potential to fragment into microplastics through burning and weathering (https://ourworldindata.org/grapher/share-of-plastic-waste-that-is-mismanaged).

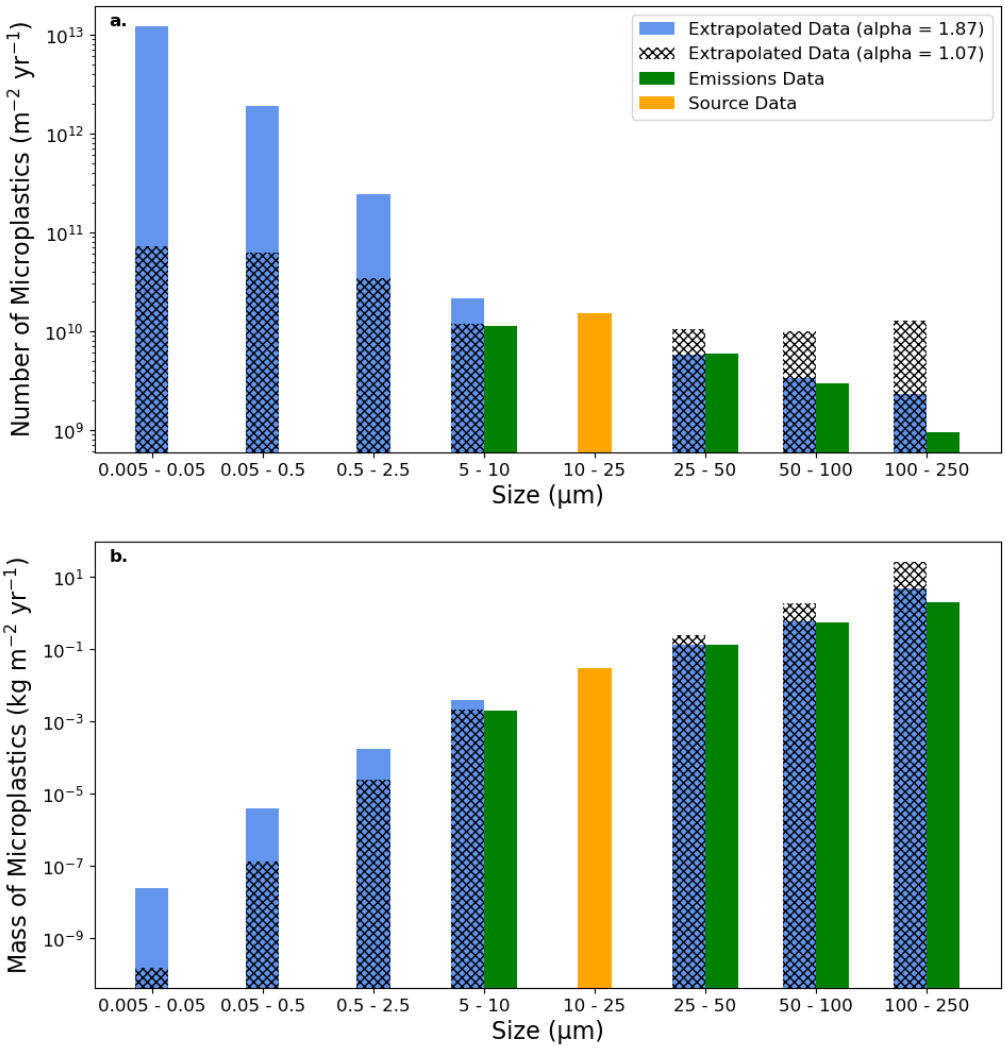
Ioanna Evangelou, Daria Tatsii, Silvia Bucci, and Andreas Stohl Environmental Science & Technology 2024 58 (22), 9741-9749 DOI: 10.1021/acs.est.4c01252

No text changes have been made here.

Comment: "When log-transformed, Leusch et al. (2023) demonstrate a linear increase in particle number with decreasing size." Please explain how this study extrapolated data from 5um down to nm. As far as I know, there is no robust way to measure microplastics at the nm scale, so you need more explanation here and justification of the use of this distribution. I should not have to read Leusch to understand this. It is possible you are just wildly extrapolating: that's fine, but state it clearly and it would be better to use a variety of extrapolations to see how sensitive your results are to the assumptions about size. Please also show comparisons of your results with available size data. If there is no size data, please discuss the implications.

Response: The Leush et al. (2023) study only assessed data down to 25 µm, so our work is assuming this relation holds into the nanometre range. Section 2.3 of the manuscript has been revised to clarify this assumption and the implications. We have also updated Appendix Figure 1 to indicate extrapolated microplastic concentrations using an alpha value of 1.07 corresponding to the lower limit of 1.44 ± 0.37 given for airborne microplastics in Leusch et al. (2023).

Tracked changes lines 206 – 216: "The Leusch et al., (2023) study demonstrated a linear increase in log-transformed microplastic number with decreasing size, but only for microplastics down to 25 µm. This is a common lower size limit in studies using µFTIR spectroscopy (e.g. Liu et al. 2019a; Chen et al. 2023; Abbasi et al. 2024), which has been a prevalent analysis methodology used to date. In this study, we extrapolate down to 5 nm under the assumption that this relation remains valid at smaller sizes of microplastics. This introduces uncertainty, particularly in the absence of robust observational data below 1 µm to validate against: to the best of our knowledge, no one has yet published a comprehensive size distribution for microplastics smaller than 10 µm. We acknowledge that differing values of α or a different relation than assumed in Equation 1, could lead to significantly different extrapolated microplastic concentrations. As such, our extrapolation should be treated as speculative. Adjusting the value of α to better represent these smaller-sized particles will be straightforward as and when new data become available. Nonetheless, the chosen value of α =1.81 indicates strong agreement between the extrapolated estimation and the emissions dataset, while the lower limit of α = 1.07 provides a poorer match (Supplementary Figure 1)."



Revised Appendix Figure 1: Microplastic emissions extrapolated across varying size bins for (a) number concentration and (b) mass concentration. Green bars represent reference emissions data from Evangeliou et al., (2022). The orange bar indicates the bin from which emissions were extrapolated for input to the model (blue bars, alpha = 1.87). The hatching indicates the lower range of the extrapolation uncertainty (alpha = 1.07).

Comment: "This suggests microplastic fibres need to be treated differently within the model, however they currently have the same spherical shape and settling velocities as the microplastic fragments." It sounds like you are ignoring the low settling velocity of microplastic fibers? Why don't you just pretend that they are much smaller particles instead for the settling velocity? This is really a killer: why even bother to model it if you aren't going to make sure you are modeling it as correctly as possible.

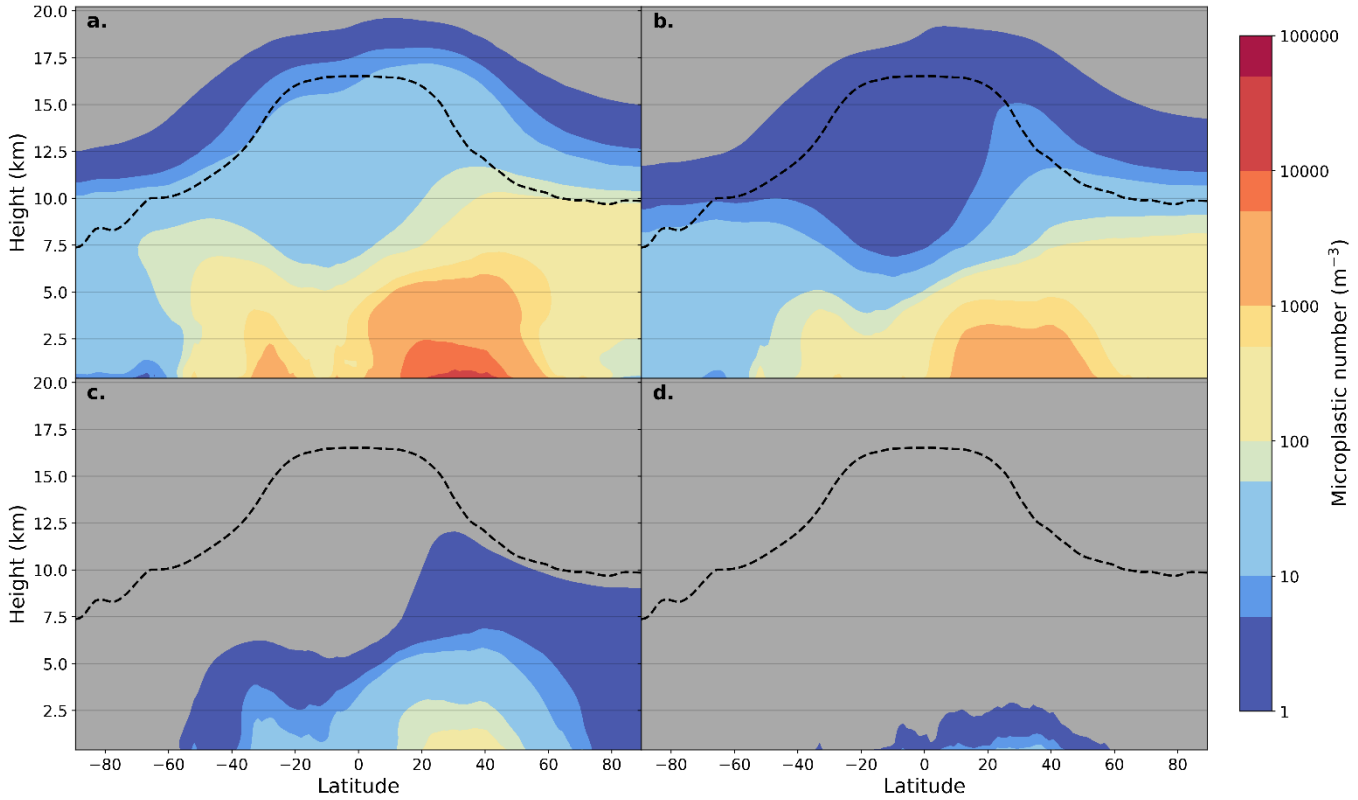
Response: We agree with the reviewer and have updated the model to include better representation of microplastic fibres. They are now modelled as volume-equivalent spheres, and have their settling velocities reduced by 60% based on the findings of Tatsii et al., (2024). Section 2.4 has been revised to reflect these changes, and discussion of fibres in the results section rewritten based on their new representation.

Tatsii, D., Bucci, S., Bhowmick, T., Guettler, J., Bakels, L., Bagheri, G., and Stohl, A.: Shape Matters: Long-Range Transport of Microplastic Fibers in the Atmosphere, Environmental Science & Technology, 58, 671–682, https://doi.org/10.1021/acs.est.3c08209, 2024.

Tracked changes lines 234 – 244: "Atmospheric transport and lifetime of microplastic fibres is influenced by their shapes (Tatsii et al., 2024; Xiao et al., 2023). Because of their non-spherical shape, microplastic fibres may be transported higher into the atmosphere than microplastic fragments. Tatsii et al. (2024) concluded that on average, microplastic fibres have settling velocities 60% lower when compared to spheres of an equivalent volume. To represent this behaviour within UKESM1.1, microplastic fibres are first modelling as volume-equivalent spheres based on their length and width from the emissions dataset. This changes the size distribution of fibres from having lengths between 10 – 3000 µm and widths between 1 – 10 µm, to spheres with diameters between 2.4 - 77 µm. This represents microplastic fibres in three modes, super-coarse insoluble, coarse insoluble, and coarse soluble through the GLOMAP ageing process. Secondly, the settling velocities of volume-equivalent spherical fibres is reduced within UKESM1.1 by 60% based on the work of Tatsii et al. (2024)."

Tracked changes lines 366 – 375: "Results shown across Figures 3, 4 and 5 agree with previous work (Tatsii et al., 2024; Bucci et al., 2024) modelling the vertical transport of microplastics, which also found that microplastics reach into the stratosphere. Tatsii et al. (2024) uggested that due to their reduced settling velocities, microplastic fibres ascend higher in the atmosphere and have increased global atmospheric transport than equivalent sized microplastic fragments. Even though microplastic fibres are modelled as volume-equivalent spheres with 60% reduced settling velocities, they show lesser maximum vertical extents than for the corresponding size modes for fragments. This is likely due to the lesser emissions of microplastics fibres than fragments from the source dataset (Figure 1). UKESM1.1 testing shows that microplastic fibres with 60% reduced settling velocities have greater vertical ascent than those with normal settling velocities (not shown)."

Old fibre vertical distribution (panel d only):



New figure showing fibre vertical distribution:

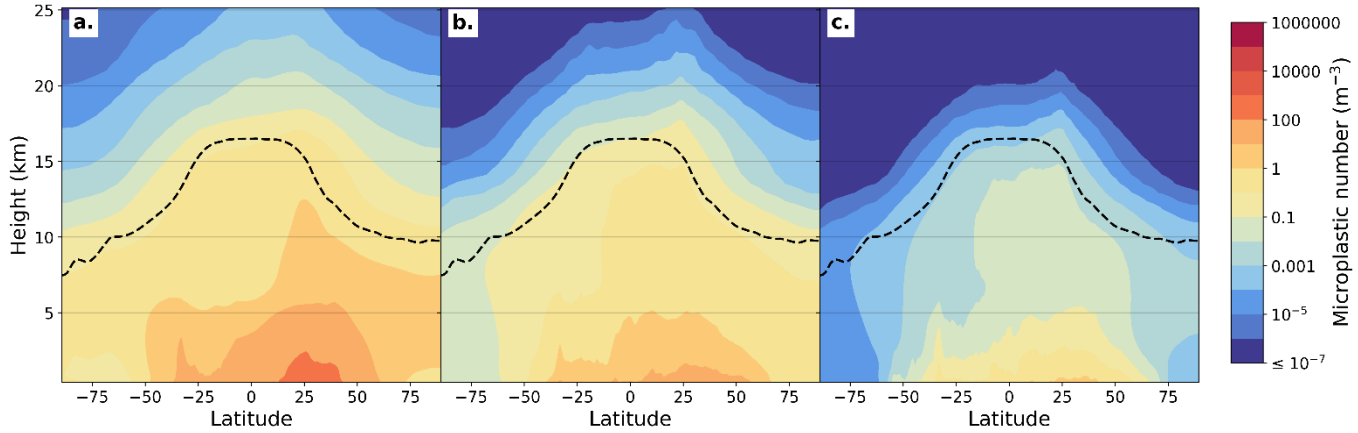


Figure 5: Vertical profile of annual zonal mean microplastic fibres concentrations (2005–2014) for (a) coarse soluble, (b) coarse insoluble, (c) super-coarse insoluble modes. Number concentrations less than or equal 1 x 10^{-7} m^{-3} are shown using the same colour scale as negligible values. The dashed line indicates the model's annual-mean tropopause height.

Comment: "Microplastic observations for model evaluation". Please show a table of the data you have collected using your method.

Response: A table of observational studies assessed has been added to the appendix. A full table is available in the Zenodo repository alongside the data used to produce the figures. Section 2.6 has been updated to iterate this more clearly.

Tracked changes lines 278 – 280: "Table A1 displays the author, name and year of studies used for comparison with the UKESM1.1 model. A complete table with concentration and latitude/longitude data is available at McErlich (2025)."

Comment: "For active sampling 330 studies, Figure 5a shows a regional bias with most studies undertaken in Europe and Asia. The model generally simulates greater microplastic concentrations than the observations, often by a few orders of magnitude, and with a poor correlation coefficient of r = 0.35 and RMSE of 5.09 (Figure 5b). This is particularly evident across studies reporting low observed concentrations, where the model simulates a large range of microplastic concentrations." The model does a really bad job! Since you are overpredicting the deposition rates, and overpredicting against the observations, this suggests you have a serious problem with your emissions and they should be much less.

"The disagreement between the model and observations is unsurprising, as observations represent a point source while the model output is the average over each latitude/longitude grid cell. Regions of high spatial variability such as around urban population centres would be most impacted by this discrepancy. Furthermore, many of the observational studies to date used micro-Fourier Transform Infrared Spectroscopy (µFTIR), which can only analyse microplastics of diameter 11 µm and larger (Allen et al., 2022), i.e. it cannot resolve microplastics down to the 2.5 µm threshold of the UKESM1.1 super-coarse mode (Table 2). This also accounts for some of the differences between the observations and the model." These reasons really aren't convincing. Please only compare the model output to the observations in the same size bin. But most of the mass should be in the larger size bins.

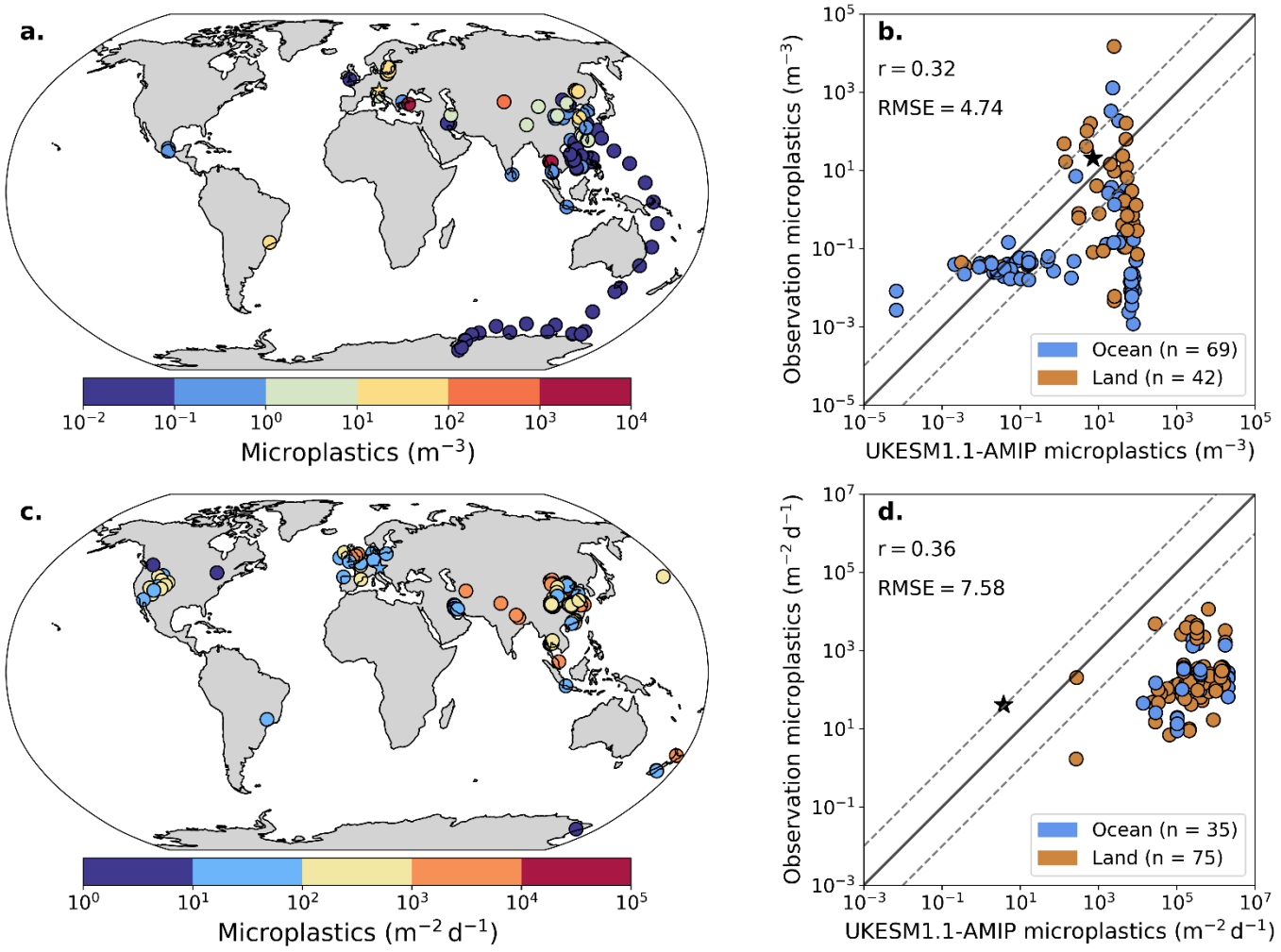
Response: We only compare data from the largest size bin in the model (> 2.5 µm) to the collated observational dataset. Further sorting the observational data to only include studies that resolves microplastics down to 2.5 µm greatly reduces the number of comparable data points, as remaining studies are mostly limited to those that used µRaman. The sample size reduces to 15 data points for sampling studies and 26 for deposition studies, compared to ~100 data points for each when including all studies. We acknowledge that model emissions and representation of microplastics processes are large sources of uncertainty impacting the comparison between the model and observations. Section 3.5 has been expanded to include discussion of this point. We

Once there are more established observational datasets available (with comparable methods being used, and better temporal and spatial coverage), the models can be rerun to keep comparing and better constrain representation of microplastics, as right now there is an abundance of limitations in the field observations, mainly that barely any of the studies are reporting small microplastics. We have added to Figure 6 (Figure 5 in the original manuscript) two data points representing studies reporting nanoplastics. The model compares okay with these studies.

Tracked changes lines 474 – 490: "The general disagreement between the model and observations is unsurprising, as the microplastic emissions going into the model have high levels of uncertainty. Physical processes that are relevant for microplastics such as wet deposition, CCN/INP capabilities, and impacts of ageing on microplastics are also poorly constrained, so may not be accurately represented within UKESM1.1 currently. Additional difficulty in comparing the model with observations arises from observations representing a point source while the model output is the average over each latitude/longitude grid cell. Regions of high spatial variability such as around urban population centres would be most impacted by this discrepancy. Furthermore, many of the observational studies to date used micro-Fourier Transform Infrared Spectroscopy (µFTIR), which can only analyse microplastics of diameter 11 µm and larger (Allen et al., 2022), i.e. it cannot resolve microplastics down to the 2.5 µm threshold of the UKESM1.1 super-coarse mode (Table 1). This also accounts for some of the differences between the observations and the model. The observations themselves are also uncertain due to variability in sampling and analysis methodologies and experimental setups. For example, deposition studies are sensitive to the sampled period microplastics are collected for (Aves et al., 2024).

The two nanoplastic studies show relatively good agreement with UKESM1.1-AMIP output, although the model simulates lower concentrations than those observed. Agreement with the current best estimates of microplastic observations at this size range provides some support for the extrapolated emissions methodology. However, the comparison remains highly uncertain due to the limited number of available studies and the estimated nature of modelled values at the nanoplastic scale."

Revised Figure 6: (a) Available observational microplastic number concentrations from active sampling studies (b) Comparison of observed concentrations from (a) with UKESM1.1-AMIP surface microplastic number concentrations at the nearest model grid cell (c) Available observational microplastic deposition fluxes from deposition studies (d) Comparison of observed deposition fluxes from (c) with UKESM1.1-AMIP microplastic deposition rates (combined wet and dry) at the nearest model grid cell. The correlation coefficients (r) and root mean square errors (RMSE) across (b) and (d) are calculated in log space, for the ocean and land measurements combined. The 1:1 (solid) and 1:10/10:1 (dashed) lines are plotted on (b) and (d) for reference. The star markers represent the two nanoplastics studies and related UKESM1.1-AMIP values, with units of ng m^{-3} for active sampling and kg km^{-2} yr^{-1} for deposition.



Comment: "This degradation forms microplastics (plastic particles 1- 5000 µm) and nanoplastics (particles smaller than 1 µm), which have the potential to cause ecological damage". Microplastics can be input to the atmosphere or other systems through other mechansims: burning or washing can also release.

Response: This information has been added to the introduction.

Tracked changes line 20: "Microplastics can also be released through burning (Luo et al., 2023) and washing (Šaravanja et al., 2022)."

Comment: "As a form of atmospheric aerosol, microplastics can contribute to climate change by interacting with incoming solar and outgoing thermal radiation. This in turn has an impact on the radiative balance of the atmosphere (Revell et al., 2021). Aerosols such as microplastics can also have indirect effects on radiative balance through cloud interactions and by acting as cloud condensation nuclei (CCN) (Aeschlimann et al., 2022). Clouds play an important role in the climate system (Forster et al., 2021) by reflecting sunlight to space (which has a cooling effect on Earth's surface) and trapping thermal radiation emitted by the Earth (which has a warming effect). In

general, clouds that have been perturbed by aerosols consist of more numerous and smaller cloud droplets, so that they reflect more sunlight and are longer lived (Twomey, 1977; Albrecht, 1989). " This paragraph should be rewritten. Revell et al., 2021 clearly showed that microplastics can be neglected for their radiative impact, and even if the impact is 100x higher (which would be hard), it will stay pretty much irrelevant. Similarly the number concentration of microplastics is unlikely to make it important for cloud condensation nuclei. So the only possible interaction with climate that can be important is Ice nucleation. Please rewrite this paragraph and combine it with the next so it doesn't pretend that microplastics are more important than they are for climate interactions.

Response: These paragraphs have been rewritten and combined as suggested.

Tracked changes lines 32 – 59: "As a form of atmospheric aerosol, microplastics could contribute to radiative forcing by interacting with incoming solar and outgoing thermal radiation. Revell et al. (2021) demonstrated that airborne microplastics exert a very small negative radiative forcing, given limited assumptions about microplastic size distribution, colour, surface concentration and vertical profile. Recent studies have shown that airborne microplastics may contribute to radiative forcing via their role as ice nucleating particles (INP; Ganguly and Ariya 2019; Busse et al. 2024; Brahana et al. 2024; Seifried et al. 2024), indicating that microplastics can potentially seed cloud formation. Research remains conflicted about how the ageing impacts the nucleation ability of microplastics, with studies indicating both increases Brahana et al., 2024) and decreases (Busse et al., 2024; Seifried et al., 2024) in the ice nucleation activity of microplastics due to ageing. When modelling atmospheric microplastics, Tatsii et al. (2025) found they contribute to INP concentrations. This impact was greatest under high microplastic emissions scenarios in pristine regions where other INP particles are scarce such as Antarctic and the Southern Ocean. Microplastics have also been collected in cloud water in cloud water (Xu et al., 2024c; Wang et al., 2023), indicating that their uptake into clouds occurs and that microplastics potentially act as cloud condensation nuclei (CCN). However, the present-day concentration of microplastics is unlikely to make them a significant source of CCN."

Comment: "Microplastics have also been collected in cloud water (Xu et al., 2024; Wang et al., 2023), indicating their uptake into clouds occurs and that microplastics potentially act as cloud condensation nuclei (CCN)." Or, more likely, are taken up by falling rain drops.

Response: While washout with falling raindrops will be relevant for microplastics, both studies used fog/cloud collectors to retrieve their samples and collected cloud water rather than rainfall.

Comment: Table 3: how does your model compare to other modeling studies in the literature, some of which are only regional, but many of which exist? For lifetime and sources, etc, and for different regions and concentrations. Please place your study in the context of the existing literature.

Response: The end of section 3.3 has been updated, introducing a new table to compare to existing literature.

Study	Emissions (Tg/yr)	Burden (Tg)	Lifetime (days)	Deposition (Tg/year)	Sizes (µm)	
This study	73.4	0.0136	0.04 - 16.9	73.26	0.005 - 250 (fragments)	2.4 - 76 (spherical fibres)
Yang et al. (2025)	10	0.03	0.9 - 365	10	0.5 - 70	
Brahney et al. (2021)	8.6	*	0.04 - 6.5	*	0.3 - 70	
Evangeliou et al. (2022)	16.1	*	2.5 - 8.3	16	10 – 250 (fragments),	10 – 3000 (fibres)
Fu et al. (2023)	0.324	0.0006	10.68	0.000322	0.3 - 70	

Table 4. Estimated emissions, burden, lifetime and deposition of microplastics, comparing this study with previous microplastic modelling studies. Entries with a asterisk denote values not reported in that study.

Tracked changes lines 409 – 424: "Table 4 compares the estimated emissions, burden, lifetimes, and deposition of microplastics within UKESM1.1 to previous microplastic modelling studies (Brahney et al., 2021; Evangeliou et al., 2022; Fu et al., 2023; Yang et al., 2025), as well as the size ranges modelled. Emissions (73.4 Tg/year) and deposition fluxes (73.26 Tg/year) in this study are greater than those modelled previously and are around ~4.5x greater than the previous version of the emissions modelled in Evangeliou et al. (2022). However, most of this burden corresponds to microplastics in the 100 – 250 µm size bin from the emissions dataset, which are not included in

any of the other microplastic modelling studies, and are not atmospherically relevant. When this size bin is excluded, the emissions decrease to 16 Tg/year. Despite large emissions and deposition rates, the observed microplastic burden (0.0136 Tg) is smaller than the burden reported in Yang et al. (2025), who see significantly longer microplastic lifetimes. Braheny et al. (2021) estimate microplastic lifetimes from 0.04 to 6.5 days when assessing microplastics of sizes between 0.3 and 70 µm. This matches well with the results seen for the coarse (1.9 - 5.6 days) and super-coarse (0.04 - 0.7 days) modes that align with their modelled size ranges."

Comment: Figure 5: why do you not use the Brahney et al,. 2020 deposition data to compare against your observations? Please list all the citations of the measurements in table.

Response: The methodology for collating the observational data using a focussed Scopus search was done for reproducibility, with a requirement to have microplastics in the title. Unfortunately, Brahney et al., (2020) was missed by the search as it doesn't have 'microplastics' in the title. Section 2.6 has been reworded slightly and Brahney et al., (2020) is now included in the list of observations, and represented on Figure 6. Citations for all the observational studies assessed have been added to the paper, and a table of studies has been introduced in Appendix Table 1. We have also added two studies reporting nanoplastic concentrations to the observations.

Tracked changes lines 275 – 278: "Brahney et al. (2020), which wasn't identified in the Scopus search because it doesn't have the word 'microplastics' in the title, has been added to the collated observational dataset, as our microplastic emissions are derived from their deposition measurements. In addition, we also include two studies reporting atmospheric nanoplastics (Kau et al., 2024; Materic et al., 2021), that were not identified by the Scopus search.

Comment: "Assessing the vertical transport of microplastics indicates that the smaller microplastics are well mixed in the troposphere," well mixed means that there is no vertical gradient: I don't think that's what you show. Please remove that phrase.

Response: This wording has been changed here and elsewhere in the paper.

Tracked changes lines 494 - 495: "Assessing the vertical transport of microplastics indicates that the smaller microplastics are present throughout troposphere"

Comment: "Compared to total aerosol number concentrations, microplastics currently contribute a minor fraction (Table 4)." Minor is an overstatement: microplastics are neglible.

Response: The new microplastics model runs have produced an increased microplastics total aerosol fraction that is 40x higher, although still very small. Table 4 has been updated with these results.

Reviewer 2:

General comments:

The paper presents a pioneering attempt to incorporate microplastic aerosols into a climate model. This is a valuable and timely work, as such components may become increasingly relevant and will have to be included in atmospheric modeling. However, I have several major concerns regarding the current version of the manuscript. Firstly, a number of key elements are insufficiently described, making it difficult to assess the robustness and validity of the methodology and results, as well as hindering the reproducibility. Additionally, the study relies on many assumptions, many of which are not adequately justified or explored in terms of their uncertainties, which significantly weakens the impact of the findings. A sensitivity analysis or scenario-based approach to explore the possible range of outcomes would strengthen the paper's scientific contribution. In particular, it would be important to avoid drawing conclusions on microplastic behaviours at this early stage, such as the longer atmospheric lifetime of small soluble microplastics or the assertion that dry deposition is the dominant removal pathway. These results follow directly from the assumptions built into the model, and we currently lack empirical validation for such assumptions. Furthermore, the manuscript would greatly benefit from a more transparent discussion of the construction of the emission inventories, including the rationale behind the choices made. In summary, while the work goes into a promising direction, the manuscript requires additional information to improve clarity, justify assumptions, and better quantify uncertainties before it is suitable for publication.

Specific comments:

Comment: Line 46: "If present in high enough concentrations.." can you be quantitative?

Response: This part of the sentence has been removed, and the whole paragraph restructured based on a comment from Reviewer 1.

Tracked changes lines 32 – 59: "As a form of atmospheric aerosol, microplastics could contribute to radiative forcing by interacting with incoming solar and outgoing thermal radiation. Revell et al. (2021) demonstrated that airborne microplastics exert a very small negative radiative forcing, given limited assumptions about microplastic size distribution, colour, surface concentration and vertical profile. Recent studies have shown that airborne microplastics may contribute to radiative forcing via their role as ice nucleating particles (INP; Ganguly and Ariya 2019; Busse et al. 2024; Brahana et al. 2024; Seifried et al. 2024), indicating that microplastics can potentially seed cloud formation. Research remains conflicted about how the ageing impacts the nucleation ability of microplastics, with studies indicating both increases Brahana et al., 2024) and decreases (Busse et al., 2024; Seifried et al., 2024) in the ice nucleation activity of microplastics due to ageing. When modelling atmospheric microplastics, Tatsii et al. (2025) found they contribute to INP concentrations. This impact was greatest under high microplastic emissions scenarios in pristine regions where other INP particles are scarce such as Antarctic and the Southern Ocean. Microplastics have also been collected in cloud water in cloud water (Xu et al., 2024c; Wang et al., 2023), indicating that their uptake into clouds occurs and that microplastics potentially act as cloud condensation nuclei (CCN). However, the present-day concentration of microplastics is unlikely to make them a significant source of CCN."

Comment: Lines 50-51: Please double-check the structure of this sentence. Do you mean "found in their modelling that atmospheric microplastic, under high emissions scenarios (how high?), can potentially contribute significantly to INP concentrations". From their paper, it seems that this is true only in specific parts of the globe, where other aerosols are not present. To not be misleading, I would specify it here, for example "…in regions where other aerosol species are absent".

Response: This sentence has been rephrased for clarity.

Tracked changes lines 55 – 56: "This impact was greatest under high microplastic emissions scenarios, and in pristine regions where other INP particles are scarce, such as Antarctic and the Southern Ocean."

Comment: Line 53-54: This sentence can be misleading in terms of cause and consequences. The contributions and implications of microplastics for total aerosol loading and climate change are not known because we currently do

not yet have good information to possibly include them in models. As the authors also discuss, the data are not yet covering enough of the globe and are not coherent. Same with their soluble or insoluble properties, which are still not understood. Hence, assuming their emissions and their behavior in the atmosphere is, indeed, only an assumption, and it has to be treated as such. I think this has to be made clear by the authors here, and throughout the paper.

Response: Thank you for this suggestion. The last paragraph of the introduction has been rewritten to highlight the uncertainties around microplastic sampling and analysis, and the subsequent uncertainties it would add to their implementation into a climate model.

Tracked changes lines 61 – 74: "As an emerging aerosol species, microplastics emissions are highly uncertain due to limited spatial and temporal coverage of observations. Difference in sample collection and analysis methods make comparisons between studies difficult. Because these methods are not yet standardized, variation within individual techniques further hinders comparisons between studies. While drawing firm conclusions about the atmospheric behaviour of microplastics is difficult given a lack of empirical data (for example their lifetime, transport and deposition pathways), models are useful tools to help interpret observations, and to inform future sampling and laboratory studies (e.g. identifying the most uncertain processes or emissions regions), thereby advancing the field as a whole. Here we describe the addition of microplastics as a new aerosol species in the United Kingdom Earth System Model. The model and microplastics scheme are described in Section 2. In Section 3 we present simulations of the global airborne microplastics loading and deposition to marine and terrestrial environments. We also evaluate the model against current observational data. Given the limitations described above, the model will inevitably need to be improved as new empirical studies or emission inventories emerge. Nonetheless, we anticipate this model to be a useful tool for airborne microplastics research."

Comment: Lines 118-124: This section needs to include many more details on how the emissions have been created, otherwise preventing the reader from understanding the significance of the results presented. For example, the authors state that the inventories have been updated with respect to previously published emissions (Evangeliou et al. 2022) by considering the ocean gyres, but it is unclear how has this been done. Did you normalize the emissions on the yearly spatial distribution? What was the procedure? Also, from the figure 1a it seems the ocean emissions have a different spatial resolution with respect to land emissions, why is it so?

Response: More detail has been added to section 2.2 describing the emissions, which has been updated again since the initial submission. In the original emissions dataset, oceanic microplastic were estimated to using an emissions dataset of sea spray. In the updated emissions, the positions of ocean gyres and their associated great garbage patches are also used provide a better estimation. The ocean emissions for fragments have the same resolution as land in the revised manuscript, with an interpolation done to extend coverage in places that previously had no emissions over the ocean. We uncertain what the Reviewer means by "Did you normalize the emissions on the yearly spatial distribution". The emissions were constrained monthly for 12 months using an inversion algorithm as explained in Evangeliou et al. (2022). This method is common and has been applied for several other species pollutants (see Jia et al., 2023: https://doi.org/10.1016/j.jenvman.2023.117735, Kim et al., 2021: https://agupubs.onlinelibrary.wiley.com/doi/10.1029/2021JD034888, Thompson et al., 2017: https://acp.copernicus.org/articles/17/3553/2017/).

Tracked changes lines 130 – 154: "The updated microplastic inventory is based on airborne microplastic deposition measurements collected across 11 National Park and Wilderness sites between 2017 and 2019 in the Western USA (Brahney et al., 2020). So far, this is the only consistent measurement dataset suitable for top-down estimates, because it comprises weekly to bi-weekly samples from background sites over a long period. The dataset is limited by its small spatial coverage, as well as the analysis method used for identification of microplastics, which restricts the size range of observed microplastics to 4 µm. The updated emissions inventory used a new version of the Lagrangian particle dispersion model FLEXPART, version 11 (Bakels et al., 2024). FLEXPART version 11 uses an updated settling scheme that, unlike other models, differentiates between spherical and non-spherical shapes (e.g fragments and fibres), that have been reported to disperse differently (Tatsii et al., 2024). The optimization

procedure of measurements from Brahney et al. (2020) and source-receptor matrices were based on the Gibbs sampling method (Gelfand, 2000). We constructed a hierarchical Bayesian model, whose parameters are optimized using the Gibbs sampler. Gibbs sampling is beneficial for optimization in high-dimensional or complex problems where traditional methods struggle. It simplifies the process by sampling from conditional distributions, avoiding full joint evaluations. This makes it especially effective for Bayesian models and correlated parameters. Unlike gradient-based methods, it handles multimodality well. Its modularity also allows easy integration with other sampling strategies. This method was used to estimate microplastics emissions in the form of samples from posterior distributions, quantifying effectively uncertainties of estimated mean and median values. The respective posterior emissions estimated at a domain that cover most of the US (yet unpublished) were then extrapolated globally using emission patterns of other sectors. Sea spray, agriculture (plastic nets), resuspension from mineral dust in bare soil and road dust were assumed to be the main sources of microplastic fragments. For the microplastic fibres, their main source was assumed to be largely from clothing and linked to the distribution of the global population. Thus, fibre emissions are absent from the ocean. Yang et al. (2025) assessed the oceanic emission potential of microplastics and found that 100 µm long microplastic fibres of various widths did not produce an oceanic emissions flux due to size, density and shape."

Tracked changes lines 155 – 170: "In the original publication, emissions of microplastics were estimated to be equal to 0.82 Tg y^{-1} for sizes between 5 – 25 µm, while fibres were 6.5 Tg y^{-1} for all sizes (10 – 3000 µm). In the updated inventory, microplastic emissions at the smallest and most highly dispersed size bin (5-25 µm) were 0.74 Tg y^{-1}, in contrast to 0.82 Tg y^{-1} in the original inventory, with greater differences at the largest sizes. Microfibre emissions are kept the same. The difference in the updated inventory is that the positions of ocean gyres ('great garbage patches') are now considered alongside the sea spray inventory when determining oceanic emissions for microplastic fragments (Isobe et al., 2021), as well as the shape-oriented dispersion that is obtained using the newer model version of FLEXPART v11. Furthermore, the high emissions observed across polar regions in Evangeliou et al. (2022) have been reduced in the upgraded version, using the sea-ice extent adopted from the fifth generation ECMWF (European Centre for Medium-range Weather Forecasts) atmospheric reanalysis produced by the Copernicus Climate Change Service (C3S; Hersbach et al., 2020). The new emission inventory is more realistic and in line with the latest knowledge, such as new insights into low oceanic microplastic emissions (Yang et al., 2025). To create emissions data files for UKESM1.1-AMIP the updated inventory was re-gridded with a resolution of 1.25◦ latitude × 1.85◦ longitude. One year of emissions data is available for 2018, based on when the airborne microplastic deposition measurements were collected (Brahney et al., 2020). Figure 1 shows microplastics emissions for both fragments and fibres using the updated emissions inventory.'

Comment: Similarly, in which way the polar region emissions have been reduced and based on which information? And how, and why have the land emissions been increased? Which of the sectors (agriculture, mineral dust, road dust), all of them as a sum? It will be important to describe these aspects to get a better idea of what is actually computed in the model.

Response: Polar emissions have been reduced based on sea-ice extent information from ERA5 reanalysis, which is now reflected in the text. For land emissions, each sector has been adjusted based on new knowledge regarding emissions inventories for each sector. For example, increases across Northern Africa are associated with resuspension from mineral dust in bare soil, whereas increases in populated regions are associated with road dust and microplastics emissions from vehicles. Land and all emissions, in general, have been changed here, due to (a) the different dispersion in the Source-Receptor Matrices calculated by the new FLEXPARTv11 version that was used to constrain posterior emissions, and (b) the more precise inversion algorithm with the Gibbs sampler used in the top-down method. We have revised Section 2.2 for clarity in the revised version of the manuscript.

See above response for emissions-related text changes.

Comment: Lines 143-145: Do you choose this bin because it is the largest number of microplastics from your emission inventories or from observational data?

Response: The bin was chosen as it contained the largest number of microplastics within the emissions inventory. This paragraph has been updated for clarity.

Tracked changes lines 190 – 193: "Microplastic fragments were extrapolated using the 10 -- 25µm size bin as a reference, as it contained the largest number of microplastics within the emissions inventory. Using a single size bin to extrapolate provided four remaining bins in the emissions dataset to validate the extrapolated microplastic concentrations against."

Comment: Lines 156-160: How is the solubility process represented from MP in the model? Is it the same as any specific aerosol included in the model? It has to be emphasized that for MP this is also a process of which, in reality, we have very little knowledge, so the resulting wet removal or CCN outcomes are also speculative.

Response: Within UKESM1.1 the solubility is reflecting whether the aerosol is hydrophilic or hydrophobic and is represented the same for microplastics as other aerosol species like black carbon and dust. Sections 2.1 and 2.4 has been updated to clarify this point, and microplastics are also now referred to as hydrophilic/hydrophobic through the paper to reinforce this understanding. However, we acknowledge that this representation is speculative.

Tracked changes lines 105 - 106: "Aerosols in the soluble modes are hydrophilic and can be incorporated into cloud droplets and affect the formation of clouds"

Tracked changes lines 107 "Aerosols in the insoluble modes are hydrophobic and do not act as CCN."

Tracked changes lines 220 – 224: "Microplastics are emitted as hydrophobic aerosol into the insoluble Aitken, accumulation, coarse and super-coarse modes. However, microplastics can be transferred to the model's soluble modes and become hydrophilic through the existing aerosol ageing within GLOMAP which also applies to existing aerosol species such as black carbon and dust."

Tracked changes lines 229 – 231 "While Wang et al., (2023) identified microplastics bearing hydrophilic groups in cloud water, we acknowledge that representing microplastics as hydrophilic aerosol is uncertain."

Comment: Lines 168-169: This is a bit unclear: what size and size distribution do you then assume for fibers? You mention an original emission inventory with fibers of lengths between 10-3000 um and diameter 1-10 um. How do you convert that here?

Response: Microplastic fibres have been updated with better representation. They are now modelled as volume equivalent spheres based on their length and width. Their settling velocities are also reduced to account for shape, based on the findings of Tatsii et al., (2024). Section 2.4 has been revised to reflect these changes. Fibres are not extrapolated down to smaller sizes like the fragments.

Tatsii, D., Bucci, S., Bhowmick, T., Guettler, J., Bakels, L., Bagheri, G., and Stohl, A.: Shape Matters: Long-Range Transport of Microplastic Fibers in the Atmosphere, Environmental Science & Technology, 58, 671–682, https://doi.org/10.1021/acs.est.3c08209, 2024.

Tracked changes lines 234 – 244: "Atmospheric transport and lifetime of microplastic fibres is influenced by their shapes (Tatsii et al., 2024; Xiao et al., 2023). Because of their non-spherical shape, microplastic fibres may be transported higher into the atmosphere than microplastic fragments. Tatsii et al. (2024) concluded that on average, microplastic fibres have settling velocities 60% lower when compared to spheres of an equivalent volume. To represent this behaviour within UKESM1.1, microplastic fibres are first modelling as volume-equivalent spheres based on their length and width from the emissions dataset. This changes the size distribution of fibres from having lengths between 10 – 3000 µm and widths between 1 – 10 µm, to spheres with diameters between 2.4 - 77 µm. This represents microplastic fibres in three modes, super-coarse insoluble, coarse insoluble, and coarse soluble

through the GLOMAP ageing process. Secondly, the settling velocities of volume-equivalent spherical fibres is reduced within UKESM1.1 by 60% based on the work of Tatsii et al. (2024)."

Tracked changes lines 203 – 205: "Microplastic fibres were not extrapolated due to their thread like shape; Once the length of microplastic fibres approaches the nanometre range their aspect ratios (length/diameter) become small enough they essentially behave more as microplastic fragments."

Comment: Line 176: "homogeneous in composition" means that you assume they are all assimilated in a single polymer? If that's the case, which one do you use and why?

Response: We do not use a single polymer but use an average plastic density. Microplastic are emitted into the model with a density of 1000 kg/m^3. This is an average plastic density based on what previous studies have used, and molar mass of 0.12 kg/mol corresponding to the molar mass of carbon. This information has been added to the end of section 2.4.

Tracked changes lines 251 – 255: "…all microplastic particles are assumed to be spherical and homogeneous in composition. Microplastic in UKESM1.1 have a density of 1000 kg/m3. This is and average plastic density similar those used in previous studies (Brahney et al., 2020; Evangeliou et al., 2022; Tatsii et al., 2024) and agrees with the work of Stride et al. (2024), who found that microplastics have densities between 940 kg/m3 and 1320 kg/m3. The molar mass of microplastics was set to 0.12 kg/mol, corresponding to the molar mass of carbon.

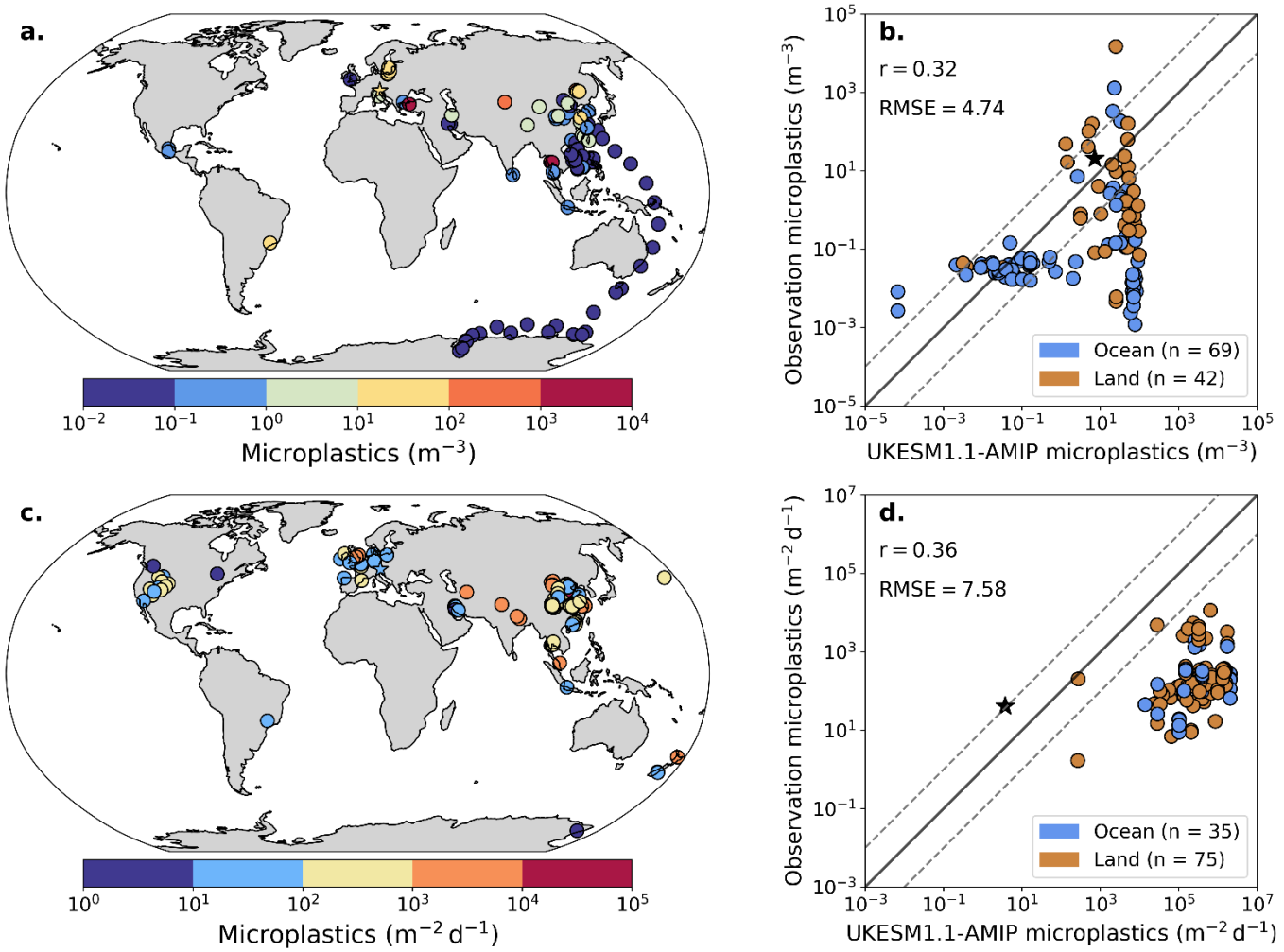
Comment: Lines 183-185: Just to clarify: The Mulchay et al paper refers to other aerosols simulations, do you mean that the model have already been well tested for that same period for other aerosols, so that you expect it to behave properly also for microplastic transport?

Response: Yes, this refers to the simulation periods, which was chosen to align with the 'historical' period in UKESM1.1-AMIP where forcings such as greenhouse gas concentrations are well constrained, as opposed to the 'future' simulations in UKESM1.1 (2015 and onwards in the model) where forcings are more speculative. Section 2.5 has been revised to clarify this point.

Tracked changes lines 258 – 263: "Simulations were performed with the atmosphere-only configuration of the model (UKESM1.1-AMIP) and run for a period of 11 years, from January 2004 to December 2014. The first 12 months were discarded as spin-up and we focus our analysis on the 10 years from January 2005 to December 2014. Microplastic emissions for the 12 months of available data have been repeated for each year of the simulations. While this predates atmospheric microplastic observations and the emissions inventory, it corresponds to the historical period of UKESM1.1-AMIP where ancillary data such as greenhouse gas emissions are well constrained (Mulcahy et al., 2023)."

Comment: Paragraph 2.6: It would be necessary to get more information on this section to get a proper interpretation of your model validation. For example, can you provide the actual list of the papers you got from your Scopus search and which ones you selected? And how many data points did you get for validation at the end for land and for ocean?

Response: A table of all the observational studies used has been added to the Appendix, with the full data provided in the Zenodo repository alongside the figure data. Figure 5 (now Figure 6 in the revised manuscript) has been amended to include the number of ocean and land data points.



Revised Figure 6: (a) Available observational microplastic number concentrations from active sampling studies (b) Comparison of observed concentrations from (a) with UKESM1.1-AMIP surface microplastic number concentrations at the nearest model grid cell (c) Available observational microplastic deposition fluxes from deposition studies (d) Comparison of observed deposition fluxes from (c) with UKESM1.1-AMIP microplastic deposition rates (combined wet and dry) at the nearest model grid cell. The correlation coefficients (r) and root mean square errors (RMSE) across (b) and (d) are calculated in log space, for the ocean and land measurements combined. The 1:1 (solid) and 1:10/10:1 (dashed) lines are plotted on (b) and (d) for reference. The star markers represent the two nanoplastics studies and related UKESM1.1-AMIP values, with units of ng m^{-3} for active sampling and kg km^{-2} yr^{-1} for deposition.

Comment: Line 205: The microplastic fibers distribution of concentrations is, for example, not easy to interpret without more information on how you considered these particles. Aside the settling velocity correction to be implemented for the shape, it would be important to know also which size distribution you associated with them, since big spherical particles would of course settle down very close to the emission points.

Response: As mentioned above, the representation of fibres has been changed to makes them smaller and take longer to settle. Figures have been updated, and results of the new model runs have been interpreted with text revisions where necessary.

Comment: Line 214: What is meant by "surface concentration" (up to which altitude)?

Response: The surface height is defined as the lowest vertical level in UKESM1.1-AMIP and corresponds to ~33 m above the land surface. This information has been added to Section 3.1.

Tracked changes line 285: “The surface height is defined as the lowest vertical level in UKESM1.1-AMIP and reaches up to ~33 m above the land surface.”

Comment: Line 227: I am also not sure to have understood how the solubility or insolubility of the particles is chosen. For the microfibers for example, are they only in the insoluble mode because they belong to the biggest particle size, which in the model is currently only associated to insoluble dust?

Response: As mentioned above, UKESM1.1 reflects whether the aerosol is hydrophilic or hydrophobic. The new representation of microplastic fibres gives them particle sizes corresponding emissions into the coarse and super-coarse modes. We have made the choice that microplastics are emitted as hydrophobic but can become coated in soluble material and become hydrophilic. Section 2.4 has been revised to make this clear.

Tracked changes lines 220 – 231: “Microplastics are emitted as hydrophobic aerosol into the insoluble Aitken, accumulation, coarse and super-coarse modes. However, microplastics can be transferred to the model's soluble modes and become hydrophilic through the existing aerosol ageing within GLOMAP which also applies to existing aerosol species such as black carbon and dust. This ageing occurs due to a build-up of soluble material such as sulfate on the surface of the aerosol (Mulcahy et al., 2018). Once the soluble material builds up to a size of 10 monolayers, the aerosol particles are transferred to the corresponding soluble mode. Because microplastics in the soluble modes are hydrophilic, it allows them to act as CCN within UKESM1.1 as they remain aloft in the atmosphere. Because the model does not contain a super-coarse soluble mode, microplastics from the super-coarse insoluble mode age into the coarse soluble mode instead, while retaining their larger mass. While Wang et al., (2023) identified microplastics bearing hydrophilic groups in cloud water, we acknowledge that representing microplastics as hydrophilic aerosol is uncertain.”

Comment: Line 244: Why do you say that number concentrations of less than 1/m3 are unrealistic? There are several studies that measured concentrations below 1 (e.g. Abbasi et al. 2022, Chandrakanthan et al 2023, Jia et al. 2025)

Response: Thank you for bringing these studies to our attention. We have changed the masking on the figures so that microplastic number concentrations less than or equal 1 x 10^{-7} m^{-3} are shown using the same colour scale, representing negligible concentrations.

Comment: Line 289: This is a bit counterintuitive. Can you give an explanation on why the soluble particles are actually the ones that have the longer lifetime? Also, I was wondering if coagulation in any way acting on the “removal” of these particles? You mention that the MP class is also able to coagulate in your model (being assimilated to the behavior of which particles? And how realistic is that?), but you didn’t discuss the effect of this process on them.

Response: The soluble microplastics appear to have longer lifetimes as they are hydrophilic and have uptake into clouds and water vapour, which enables longer transport. Section 3.3 has been reworded to make this clearer. As for the coagulation, it just means that microplastic aerosol in UKESM1.1 can coagulate to each other to form larger microplastic particles, so remain as microplastics. This is only a small flux transferring microplastics between the modes so has not been discussed further. The sentence mentioning coagulation in Section 2.4 has been revised to make this clear.

Tracked changes lines 384 – 401: “The predominant microplastic removal pathway across all size modes is dry deposition, with an average of 61% microplastic removal through this pathway. Wet deposition pathways indicate some interactions with cloud processes. Soluble mode hydrophilic microplastics show greater loss through nucleation scavenging (rainout) compared to the insoluble size modes with hydrophobic microplastics. Accumulation soluble mode microplastics show the greatest loss (52%) through nucleation scavenging. This reflects the ability of hydrophilic accumulation soluble mode microplastics to become incorporated into cloud droplets as CCN before wet deposition removes them. The coarse insoluble mode for both fragments and fibres shows the

greatest loss through impaction scavenging (washout), of 51% and 70% of total removal through this pathway respectively. Atmospheric lifetimes are longer for smaller particles as expected (Seinfeld and Pandis, 2016) with the greatest atmospheric lifetime occurring in the Aitken soluble mode (16.9 days). Atmospheric lifetimes are also longer for hydrophilic microplastics. Greater atmospheric lifetimes of hydrophilic microplastics (Table 3) potentially highlighting their ability to be incorporated into clouds and water vapour, after which they are carried with the subsequent atmospheric movement. This enables them to travel longer distances, especially if they are embedded in large weather systems like cyclones or fronts (Ryan et al., 2023)"

Tracked changes lines 231 – 232: "Similar to other aerosol species in GLOMAP, microplastics can undergo wet and dry deposition and coagulate with other microplastics into larger size modes.."

Comment: Lines 324-325: This is one of the reasons why you should indicate which studies you included in your comparison. From what you say, it seems that you take the super-coarse class from the model and compare the number concentrations measured in the various studies, but some will have upper limits at certain diameters, and others at different ones. This can lead to quite some big differences in the concentrations. How do you handle that? That would also be quite complicated if you also include the data on fibers.

Response: We have added a table to the Appendix summarising the studies used for comparison in Figure 6. Masking out the studies to those with detection limits down to 2.5 µm reduces the sample size greatly, making comparison more difficult. In our comparison, we combine super-coarse mode model output for fragments and fibres together when comparing with the observations, which often report their concentrations of fragments and fibres combined. Section 3.5 has been revised to also discuss the uncertainties within the model, and highlight the reasonable match between the two nanoplastic studies and the model output.

Tracked changes lines 474 – 490: "The general disagreement between the model and observations is unsurprising, as the microplastic emissions going into the model have high levels of uncertainty. Physical processes that are relevant for microplastics such as wet deposition, CCN/INP capabilities, and impacts of ageing on microplastics are also poorly constrained, so may not be accurately represented within UKESM1.1 currently. Additional difficulty in comparing the model with observations arises from observations representing a point source while the model output is the average over each latitude/longitude grid cell. Regions of high spatial variability such as around urban population centres would be most impacted by this discrepancy. Furthermore, many of the observational studies to date used micro-Fourier Transform Infrared Spectroscopy (µFTIR), which can only analyse microplastics of diameter 11 µm and larger (Allen et al., 2022), i.e. it cannot resolve microplastics down to the 2.5 µm threshold of the UKESM1.1 super-coarse mode (Table 1). This also accounts for some of the differences between the observations and the model. The observations themselves are also uncertain due to variability in sampling and analysis methodologies and experimental setups. For example, deposition studies are sensitive to the sampled period microplastics are collected for (Aves et al., 2024).

The two nanoplastic studies show relatively good agreement with UKESM1.1-AMIP output, although the model simulates lower concentrations than those observed. Agreement with the current best estimates of microplastic observations at this size range provides some support for the extrapolated emissions methodology. However, the comparison remains highly uncertain due to the limited number of available studies and the estimated nature of modelled values at the nanoplastic scale."

Comment: Lines 334-335: Looking at the scatter plot, though, there are high deviations also for the observations over land, which suggests that the model gives higher values with respect to the data overall.

Response: Figure 5 (now Figure 6) has been updated with the new model runs, which has enhanced the trend of the model showing greater microplastic concentrations than the observations. Section 3.5 has been rewritten to reflect the new results.

Tracked changes lines 460 – 472: "For active sampling studies, Figure 6a shows a regional bias with most studies undertaken in Europe and Asia. The model generally simulates greater microplastic concentrations than the observations, often by a few orders of magnitude, and with a poor correlation coefficient of r = 0.32 and RMSE of 4.74 (Figure 6b). The separation between ocean and land shows two distinct behaviours. Over land the model simulates narrow range of concentrations (10^0 to 10^2 m−3), while the observations show a wide range of concentrations (10^{-3} to 10^4 m−3) Over the oceans, the model shows a large range in concentrations (10^{-4} to 10^2 m−3) while the observations show a narrower range (10^{-3} to 10^{-1} m−3). Many points sampled over the ocean correspond to a single observational study by Chen et al. (2023) and may not be representative of the microplastic concentrations in these regions. Figure 6c also shows that the observations of microplastic deposition are biased towards European and Asian locations. UKESM1.1-AMIP simulates greater deposition fluxes as compared to observations at almost all data points. Comparisons with the model shows slightly higher correlation coefficient of r = 0.36, but an increased RMSE of 7.58 (Figure 6d)."

Comment: Lines 339-340: One of the main reasons of discrepancy is also the modelling itself. It is important to recognize that our knowledge at the moment is limited, hence the assumptions that are needed to feed the model may contain important biases (e.g. on the transport behavior, such as the solubility or coagulation properties, the distribution of the sources, their intensity, the size distribution). While the points raised by the authors are valid, those modelling uncertainties are most likely the biggest reason for the discrepancies with data. This is also why I would invite the author to add sensitivity studies or some assessment of the uncertainties related to the various assumptions introduced with this new aerosol species.

Response We acknowledge the biggest source of uncertainty is in the emissions, and section 3.5 has been expanded to include discussion of this point as mentioned above. Assessing the model sensitivity is outside scope of this paper, which aims to present the new microplastic Earth System modelling capabilities but makes an interesting avenue for future work. However, we have added a new table comparing our modelling results to previous microplastic modelling studies.

Study	Emissions (Tg/yr)	Burden (Tg)	Lifetime (days)	Deposition (Tg/year)	Sizes (µm)	
This study	73.4	0.0136	0.04 - 16.9	73.26	0.005 - 250 (fragments)	2.4 - 76 (spherical fibres)
Yang et al. (2025)	10	0.03	0.9 - 365	10	0.5 - 70	
Brahney et al. (2021)	8.6	*	0.04 - 6.5	*	0.3 - 70	
Evangeliou et al. (2022)	16.1	*	2.5 - 8.3	16	10 – 250 (fragments),	10 – 3000 (fibres)
Fu et al. (2023)	0.324	0.0006	10.68	0.000322	0.3 - 70	

Table 4. Estimated emissions, burden, lifetime and deposition of microplastics, comparing this study with previous microplastic modelling studies. Entries with a asterisk denote values not reported in that study.

Tracked changes lines 409 – 424: "Table 4 compares the estimated emissions, burden, lifetimes, and deposition of microplastics within UKESM1.1 to previous microplastic modelling studies (Brahney et al., 2021; Evangeliou et al., 2022; Fu et al., 2023; Yang et al., 2025), as well as the size ranges modelled. Emissions (73.4 Tg/year) and deposition fluxes (73.26 Tg/year) in this study are greater than those modelled previously and are around ~4.5x greater than the previous version of the emissions modelled in Evangeliou et al. (2022). However, most of this burden corresponds to microplastics in the 100 – 250 µm size bin from the emissions dataset, which are not included in any of the other microplastic modelling studies, and are not atmospherically relevant. When this size bin is excluded, the emissions decrease to 16 Tg/year. Despite large emissions and deposition rates, the observed microplastic burden (0.0136 Tg) is smaller than the burden reported in Yang et al. (2025), who see significantly longer microplastic lifetimes. Braheny et al. (2021) estimate microplastic lifetimes from 0.04 to 6.5 days when assessing microplastics of sizes between 0.3 and 70 µm. This matches well with the results seen for the coarse (1.9 - 5.6 days) and super-coarse (0.04 - 0.7 days) modes that align with their modelled size ranges."

Technical comments:

Comment: Line 37: I'd suggest removing the parenthesis and say "..by the Earth with a warming effect."

Response: This has been removed as part of the revisions to the introduction.

Comment: Line 95: space missing after 3.3um.

Response: This has been fixed.